

Advances in Printing and Media Technology

Vol. XLII(II)

Edited by Patrick Gane Associate editor: Cathy Ridgway

> Darmstadt MMXV

Advances in Printing and Media Technology

Proceedings of the 42nd International Research Conference of iarigai

Helsinki, Finland, September 2015

Published by the International Association of Research Organizations for the Information, Media and Graphic Arts Industries Darmstadt, Germany, 2015

> Edited by Patrick Gane, Aalto, Finland

Scientific Committee Anne Blayo (Grenoble) Timothy Claypole (Swansea) Edgar Dörsam (Darmstadt) Nils Enlund (Helsinki) Wolfgang Faigle (Stuttgart) Leopoldina Fortunati (Udine) David Frolich (Guildford) Patrick Gane (Aalto) Claire Gauzente (Nantes) Gorazd Golob (Ljubljana) Jon Hardeberg (Gjøvik) Jukka Hast (Espoo) Ulrike Herzau-Gerhardt (Leipzig) Gunter Hübner (Stuttgart) John Kettle (Espoo) Helmut Kipphan (Schwetzingen) Yuri Kuznetsov (St. Petersburg) Magnus Lestelius (Karlstad) Erzsebet Novotny (Budapest) Anastasios Politis (Athens) Wolfgang Schmidt (Osnabrück) Anu Seisto (Espoo) Karl-Heinz Selbmann (Bern) Johan Stenberg (Stockholm) Renke Wilken (Munich) Scott Williams (Rochester)

The facts published in this book are obtained from sources believed to be reliable. However, publishers can accept no legal liability for the contents of papers, nor for any information contained therein, nor for conclusions drawn by any party from it.

No part of this publication may be reproduced, stored in a retrieval system or transmitted in any form or by any means of electronic, mechanical, photocopying, recording or otherwise without the prior written permission of the publisher.

> Online edition ISBN 978-E-870 NBS ISSN 2409-4021

> > Copyright © iarigai 2015

Contents

Preface

	Half a century of research of the printing technology and media in iarigai Gorazd Golob	1
1A	Communication interfacing	
	From Digital to Print: RFID and QR-Code integration in Calabria (Southern Italy) Wood Chain Logistics Corrado Costa, Paolo Menesatti, Gianfranco Scrinzi, Giacomo Colle, Marco Bezzi, Federico Pallottino, Simone Figorilli, Francesca Antonucci, Giuseppe Scarascia	5
	Enhancing Product Security by using direct Marking and QR Codes Liisa Hakola, Thea Sipiläinen-Malm, Hanna-Leena Alakomi	11
	Experiments on Producing an Interactive Campus Map Angelina Barbonelova, Jiayuan Yin, Aarne Klemetti	17
	Typographic Test Form: investigating the performance of free fonts <i>Natalia Lumby</i>	25
	Innovative Combinations of Print & Digital – attitudes towards change in the European printing industry Markéta Držková, Sanne Tiekstra, Paula Miranda, Pedro Isaías, Igor Karlović, Rozália Szentgyörgyvölgyi, Tadeja Muck, Kaisa Vehmas, Claire Gauzente, Anu Seisto	33
1 B	Printing process	
	Ink-Water Balance during Emulsification and its Relation to Substrate Transfer and Optical Properties of Prints Carl-Mikael Tåg, Pertti Silfsten, Cathy Ridgway, Kai-Erik Peiponen, Patrick A.C. Gane	43
	A Memory Effect in Sheet-Fed-Offset Printing Wolfgang Fuchs, Michael Dauer, Ulrich Hirn, Wolfgang Bauer	53
	A new Concept to simulate commercial Print Trials on Lab Scale Part introduction of a lab printing device <i>Dietmar Wolf, Dirk Kisters, Patrick Gane</i>	65
	Evaluation of Sensors for Inline Viscosity Measurement in Gravure Printing Henrik Knauber, Peter Schöffler, Thomas Sprinzing, Armin Weichmann	77
	The Statement and Investigation of the Problem of Separation of a Paper Sheet from the Offset Cylinder after Printing. Rostislav Moginov, Alexander Vorozhtsov	89

2A	Printed	functionality	1

All Inkjet-Printed transparent Piezoelectric Polymer Actuators for Microfluidic Lab-on-a-Chip Systems Oliver Pabst, Maximilian Reif, Erik Beckert, Ramona Eberhardt, Andreas Tünnermann	99
Inkjet-Printed Reaction Arrays on Pigment Coated Substrates Risto Koivunen, Eveliina Jutila, Roger Bollström, Patrick Gane	107
Improving the Electrical Performance and Mechanical Properties of Conductive Ink on thin compound Substrate Andreas Willfahrt, Thomas Fischer, Gunter Hühner	115
A new FEM Simulation Method of Paper Materials by using a Gasket Model Jian Chen, Jann Neumann, Hans Martin Sauer, Edgar Dörsam	125
2B Print for packaging	
Critical Evaluations of Liquid Absorption Testing Methods for Package Printing Li Yang and Sofia Thorman	139
The Choice of a LCIA Method from a Practitioner's Perspective – an Example of Laminating Films in the Printing and Packaging Industry <i>Kirsten Radermacher, Ulrich Jung, Joachim M. Marzinkowski</i>	147
Does the use of Black Ink still comprise the "Darkest" Issue of CMYK Printing? Y. Kuznetsov, M. Ermoshina	157
3A Printed functionality 2	
Co-Innovating novel Printed-Electronic Products Kaisa Vehmas, Thea Sipiläinen-Malm, Anu Seisto, Maria Smolander, Himadri Majumdar	167
Fully printed Biodegradable Nanocellulose-Based Humidity Sensor for SMART LABEL Applications Tomáš Syrovy, Vinay Kumar, Martti Toivakka, Chunlin Xu, Lucie Syrová, Jan Řeboun, Douglas W. Bousfield	175
Printed Batteries, Overview, Status, Recent Developments, Future Perspectives Gunter Hübner, Martin Krebs, Patrick Rassek, Andreas Willfabrt	183
Zinc Oxide-Based Inks for Semi-Conductive Applications Shreyas Pathak, Alexandra Pekarovicova, Paul D. Fleming	195

3B	User and audience experience 1	
	Formatting Print Layouts with CSS 3 Christin Götz, Ulrich Nikolans	207
	The influence of location-related Factors on the Perception of Billboard Advertising <i>Janine Spieß, Ulrich Nikolaus</i>	213
	RocReadar – a system for collaborative transmedia publishing Yujue Wang, Saunil Desai, Elena Fedorovskaya	223
	Library Patrons' Preferences for Social Reading of ebooks Olli Nurmi	231
	Studies on the social Judgment of paper-based Communication in a special Environment <i>Csaba Horváth, Erzsébet Novotny</i>	239
5A	Coatings	
	Modelling Reflectance Spectra for Special Effect Pigment Coatings Nina Rogelj, Marta Klanjšek Gunde	247
	Thin Layer Chromatographic Behaviour of Dyes during Microfluidic Transport in Functionalised Calcium Carbonate Coatings <i>Eveliina Jutila, Risto Koivunen, Roger Bollström, Patrick Gane</i>	255
	Yellowing of Thick-Film Coatings printed with UV-Curable Inkjet Varnishes Manfred Schär, Tjaša Krivec, Gorazd Golob, Karl-Heinz Selbmann	265
	Effect of Coating Structure and Cationic Charge Level on Inkjet Printability Frank Gref, Päivi Miettinen, Jaana Heinonen, Mika Sundquist, Martti Toivakka	275
5B	User and audience experience 2	
	Why do certain Consumers avoid new Media Developments? An Investigation of three prudent users' Subjectivity <i>Stéphanie Gauttier, Claire Gauzente</i>	287
	Experimental Comparison of the User Experiences of Different Digital and Printed Newspaper Versions Janne S. Laine	295
	Paper or Digital? A Cross-Cultural Exploration of Students' use of Paper and Pen versus Digital Media in University Environments Jane Vincent, Leopoldina Fortunati	305
	How Today's Audience is searching for Contact with traditional Media via Facebook Liis Velsker, Ragne Kõuts	313
Inde	ex of authors	319

Half a century of research of the printing technology and media in iarigai

Gorazd Golob

Editor-in-Chief of the Journal of Print and Media Technology Research

E-mail: gorazd.golob@jpmtr.org journal@iarigai.org

> Another successful conference, at which we celebrated successful 50 years of iarigai -The International Association of Research Organizations for the Information, Media and Graphic Arts Industries, is behind us. We have seen many changes brought about by developments in technology, changing habits and expectations of end users and providers of technical solutions and services. The conference contributed to research and development in specific areas which for us are important.

> The mission statement of iarigai pointed out in the first place to the permanent support for redefining our scientific field. Every year we make a step forward to the new knowledge and understanding of the concepts that define us. It began with conventional printing and print media, however today the focus has shifted to communication in digital media, functional printing, packaging, coatings and user experience of the modern user of the media, especially interactive digital media.

> The conference was held in Helsinki, Finland, with a symbolic return to the country where iarigai was born in 1965 and began a successful series of the research conferences in Aulanko. In recent years, we tried to find the path to greater success and visibility of the conferences and the dissemination of research results.

The 42th conference was held as a joint event of iarigai, with VTT as the main organizer and important institution in the field of research and development of graphic and media technologies, and the members of COST Action FP1104 "New Possibilities for print media and packaging - Combining print with digital". The conference was final step of the project, giving opportunities to the members of the project team for the presentation of the results of their work to other researchers and to the general public.

In recent years the publishing model of publications, issued by iarigai, has also changed. Well known scientific book Advances in Printing and Media Technology, Vol.38, published in 2011 was upgraded to a serial publication with ISSN number assigned to traditional ISBN. In the year 2014, it was firstly published as a digital publication, with a content slightly modified with a combined publication of full conference papers and only abstracts from the authors who have decided to publish a full, completed paper in the Journal of the Print and Media Technology Research.

This year's edition of the Advances in Printing and Media Technology: Proceedings of the 42th International Research Conference of iarigai, includes extended abstracts of all papers presented at the conference. In terms of content and form they serve the basic purpose, disseminating the first and basic information on the authors' research achievements. The conference gave the opportunity to improve, extend and upgrade authors' contributions to full papers, suitable for publication in the Journal of Print and Media Technology Research.

1

The book, Advances of Printing and Media Media Technology, is available only in digital form, but equipped with all necessary reference data for publication into the bibliographic databases. The international nature of the publication is preserved and the quality of published contributions is assured with reviewing process, based on the reviews by internationally recognized experts and the final assessment of the conference Scientific committee.

The contributions of the authors, published in the Advances in Printing and Media Technology represent an important step forward towards the promotion of research in the field, which is still at the beginning of the recognition and confirmation of its role in the international scientific and research community.

The main features of this field are interdisciplinary and above mentioned quick changes of the main object of research. It requires a special effort due to specialization and visibility of this exciting and emerging research area and scientific discipline. For all of us the research results, including those published in this publication, are a source of satisfaction, however they are also a challenge for the continuation of our work and cooperation in the future. 42nd International Research Conference, 06 to 09 September 2015



Session 1A

Communication interfacing

Monday, 7 September 2015 11:05 – 12:45

Chair: Frans Mäyrä

From Digital to Print: RFID and QR-Code integration in Calabria (Southern Italy) Wood Chain Logistics

Corrado Costa¹, Paolo Menesatti¹, Gianfranco Scrinzi², Giacomo Colle, Marco Bezzi, Federico Pallottino¹, Simone Figorilli¹, Francesca Antonucci¹, Giuseppe Scarascia⁴

¹ Consiglio per la ricerca in agricoltura e l'analisi dell'economia agraria (CRA), Unità di ricerca per l'ingegneria agraria - Via della Pascolare 16, 00015 Monterotondo scalo (Rome), Italy.

² Consiglio per la ricerca in agricoltura e l'analisi dell'economia agraria (CRA), Unità di ricerca per il monitoraggio e la pianificazione forestale - Piazza Nicolini, 6 Loc. Villazzano, 38100 Trento, Italy

³ Effetreseizero s.r.l. - Piazza Nicolini 6, 38123 Trento, Italy.

⁴ Università della Tuscia, Dipartimento DIBAF - Via San Camillo De Lellis s.n.c., 01100 Viterbo, Italy.

E-mail: corrado.costa@entecra.it; paolo.menesatti@entecra.it; gianfranco.scrinzi@entecra.it; giacomo.colle@f360.it; marco.bezzi@f360.it; fedepall@yahoo.it; simone.figorilli@infone.it; francescaantonucci@hotmail.it; gscaras@unitus.it

Short Abstract

The activity with title "From digital to print: RFID and QR-code integration in southern Italy wood chain logistics (Calabria)" is being developed within the project "Ambi.Tec.Fil.Legno: Tecnologie innovative ad elevata sostenibilità ambientale nella filiera foresta -legno-energia" aiming also to improve the wood chain logistic in Calabria throughout the introduction of innovative technologies based on RFID. In particular these systems will be specifically adapted, developed and finally tested in field regarding three main applications regarding standing trees designated to be cut, the chain of custody and monumental trees in order to: create effective tools for collaboration through the wood supply chain; innovate for improving harvest planning and operations; integrate planning, operations, harvesting and sales; front key issues facing wood and timber freight, shipping and transport; fulfil traceability requirements needed in order to obtain product certification and satisfy the consumer demand increasing in terms of product information in an always more competitive multimedia world.

Keywords: forestry, infotracing, traceability, wood quality.

1. RFID and QR-code

Radio Frequency Identification (RFID) is a technology providing great opportunities to improve the management of information flow, security and thus quality within the wood chain. A typical RFID structure (Roberts 2006; Costa et al. 2012) including an RFID tag, an antenna (reader) and a database system used to store the information gained through the routine such as the tag ID and stored information till that point within the chain. The tags can be made of a variety of materials depending on the application needed (glass different epoxy resins, others), typically are composed by an antenna, a microchip containing memory storage and the material constituting its shape and body. The tags must have a univocal identification code. On the market are present read-only or read–write tags. These last can be useful where there is the need to add information along the chain in order to generate the production and logistic history of the product in a final database. Moreover tags can be active, carrying battery, or passive, where these last are not able to communicate without the powering field of interrogators and are activate just within its range (Costa et al., 2012).

An RFID antenna (or interrogator) on its side recognizes the tag ID and information and can be portable or just a fixed devices positioned in strategic places of the chain. Following the frequency used for the data interchange change the reading distance and the possibility for the signal to pass through certain materials.

RFID systems have many advantages over the more traditional bar code or the newer Quick Response code (QR-code) being more resistant to atmospheric agents, carrying more information, being rewritable at several steps along the chain, having a higher information flow speed thus being safer and more cost-effective. However, while to read a RFID tag a specific antenna is needed, any QR-code is made to be read with all the smartphone on the market making it preferable for application consumer oriented. Indeed, a QR-code is an optical label readable by a device including a camera and decoding software and that carry data about the good to which it is attached (product, URL, link, etc.).

2. Wood chain logistic in southern Italy (Calabria)

The logistic wood chain and forestry mechanization in Calabria region, with some exception, is substantially at initial level. Such condition often creates problems related to operators' safety, timber information, control of the chain of custody and proper soil and forestry management. Especially in order to correctly manage the chain of custody traceability is fundamental and represents the ability to trace the history, application or location of an item or activity, by means of its recorded identification. Estimation made by the Unità di ricerca per la selvicoltura in ambiente mediterraneo of the Consiglio per la Ricerca e la Sperimentazione in Agricoltura and the (Cosenza, CRA-SAM) regional forestry plan (2007-2013) evaluate about 1.4 milion m³/year the biomass that is possible to harvest without damaging the natural stocks underlining conspicuous financial resources. This data contribute to evidence the fact that forest production exploitation is not fully developed in Italy according to its potential following different problem among which: the supply chain fragmentation, the landowner inertia, the economic nature of public good of many products and services provided by the forests, the lack of governance and cooperation. In the last decades, the increase of ecosystem service demand and the gradual abandonment of many mountainous areas have caused a certain decrease of wood harvesting and significant changes in land management. Harvesting rate has been maintained unchanged only in coppices, mainly targeted to fuelwood production. Lately certification systems are gathering more and more importance with the aim of ensuring that the product originates from sustainably managed forests. However, these approaches are mainly oriented to process rather than to the product, whose precise geographical origin is often lost in the downstream sawmill because of hard practicality of processes of space-temporal segregation of wood assortments. At the moment the reduced information flow present is managed by hand writing data on paper increasing errors rate, reducing data recording speed and data recorded amount. Automatic technologies, such as RFID systems, look increasingly concrete, especially from forest to sawmill. Costs must be balanced with product value, *i.e.*, the application is essentially targeted for valuable timber even if lately the costs of tags and antenna suitable for such scope are lowering (Timpe, 2006). These promising technologies can support a unique identification of the primary object (tree), along with management planning information (Hakli et al., 2010). The association of derived objects (trunks) could then gradually be already in use, transmitting possibly more information for measurement, quality, defects, even to guide subsequent processing and eliminate further accounting in the camp timber apron and relative risks for the safety of operators. It could also become an important advantage, both for the landowner, the harvesting company and the manufacturer, in order to perform backwards inferences on products with individual traceability at every stage of processing. Moreover, the region own another patrimony represented by the presence of many monumental trees that enrich its territory attracting tourists.

3. Potential intervention initiatives

Within this framework the Ambi.Tec.Fil.Legno group of research, in relationship with the acknowledged research project planned three main points of intervention, where will be possible to insert RFID systems in order to generate increased value products (*e.g.*, certifications), reduce environmental and operators' risks, create effective collaboration tools through the wood supply chain; innovate for improving harvest planning and operations and create a robust traceability system.

The first intervention regards the chain of custody as many organisms on the base of governments and consumers requirements do certify wooden product on the base of their geographical origins, processing and transport. Such a operation implies the use of RFID within different points along the chain, as shown in Figure 1, starting from the forest (standing trees designated to be cut) till the primary transformation (sawmill). Selected info can be replicated and attached to the final product, (*e.g.*, stored on a website and accessible by the consumer through the use of an easy readable QR-code), when convenient.

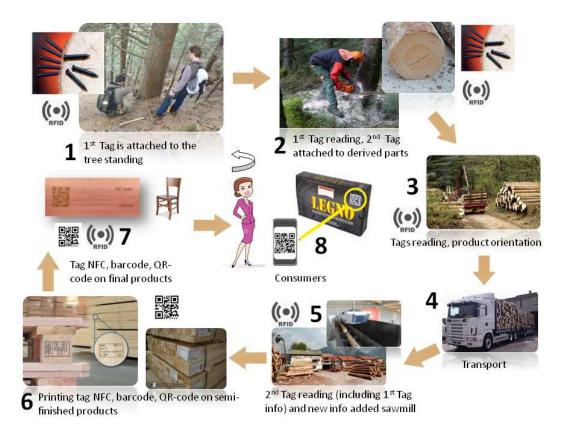


Figure 1 - 1) Specifically designed tag attached to standing trees designated to be cut; 2) reading info from first tag copied on second ones placed on each log, added eventual new info; 3) tags reading during logging for product sorting and merceological class attribution; 4) transport; 5) total info are read again in sawmill and for correct logs management and storage new data are added; 6) info are partially printed on semi-finished products or nail tag are used for valuable pieces; 7) info reach the final products, following its value, with NFC tag, RFID tag, barcode or QR-code; 8) consumers can scan the final products through a smartphone to reconstruct its history or to acquire deeper knowledge of its characteristics.

During each step (see relative numbers in Figure 1) several data are recorded and added to the previous one as follows:

- 1. Date (designation of the tree to the cut), GPS coordinates, specie, tree diameter at breast height, qualitative class, other information;
- 2. Harvesting date, log length, log average diameter, qualitative class, other information;
- 5. Date of entry into the sawmill, dimensions, quality, other information.

The second potential application provides RFID-based solutions to store and manage information during monitoring and inventories field surveys. The adoption of digital solutions in the field activities concerning forest management has to be a necessary step in order to achieve the maximum results in the consequent chain of custody RFID traceability. The integration between mobile GIS software and RFID technology allows several advantages in survey's data logging: direct in-field data validation,

fast and secure localization of single trees, improved GPS-based positioning of stands or sample's plots and many others.

Specifically, the following solutions are benefits of an RFID integration. First of all the opportunity to store directly in the RFID the observed data or a significant subset of them, this should be done tree by tree or a single RFID for the entire sample plot. Store the data directly in each tree facilitates the acceptance test plan allowing a deeper analysis of the discordances between the collected data and the test. The RFID technology allows also the re-use of the observed data to other research groups for future surveys. The info that will be gathered will be the GPS coordinates, the species, the date of each survey, the diameter at chest height, the tree height using stereovision or clinometer.

Another benefit of the RFID technology in field surveys is it's possibility to mark a tree in an invisible way, differently from standard mark techniques like color spray or plastic plaques. In monitoring surveys this advantage should avoid the common problem of the mark removals. Moreover, the invisibility of the markers guarantees the secrecy of the test areas and minimizes the influence of the survey on the activities of the forest stakeholders.

The last intervention regards monumental trees as fundamental part of our territory representing the memory of our past. The QR-code technology can be applied to enhance the appreciation of monument trees as high relevant value of Calabria forests and to establish a database for the recording of dimension and characteristics of those trees.

The web-database implemented in a Web-GIS will be consulted using a dedicated App for iPhone and Android systems or from a web-site. The App will help the tourist who wants to discover the tree in the forest by providing different tours based on geographical information such as roads and paths to reach each single tree. The database will store all the information related to each individual tree: age, height, volume, characteristics of the species and high quality pictures of the tree during different seasons and years.

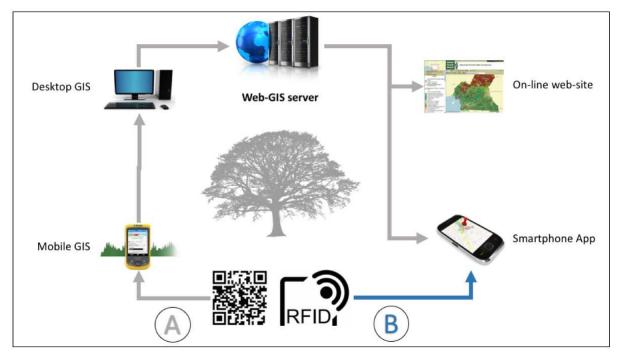


Figure 2 – Web-GIS dataflow. A: the field operator collects data by a mobile-GIS software and mark each tree of the survey with a RFID or QR-code tag. After software data validation, directly in field by means of the mobile application or in office with a GIS desktop software, data are stored in a web-GIS server. B: the user scans the QR-code or the RFID tag with a smartphone app, the software identify the tree and provides the data stored in the tag and, if necessary, download from the web-GIS additional data or database queries results.

9

Each historical tree will be marked with an unique RFID to store their main characteristics and a QR-Code to allow tourists to consult them with a smartphone and to access supplementary information from the on-line database.

4. Economic and technical feasibility

An analysis of the processes in the supply chain from forests to mills (Timpe, 2006) revealed that there is a potential to streamline operations and making a more efficient use of the resources by implementing a RFID-based log tracking system in the chain. This system should be an open loop, using inexpensive, passive RFID devices.

On 2006, Timpe calculated, in a concrete example when moving from one tag *per* stack to two tags *per* stack, that the break-even price *per* RFID is halved. Hence, he assumed that the economic feasibility for a passive RFID tag is lower than 0.43 USD. From that time some tags has been developed and patented for specific uses in forestry (Hakli et al., 2010; Björk et al., 2011; Hogg, 2012).

Moreover, the implementation of open-source technologies, such as Arduino and 3d printing technologies, in logistics (Menesatti et al., 2014) is opening the market to low-cost sensors and actuators with high reliability simplifying the implementation and the management and allowing a high flexibility in the hardware production. Open source technologies in logistics, for example for antennas, could allow a significant cost reduction from one third, for standard sensors, to two third, for more specific and evolved sensors.

Acknowledgments

Finalized research project Ambi.Tec.Fil.Legno project: "Tecnologie innovative ad elevata sostenibilità ambientale nella filiera foresta-legno-energia" (D.M. 952 Miur del 17/03/2014) supported by the Italian Ministry of University and Research.

References

Björk A, Erlandsson M, Häkli J, Jaakkola K, Nilsson Å, Nummila K, Puntanen V, Sirkka, A (2011) Monitoring environmental performance of the forestry supply chain using RFID. Computers in Industry, 62(8), pp. 830-841.

Costa C, Antonucci F, Pallottino F, Aguzzi J, Sarrià D, Menesatti P (2013) A review on agri-food supply chain traceability by means of RFID technology. Food and Bioprocess Technology, 6, pp. 353-366.

Hakli J, Jaakkola K, Pursula P, Huusko M, Nummila K. (2010) UHF RFID based tracking of logs in the forest industry. In RFID, 2010 IEEE International Conference on, pp. 245-251.

Hogg G (2012) RFID Tags-A Method For Tracking Sawtimber Through The Supply Chain. ICFR Technical Note 08/2012.

Menesatti P, Costa C, Antonucci F, Pallottino F, Figorilli S, Mencarelli F, Bellincontro A, Forniti R, Ferrante A, Trivellini A, Cocetta G, Bulgari R, Burchi G, Prisa D, Pacifici S, Rinaldi R, Bandinelli R,

Raffaelli V, (2014) L'innovazione cambia la gestione e la logistica della floricoltura. Il Floricultore, 9, pp. 40-43. ISSN: 1128-0212.

Roberts C M (2006) Radio frequency identification (RFID). Computers & Security, 25(1), pp. 18-26.

Timpe D (2006) RFID IN FORESTRY: Prospects of an RFID-based log tracking system as an alternative to stamping.

Enhancing Product Security by using direct Marking and QR Codes

Liisa Hakola, Thea Sipiläinen-Malm, Hanna-Leena Alakomi

VTT Technical Research Centre of Finland, P.O. Box 1000, FI-02044 VTT, Finland

E-mail: liisa.hakola@vtt.fi, thea.sipilainen-malm@vtt.fi, hanna-leena.alakomi@vtt.fi

Short Abstract

Typical means to fight counterfeiters is to put safety elements onto product packages. Product security can, however, be enhanced by using safety features on product itself in order to guarantee user safety, authenticity and tracking. Making markings on product itself is called direct marking. Laser marking and inkjet printing are suitable digital direct marking methods. In this paper VTT has evaluated the potential of inkjet printing based direct marking to provide security elements – specifically QR codes – on food products. The focus has been on evaluating if inkjet printed QR codes remain readable during storage. The main phenomena affecting code readability is ink spreading in cold and humid conditions. Edible commercial inks were used in this study. It was concluded that inkjet printing is a suitable direct marking methods to be used for security solutions.

Keywords: direct marking, inkjet, QR code, edible ink

1. Introduction and background

Counterfeiting is an increasing problem. It is estimated that the counterfeit food industry is worth about 49 billion US dollars (approximately 35 billion euros), according to the World Customs Institute [Interlandi, 2010]. Accorging to EU, counterfeited and pirated articles threaten the health and safety of EU citizens, their jobs, community competitiveness, trade, and investment in research and innovation [EU, 2010]. In 2010, EU Customs found over 103 million counterfeit and pirated goods, a figure that has almost doubled since 2009. In medicines the cases dealt with the customs increased 51 % from 2006 to 2007. Also a significant rise of 264 % counterfeit cases was found with personal care products.

There is now evidence that counterfeiters are moving into product lines where the effect is not only economic, but it can put lives at risk. Such product areas are food, car parts, aircraft parts, medicines, electronics and electrical good. Consequences from such fake products include faulty brake pads, drugs with no active ingredient, drugs with even poisons present, unsafe or out of date food as well as repackaged food. [Metters et al, 2010]

Traceability of the food product is also important e.g. with product recalls or when dealing with liability issues. Since 2002, traceability has been obligatory in the European Union for all food and feed businesses. Studies have shown that the information loss from one link in the chain to the next is huge, in some industries documented to be 80-95% [Trace project].

Typical means to fight counterfeiters is to put safety elements onto product packages. Product security can, however, be enhanced by using safety features on product itself in order to guarantee user safety, authenticity and tracking. For example, one aspect of counterfeiting is to replace genuine products with pirated ones, but retain the genuine package. In this regard, it is important to guarantee the authenticity of the product, not just the package. Furthermore, various decoration and entertainment solutions promote the brand. These two aspects (safety and added value) can be achieved by using a single marking when information content and marking technology have been adapted to this purpose.

Digital direct marking methods, such as laser marking and inkjet printing, where a marking is made directly on the product surface without a separate label, have been mainly used for product decoration purposes, but they offer a huge potential in product safety applications [Hakola et al, 2013]. Compared to traditional stamping-type of methods these direct marking methods can be easily automated, are suitable for mass production, offer possibility for customised markings and can be integrated into existing production lines. Inkjet printing has been widely accepted for food decoration purposes by both EU and FDA and there are commercially available suitable printing systems and edible inks.

One benefit of inkjet printing for direct marking is that it is a non-contact method. This means that the printhead and the substrate don't touch each other during printing, which avoids contamination. This is specifically important when decorating food products. Because inkjet printing is a digital printing method the printing layout is read from a digital file. This means that for direct marking purposes the printing can be anything that can be presented as a digital file such as text, numbers, photograph or a code.

Two-dimensional bar codes (2D bar codes) can be used as security elements. A 2D bar code called QR Code (Figure 1), also known as Quick Response Code, was chosen for this experiment, because of its popularity and availability of mobile phone compatible reading software. QR Codes consist of black and white squares called cells. QR Code provides a large information capacity, even up to 7336 numbers or 4464 alphanumeric characters per code. A sophisticated error correction algorithm is included, which means that information is readable even if up to 30 % of the code is destroyed. 2D bar codes can serve as a link to a database similar to linear bar codes, but they can also serve as an independent database. The physical size of the code is scalable without affecting the information capacity. This means that the cell size of a particular code can be scaled. QR Codes can be easily detected and read with many mobile phones thus providing a link to a digital media and additional information.



Figure 1. QR Code.

2. Scope and motivation

The results presented in this paper are part of the EU-funded EDEN project. The concept developed is to provide a "toolbox of toolboxes" based on a web tool to give stakeholders access to interoperable capabilities they deem important, or affordable, from a certified set of applications. Through the use of the service, all countries and stakeholders, irrespective of their existing capability levels, should be able to gain immediate advantages through improved interoperability. EDEN project includes three themed end-user demonstrations (Food Industry, Multi Chemical, Radiological) covering multiple hazards (CBRNe) and phases of the security cycle, response tiers, and stakeholder, and will be assessed by end users. The Food Industry demonstrator about contamination on the food chain focuses on guarantee and assessment of food safety during extreme situations such as war, terrorism attacks and natural disasters. Therefore the motivation to use direct marking on food products is related to special security needs in critical situations. In normal conditions consumers might not want such markings on their food products.

The food chain is vulnerable to natural, accidental and deliberate contamination. Therefore security and safety of food has a major role in the global public health. Several methods are needed to safeguard biosecurity and preparedness of the food chain. The modern food chain is a complex system with several players from primary production via production and retail to the consumer. In addition, food and feed industries are nowadays global business where raw materials, ingredients and final products are transported fast from one country to another. The complexity of the food chain, from farm-to-fork, makes the food chain vulnerable to deliberate contamination in several points. According to WHO, the two major strategies against food terrorism are prevention and response [WHO, 2008]. All the players in the food chain need to be active in preparedness and in prevention of potential contamination of food and feed chains.

The results presented here are part of the development of the Food Industry demonstrator where several ways to analyse and detect food safety, origin and authenticity are developed and demonstrated. One of the developed means is the use of inkjet printing to make durable markings on food products instead of and in addition to package markings in order to guarantee proper traceability. Other means include a leakage indicator and a method to detect pathogens [Alakomi et al, 2015]. Specifically a meat product has been chosen as the demonstrator product due to its sensitivity to changed conditions. The concept evaluated in this paper is QR codes inkjet printed with edible ink on ham slices. The focus is on durability of the marking during storage in a refrigerator (+4 °C) for 1 week. The durability of the codes is affected by ink spreading and analysed by reading the codes by a mobile phone.

3. Materials and Methods

VTT's drop-on-demand piezoelectric inkjet printing equipment with industrial-scale printheads (Fujifilm Dimatix) was used for the direct marking printing experiments. The printing conditions were 80 pl drop size, 300 dpi resolution and 150 mm/s printing speed.

Four printing inks listed in Table 1 were used: three commercial solvent-based edible inks (Edible inks 1-3) and one standard solvent-based ink as a reference. The main difference between the inks is the main solvents used that affects the ink-substrate compatibility through ink spreading, drying speed and penetration, in addition to printability characteristics. Sliced ham bought from a local supermarket was used as a substrate. Copy paper (Staples Europe B.V., 80 g/m²) and a photographic paper (Intelicoat Technologies, 250 g/m²) were used as reference substrates for optimal print quality.

Ink	Trade name	Manufacturer	Main solvents	Colorant
Edible ink 1	Tapestry	Fujifilm Dimatix	Glycols, glycerol	Dye-based cyan
Edible ink 2	6120	Linx Printing Technologies	Ethyl acetate	Cyan
Edible ink 3	76000-00102	Leibinger Group	Ethanol	Cyan
Solvent based ink	Jetrion	Jetrion	Glycols	Pigment based black

Table 1. Details	of the	inks used.
------------------	--------	------------

The printing layout consisted of QR Codes with different cell size between 0.25-1.50 mm. The code contained text "www.vtt.fi" and had 21 x 21 cells. Mobile phone (Lumia 800) with a pre-installed QR code reading software was used for detecting the QR codes. The camera of the mobile phone has 8 Mpixs. After printing the ham slices were vacuum packaged or put into a pouch in order to simulate end use conditions. The slices were stored in +4 °C for one week. The QR Codes were read after printing, after packaging, after 1 day in storage and after 7 days in storage. Figure 2 shows the concept.

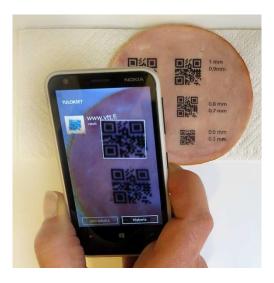


Figure 2. Code reading by mobile phone.

Optical print quality of the printed codes was evaluated visually by using Dino-lite digital microscope with 20x magnification.

4. Results and Discussion

Different QR code cell sizes between 0.10-1.50 mm were printed in different substrates with Edible ink 1 and Solvent based ink in order to define the smallest cell size that can be successfully read with mobile phone on ham slices. The smallest cell size was found to be in the range of 0.20-0.50 mm. It was expected that the QR codes would spread during cold storage and thus cell sizes between 0.25-1.50 mm were used for the final print layout.

Figure 3 shows a comparison of the inks when printed on ham slices. It can be seen that the contrast with Edible ink 3 wasn't very good probably due to ink penetration into the ham. This resulted in that even the biggest QR codes were difficult to decode. This ink wasn't used for the actual trials due to this problem that might be due to the ink designed to be used with dry products, such as cookies, instead of moist products. With both Edible ink 1 and 2 the contrast is very good, and after printing cell sizes down to 0.50 mm were decoded. Compared to the solvent based ink the print quality with edible inks is slightly poorer, because the edge sharpness isn't as good.

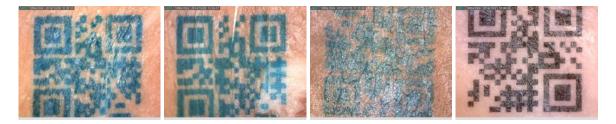


Figure 3. Comparison of the inks on ham slices: from left Edible ink 1, 2 and 3, and Solvent based ink. Cell size is 1.00 mm.

Figure 4 shows a comparison between codes printed on ham slices and on the two paper substrates with the different inks. Compared to the paper substrates print quality on ham slices is poorer as a result of ink spreading. On paper substrates all codes were readable with the mobile phone down to cell size 0.50 mm.



Figure 4. Comparison of the inks on different substrates. Cell size is 1.00 mm.

Set of samples were printed for the actual trial. QR codes were printed on ham slices with Edible ink 1 (13 parallel samples) and Edible ink 2 (8 parallel samples). Three parallel samples were printed on both paper substrates with the both inks. With Edible ink 1 five parallel samples printed on ham slices were vacuum packaged and five parallel samples were put in resealable pouches. The latter simulated a condition when the packaged is opened and stored again. With Edible ink 2 four parallel samples printed on ham slices were both vacuum packaged and put in resealable pouches. Vacuum packages (packaging) and pouches were then stored in +4 °C. All QR codes were read with the mobile phone after printing, after packaging/pouching, and after one and seven days in +4 °C. Table 2 presents the decoding results of the two edible inks. Figure 5 presents some example on how the codes spread during storage. Right after printing the codes with Edible ink 1 are easier to detect, but after cold storage the Edible ink 2 is better, because it spreads less. Codes in pouches spread more and are thus more difficult to detect probably due to more air entering the pouch than the air-tight vacuum package. Compared to copy paper and photographic paper the ham slices are equal in code readability after printing.

Ink	Edible	ink 1					Edible	ink 2				
Cell size (mm)	1.50	1.25	1.00	0.75	0.50	0.25	1.50	1.25	1.00	0.75	0.50	0.25
Ham slices												
After printing	13/13	13/13	13/13	12/13	12/13	0/13	8/8	8/8	8/8	5/8	0/8	0/8
After packaging	4/5	3/5	2/5	1/5	0/5	0/5	2/4	2/4	2/4	0/4	0/4	0/4
After pouching	1/5	4/5	2/5	0/5	0/5	0/5	4/4	4/4	4/4	4/4	0/4	0/4
Packages after 1 day in +4 °C	1/5	0/5	0/5	0/5	0/5	0/5	2/4	2/4	0/4	0/4	0/4	0/4
Packages after 7 days in +4 °C	1/5	0/5	0/5	0/5	0/5	0/5	0/4	0/4	0/4	0/4	0/4	0/4
Pouches after 1 day in +4 °C	1/5	0/5	0/5	0/5	0/5	0/5	2/4	1/4	0/4	0/4	0/4	0/4
Pouches after 7 days in +4 °C	0/5	0/5	0/5	0/5	0/5	0/5	0/4	0/4	0/4	0/4	0/4	0/4
Copy paper												
After printing	3/3	3/3	3/3	3/3	2/3	0/3	3/3	3/3	3/3	3/3	3/3	0/3
Photographic paper												
After printing	3/3	3/3	3/3	3/3	1/3	0/3	3/3	3/3	3/3	3/3	3/3	0/3

Table 2. Results for QR Code readability with different QR code cell sizes. The results are presented as "amount of successfully read codes / all codes in the series".

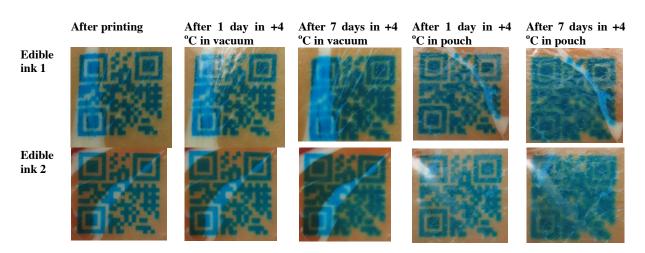


Figure 5. Spreading of edible inks during storage. Cell size is 1.50 mm.

4. Conclusions

Three edible inks were tested for suitability for making security marking on food products with ham slices chosen as the case product. The printing layout consisted of QR codes that were decoded with a mobile phone. It can be concluded that commercial edible inks are suitable for printing on ham slices and thus suitable to be used for security solutions described in this paper. However, the inks spread during cold storage, even when vacuum packaged. This means that careful optimisation of the code cell size is required since with one tested ink the codes were, however, detectable even after one week in +4 °C.

Acknowledgments

The research leading to these results has received funding from the European Union Seventh Framework Programme ([FP7/2007-2013]) under grant agreement no 313077 within the EDEN Project (End-user driven DEmo for cbrNe).

References

Alakomi H-L.; Hakola. L.; Maukonen, J.; Tsitko, I.; Storgårds, E.; Sipiläinen-Malm, T. Tools for food chain protection. Conference proceedings. NBC 2015, 9th Symposium on CBRNE treaths (2015).

Hakola, L.; Hartman, J.; Honkapää, K.; Laakso, P.; Penttilä, R.; Pitkänen, M.; Rusko, E.; Sipiläinen-Malm, T.; Vanttaja, I. New methods for improving food product safety and communication. Conference proceedings. NIP29 (2013), p. 498 – 502.

Interlandi, Jeneen. The Fake-Food Detectives. The Daily Beast from Newsweek 7.2.2010. (http://www.thedailybeast.com/newsweek/2010/02/07/the-fake-food-detectives.html)

Metters, L., Stobie, C. Using printing technologies to authenticate and fight against counterfeits. 26th International Conference on Digital Printing Technologies. Technical Program and Proceedings. Austin, USA (2010), p. 621 – 625

Report on EU customs enforcement of intellectual property rights, Results at the EU border – 2010. European Commission - Taxation and customs union, 14.7.2011. 33 pages. (http://ec.europa.eu/taxation_customs/resources/ documents/customs/customs_controls/counterfeit_piracy/statisti cs/statistics_2010.pdf)

Trace project website, www.trace.eu.org.

WHO (2008) Terrorist threats to food. Guidance for establishing and strengthening prevention and response. 52 p.

Experiments on Producing an Interactive Campus Map

Angelina Barbonelova, Jiayuan Yin, Aarne Klemetti

Helsinki Metropolia University of Applied Sciences PL 4070, 00079 Metropolia Finland

E-mails: Angelina.Barbonelova@metropolia.fi; Jiayuan.Yin@metropolia.fi; Aarne.Klemetti@metropolia.fi

Short Abstract

Several scientific experiments have been conducted on printing electronic devices with traditional printing equipment. The results have shown the feasibility of the currently applied technologies, but there is not yet a full understanding on implementing conventional printing for producing functional printouts in an uncontrolled environment setting.

This study presents research on different printing techniques in relation to their advantages and disadvantages in production of intelligent applications. Analysis of the studied literature was conducted and the findings were used as guidelines in creating an interactive campus map in an office room setting. Experiments on producing the map circuitry were done using inkjet technology and nanosilver ink. The campus locations were designed to be represented by electroluminescence light emitting elements. The light components were produced with screen printing and hand-held deposition methods. The methods and results of the performed experiments are discussed in this paper as well as the benefits of integrating printed intelligence into the traditional printing industry.

As a result, it was concluded that conventional printing techniques can be used for manufacturing of functional printouts such as interactive maps and more research and development is needed in order to reduce the production costs of intelligent applications.

Keywords: Printed intelligence, printed electronics, hybrid media technology, SW analysis

1. Introduction and background

Printing techniques are being applied in the manufacturing process of different applications varying from interactive posters and journals to luminous packages and printed biosensors. The market potential for printed, flexible and organic electronics is increasing due to the numerous beneficial factors accompanying its production line. In addition, it enables designing thin, flexible, lightweight and robust products that are able to share information with the user. According to the definition of VTT – Technical Research Centre of Finland Ltd, these products can be considered as Printed Intelligence.

Applications of printed intelligence technology are visible today. The need to get familiar with the methods used in their manufacturing process is increasing. Traditional and digital printing techniques have been studied as an approach of speeding up the production of flexible electronic components. For different applications and stages in the production chain, a variety of printing methods are used.

This study presents research on the applicability of conventional printing technologies on adding functionality to printouts and uses the findings to guide the development in creating an interactive map of Metropolia University of Applied Sciences (UAS) campuses.

The research questions in this study are:

- What is the applicability of conventional printing technologies on adding functionality to printouts?
- Is it feasible to produce functional printouts in an office environment setting?

2. Research methods

Our literature study is based on a collection of research data about the different printing techniques. The main focus is on the advantages and benefits of implementing printed intelligence technology in traditional printing equipment. SW (strengths and weaknesses) analyses based on the literature are made for traditional and digital printing methods in producing intelligent applications. These guidelines were used in the development of the interactive campus map.

In this study, the usage of inkjet, screen printing techniques for production were evaluated. Test measurements were performed using a multimeter and a paper thickness gauge (micrometre).

2.1 SW analyses on printing technologies

In this paper, SW analyses are presented as guidance in determining which of the printing processes is best for producing a certain component or a whole product. Comparing the strengths and weaknesses of each technique gives an opportunity to make faster decisions to achieve a more efficient production chain.

In their paper, Chang, Ge and Sanchez-Sinencio (2012) study and compare additive (fully printed) and subtractive (non-fully printed) processing. Their findings suggest that the additive process has higher ubiquity potential due to its cost and simplicity. However, subtractive processing is considered to be a competitor to silicon as it allows nanoscale accuracy. According to OE-A's (2013) research, analysis on resolution and throughput, shown in Figure 1, supports the findings described in the paper of Chang, Ge and Sanchez-Sinencio (2012)

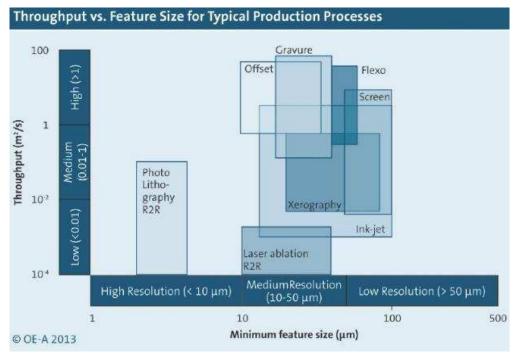


Figure 1. Resolution and throughput comparison. Reprinted from (OE-A, 2013).

Additive and subtractive processes are compared in this study by analyzing various factors such as throughput, cost efficiency, resolution, speed, registration, and performance of the printed product. Conducted SW analysis on the basis of the studied literature on the additive printing methods and the subtractive processes is shown in Table 1.

Tom Gamola el C	<i>L.</i> (2004), OE-A (2015), Li, Lu ana wong (2) Strengths	Weaknesses
	Additive processing	
Inkjet	 Reduced process steps; No use of screens or plates; 	- Reduced performance of fully printed circuits;
	 Print on demand; Accurate alignment for multi-layer 	- Resolution – estimated range 15 to 100μm (see Figure 1);
	printing; - Non contact deposition;	-Low viscosity inks (presented as weakness for producing components
	- Compatible with R2R (roll-to-roll) printing;	requiring deposition of a thick layer);
	 Low and medium throughput; Environmentally friendly due to lesser usage of materials; 	
	- Cost effectiveness	
Flexography	 Speed of R2R production is high; Low to medium ink viscosities; A count to be a sisteration; 	- Ink splitting between printing form and substrate causes somewhat inconsistent
	 Accurate layer registration; Print on different substrates; 	surface; -Medium to high throughput;
	- Large scale manufacturing;	- Low to medium resolution;
	- Fine line silver grid networks for OPV	- Complex formulation of inks;
	(Organic PhotoVoltaic) manufacturing is	- Long plate preparation time;
	promising;	
	- Cost effectiveness (R2R manufacturing); -Recyclable plates	
Screen	- Easy formulation of inks;	- Low resolution screens – estimated range
	- Allows printing on textiles and flexible	50 to 100μm (see Figure 1);
	materials;	- Registration problems in manual screen
	- Ability to adjust the printed layer	printing;
	thickness;	
	- Throughput varies depending on the screen size;	
	- High speed of R2R production;	
	- High potential due to simplicity of the	
	process;	
	- Cost effective	
	Strengths	Weaknesses
	Additive processing	
Gravure	- Speed of R2R production is high;	- Complex formulation of inks;
	- Reproducing very fine detailed elements;	- Time-consuming preparation of gravure
	- Uniform thickness layer;	cylinder;
	- Accurate layer registration;	- Medium to high throughput;
	- Medium resolution;	- High cost for fabricating gravure
	- Long runs on R2R printing;	cylinders;
	- Recyclable gravure cylinders;	
	- Cost-effective in large scale production	
Offset	- Fine printing resolution;	- Complex formulation of inks;
	- High speed of R2R production;	- Water, if used in the process;
	- Accurate layer registration;	- No personalization;
	- Cost-effectiveness in high volume	- Medium to high throughput;
	production; Recyclable plates	- High machinery cost;
	-Recyclable plates Strengths	Weaknesses
	Subtractive processin	
Photolithography		- Toner usage;
	, reproducing for, the dounted ciefficities,	· · · · · · · · · · · · · · · · · · ·

Table 1: SW analysis of printing technologies for printed intelligence applications. Data gathered from Gamota et al. (2004), OE-A (2013), Li, Lu and Wong (2010) and Suganuma (2014) publications.

laser ablation and xerography	 -Printing on flexible substrates; - Accurate registration; - High and Medium resolution; -Low throughput; -On-demand printing 	 Complex process; Non-scalability; High machinery cost
-------------------------------	---	---

2.2. Interactive Metropolia UAS Campus Map

Experimental work had to be done in a normal office environment in order to determine whether producing functional printouts in these conditions is reasonable. In order to start the experimentation work, an idea of a product that incorporates a functional printout had to be generated. Several ideas were considered and the one most suitable for the project was an interactive campus map for Metropolia UAS. This idea was chosen because of several factors, including simplicity of the structure, functionality and interactivity. As illustrated in Figure 2, the design implemented a thin film battery and EL (electroluminescence) light emitting elements attached to the circuitry layer, a foam layer and a graphics layer.



Figure 2. Design of Interactive Campus Map of Metropolia University of Applied Sciences

The foam layer was integrated into the design to eliminate the thickness difference of the components as well as to provide a non-conductive gap between the buttons and the circuit on the functional layer. The battery had a maximum thickness of 0.7 mm and in order to neutralize the bending of the top layer, the underlying layer (the foam sheet) had to be 0.7 mm thick as well. Cut-outs for the battery, buttons and light elements had to be made. For creating the prototype of the map, a paper sheet was used as an alternative to the foam layer. SMD LEDs (Surface Mounted Device Light Emitting Diode) were purchased for testing the circuit design and to provide an alternative solution for the light emitting components.

List of materials used:

- Polyimide substrate;
- Paper substrate;
- Nanosilver ink (particle size ≤ 10 nm) more information provided in Figure 4;
- EL-ink set (Electro Luminescent);

- Thin Film Battery-set: regular 1.5V; Mini 1.5V and regular 3.0V. Specifications shown in Figure 3;

- SMD LED sizes: 1608 and 2012;

SoftBattery	Reg 1,5V (Plus)	Mini 1,5V (Plus)	Reg 3,0V (Plus)	
Dimensions				
Maximum thickness	0,7 mm	0,7 mm	0,7 mm	
Outer dimensions (± 1 mm)	60 mm x 72 mm	36 mm x 46 mm	60 mm x 42 mm	
Maximum weight ⁽¹⁾	2,9 g	0,90 g	1,4 g	
Performance ⁽²⁾				
Nominal voltage ⁽³⁾	1,5 V	1,5 V	3,0 V	
Minimum initial capacity ⁽⁴⁾	90 mAh at 0,6 mA (1,2 mA)	18 mAh at 0,2 mA <mark>(0,4 mA)</mark>	10 mAh at 0,2 mA (0,4 mA)	
Initial internal resistance	~50 Ω (~25 Ω)	~150 Ω (~75 Ω)	~300 Ω (~150 Ω)	
Maximum peak current ⁽⁴⁾	8-10 mA (18-20 mA)	3-4 mA (6-8 mA)	3-4 mA (6-8 mA)	
Shelf lifetime Min. 2 years				

Figure 3. Battery specifications. Reprinted from (Enfucell, 2014).

In circuit design the choice of materials becomes essential for their chemical compatibility, adhesion and functional properties. Nano-sized metal particles provide a working solution for adhesion, especially when they form a bond with a clean and relatively rough surface.

The conducted research on the printing methods led to utilising Inkjet technology in producing the circuit as it was the ideal option for minimizing the throughput of used ink. The available equipment for this step was the OmniJet 100 Mini inkjet printing system. Consequently, the choice of materials was made according to the requirements for the piezo-electric inkjet system. The ink used at this phase was a nanosilver ink with particle size ≤ 10 nm that was purchased from Sigma-Aldrich – more information is provided in Figure 4.

Related Categories	47: Ag, Inks for Printing Applications, Materials Science, Nanomaterials, Nanopowders and Nanoparticle Dispersions, Organic and Printed Electronics, Printed Electronics Less
form	dispersion
	nanoparticle
concentration	50-60 wt. % in tetradecane
spec. resistivity	~2.7 μΩ-cm
surface tension	27-31 dyn/cm
refractive index	n20/D 1.333
particle size	≤10 nm
viscosity	7-14 cP
density	1.5-1.8 g/mL at 25 °C

Figure 4. Silver dispersion. Reprinted from Sigma-Aldrich.

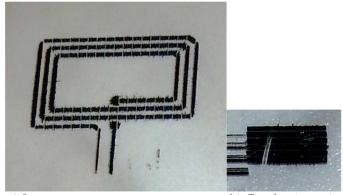
The OmniJet 100 Mini inkjet printing system allows the usage of four cartridges simultaneously and this provides an opportunity to use different functional inks. However, when this project was carried out, only one cartridge could be applied. The used cartridges were Samsung Mini 030 with a volume of 5ml and 16 nozzles. The printer was situated in a laboratory room, where UV safe yellow lighting was applied.

The design of the map included EL light elements to test if it is possible to produce them in an office environment and use them as location indicators for the map. In addition, the available EL-ink set allowed customization of the shape of the light sources and provided freedom in terms of design. The

new light elements were designed in representative shapes in accordance to the four main areas of study in Metropolia UAS. Four samples of EL light elements were produced during this project. Screen printing and hand-held deposition methods were used for applying the four necessary layers of the EL elements. The chosen substrate, onto which the layers were deposited, was a polyimide sheet in A4 paper size.

3. Results and Discussion

As the experimental work started on creating the circuitry, a few test prints were made. After a close examination of the printed shapes (see Figure 5a) and when measured with a multimeter, they presented conductive properties. The produced samples provided a positive answer to the research question about the feasibility for inkjet technology to be implemented in creating functional printouts.



a) Image pattern. b) Conductive printout. Figure 5. Inkjet printed samples.

The EL light emitting elements were produced with two different deposition methods. The structure of the electroluminescence component is built on several layers as illustrated in Figure 6. The screen printing unit was used for creating the first layer of the four light elements using the conductive ink from the EL ink set. It was produced with an estimated thickness of 12 μ m measured with a paper thickness gauge. The next layer was produced with the dielectric ink and it had to cover the bigger part of the electrode of each sample. A small line of the conductor, as shown in Figure 20a, had to be left uncovered in order to have a successful connection to a power supply. After the insulator was applied with an estimation of the thickness - 11 μ m, the following layer consisting of an active luminescent phosphor was deposited. The shape of the luminescent layer determines the shape of the lit element as seen in Figure 7b. The next deposition layer was a transparent conductor.

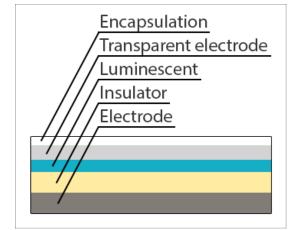
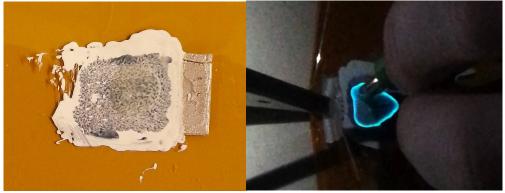


Figure 6. EL light emitting element.



a) *EL printed element*. b) *EL - 220 Volts applied*. *Figure 7. EL light emitting components created during the second phase of producing EL elements*.

The produced EL light components were tested by applying AC current gradually. From 0 to 100 volts the samples were not emitting light. When the voltage was increased to over 100 V, a flickering was noticed. The intensity of the light was increasing up to 220 V.

The measurement results from the produced EL samples led to conclude that, if they are to be used in the map prototype, the available Enfucell batteries cannot be used to provide the required power. The alternative solution to the EL elements were the SMD LEDs. The battery specifications, however, limit the use of LEDs. In the campus map design, a total of 16 light emitting components are connected to the circuitry. The maximum number of light elements connected to a button is 7 and therefore the available Enfucel batteries for this project, would not provide an efficient solution to drive standard 0.7V LEDs for extended period of time. However, recent advances in the development of Enfucell SoftBattery allow the improved batteries – Reg 1,5V Plus, Mini 1,5V Plus and Reg 3,0V Plus, to operate with double peak current and would provide an optimised battery solution for the interactive campus map (Enfucell, 2014).

A limitation was observed, when printing in the uncontrolled environment of the laboratories. Pollutants such as dust could significantly lower the conductivity of printed wires or thin elements. If a dust particle creates a bridge between the ink components, it would affect the electrical properties of the printed matter. During the experimental work done for this project, the deviation in humidity and temperature did not noticeably affect the produced printouts on glass and polyimide substrates.

The answers to the research questions are:

- The conventional printing technologies in the existing industry can be applied to printed intelligence. However, the nature of the product is important in terms of volume printing; for example, the requirement for changing the contents for an individual print (personalization) will rule out volume duplicate printing.

- As long as normal factory conditions do not damage the printouts, it is possible to produce functional printouts in an office environment setting.

4. Conclusions

The set research questions were answered, but more practical experiments need to take place in order to manufacture interactive maps in industrial scale. This study shows the feasibility of functional devices to be produced with existing graphic arts industry equipment. Since the traditional industry is mainly based on small and medium sized enterprises, the more there is to be studied on the processes of functional printouts.

The findings presented in this paper indicate there is demand for studying, finding and learning new ways of applying conventional printing technologies for developing and multiplying intelligent

devices and objects. The practical approach should include prototyping in order to understand the characteristics and behavior of required processes, substrates, and inks.

Acknowledgments

This study was carried out at the Leppävaara campus of Metropolia and partially funded by Uudenmaan liitto without whom we would not have been able to carry out our project. We are also grateful to the help and numerous discussions with Tapio Mäkelä and Kim Eiroma at VTT; Sami Veikkolainen at EditaPrima; Sami Kalliokoski, Jarmo Tuppurainen, Kari Vierinen, Maksim Korobkin, Ruslan Lagashkin, Chandra Pandey and Toni Spännäri from Metropolia.

References

Chang, J., Tong, Ge. and Sanchez-Sinencio, E., 2012. Challenges of printed electronics on flexible substrates. IEEE 55th International Midwest Symposium on Circuits and Systems (MWSCAS), 2012, [e-journal] pp.582 – 585.

Enfucell SoftBattery®, 2014. Full specs available at: <hr/>

Gamota, D., Brazis, P., Kalyanasundaram, K. and Zhang, J. eds., 2004. Printed organic and molecular electronics. London: Kluwer Academic Publishers.

Li Yi, Lu D., Wong C.P. Electrical conductive adhesives with nanotechnologies [online]. New York: Springer Science+Business Media; 2010. Available at: < http://link.springer.com/book/10.1007/978-0-387-88783-8 > [Accessed 19 April 2015].

OE-A, 2013. Resolution and throughput for a variety of processes. [image online] Available at: ">http://www.oe-a.org/web/guest/roadmap> [Accessed 12 February 2015].

Silver dispersion from Sigma-Aldrich, full spec available here, more on request: <http://www.sigmaaldrich.com/Graphics/COfAInfo/SigmaSAPQM/SPEC/73/736503/736503-BULK_____ALDRICH__.pdf> [Accessed 14 February 2015]

Suganuma K. Introduction to printed electronics [online]. New York: Springer Science+Business Media; 2014. Available at: < http://link.springer.com/book/10.1007/978-1-4614-9625-0. > [Accessed 19 April 2015].

Typographic Test Form: investigating the performance of free fonts

Natalia Lumby

Ryerson University, Toronto, Ontario, Canada

E-mail: nlumby@ryerson.ca

Short Abstract

The following study investigates the performance of free fonts. The graphic design industry is full of free crowdsourced resources such as vector illustrations, photos and fonts. However, generally we caution against the professional use of free fonts (Von Meissner, 2012). This study investigates the performance of free typefaces. Firstly, we identify current typographic trends in order to evaluate whether free fonts are consistent with what is popular. Secondly, we look at how the free fonts perform on output, using a test form designed specifically for this study. The performance of both decorative display and body text typefaces is measured and compared to paid typefaces that they mimic. In addition the 10 most popular free fonts are analysed. While free fonts did not perform as well as their paid counterparts, findings suggest that free fonts are getting closer to becoming a "safe" option from a technical standpoint so long as you understand the limitations.

Keywords: typography, free fonts, design trends, typographic test form

1. Introduction and background

The graphic design industry is under siege by the proliferation of many amateur designers providing sub-cost design services. Desktop publishing technology and our ability to crowdsource using the Internet have shifted how design work is sourced (No Spec, RGD). Furthermore, the availability and ease of use of design software, along with the popularity of graphic design degrees have significantly lowered the barriers to entry for this industry and saturated it with excess talent (Heller, 2005).

This shift has affected typography. It is very difficult to make a living as a typographer (Haley, nd). The democratization of type distribution (thanks to the Internet) has resulted in a deep pool of free typefaces on offer. Since the digitization of typography many amateur artists using various levels of skill and quality of software are contributing to this space (Shamir, 1997). Anecdotally, professionals ward off the use of free fonts for paid work (Von Meissner, 2005). There are thousands of free and paid fonts now available online, each contributing incrementally smaller differences in style (Hakki, 2013). This, along with the need to maintain tighter budgets has made the designer's mission to choose an appropriate typeface that much more difficult. With the rise of more open copyright options such as those provided by Creative Commons (creativecommons.ca), more resources are free to use, remix, and share. Right or wrong, there are supporters for this movement within typography. For example The League of Moveable Type is dedicated to encouraging artists to share fonts they design for free (with appropriate attribution). In addition, free designs have seen an overall increase in quality (Shoaf, 2014). This study will interrogate the performance of free typefaces thus helping designers make more informed font choices. While some studies exist to help designers consider the impact of the aesthetics of a typeface (Henderson, 2004; Childers and Jass, 2002; Koch, 2012), here we look at the ability of a font to perform from a technical perspective.

The research will begin with an investigation of recent typographic trends, which will allow us to determine whether free fonts are in keeping with the designs that are currently in demand. Preliminarily, this review will also allow us to look at the kinds of fonts people are buying in comparison to those they are downloading for free.

From a technical perspective, in order to test for typeface performance a tool needs to be developed. Currently there is a tremendous lack of research in typographic quality and standards (Dyer, 2014). The studies that do focus on type quality are typically divided into looking at the technical attributes of a font (Shamir, 1997; Karow, 1994), or the aesthetic attributes (Bringnurst, 2004; Jamra, 1993). Most of the technical studies emerged when typographic standards for digital fonts were emerging. Their contribution was primarily used to improve this transition, and thus aimed at typographers. This research however is aimed at designers, helping them to sift through the overwhelming variety of free typefaces using a technical lens. Other studies use software tools to categorize typefaces using a variety of attributes to aid in font selection (O'Donovan, 2014). Here the focus is on creating a test form that will allow designers to ascertain the limitations of a font they would like to use.

2. Methods

This study uses content analysis in order to establish whether free fonts are able to meet market demands. To accomplish this we compare the attributes of the most popular paid and free typefaces across several industry standard websites. Download statistics from online font libraries are a great indicator for popularity. Thus, we compare the 10 most popular fonts for 2014, sourced from veer.com and fonts.com, which host primarily professional paid fonts, with their 10 most popular counterparts from websites that focus on featuring free designs (dafont.com and 1001freefonts.com). A word cloud analysis is then used to categorize the key words describing the fonts. This allows us to interrogate whether designers are using free fonts for similarly purposed work.

One important issue around developing a method for this study is that it is difficult to point to specific criteria that determine the quality of a font. A typeface must first and foremost communicate a message (Hakki, 2013). "Letters are things, not pictures of things"—the implication is that typefaces "are meant to be read and not just looked at" (Carter, 1982). As such, many indicators of type quality focus on readability and legibility (Shamir, 1997; Hakki, 2013; Faiola, 2000: Paterson, 1931). Where legibility can be interpreted as the contrast in the letterforms and readability the consistency (Adams, 1978), we consider the former to fulfill this communication role, while the latter serves to attract the reader (Hakki, 2013). Rather than focusing on quality from a readability/legibility standpoint, this research seeks to develop a way to quickly decipher typeface limitations for the designer. It is meant to aid in font selection.

We categorize the research into two broad font categories, looking at body text and display/titling text. In addition to exploring results from the 10 most popular free typefaces on dafont.com, we also compare results of typefaces that are paid with their free tribute typefaces. This comparison is used to determine whether there is a performance difference between the free and paid typefaces using our metrics.

The following is an image of the test form created.

	ne: xx ted: mm/dd/yy sets used: xx
	SIZING *CREATE OUTLINES
8	Hamburgefonts
•	Hamburgefonts
12	Hamburgefonts

14 Hamburgefonts

24 Hamburgefonts

All ligatures Some ligatures Numbers Leading, word spacing Performs at small si Variety of weights & styles Numbers

DISPLAY TEXT

Some symbols & numbers

Performs at large sizes

Kerning

BODY TEXT

e & uppercas

Accents

All symbols

Hamburgefonts

^a Hamburgefonts

Hamburgefonts Hamburgef LIGATURES LEADING, WORD SPACING (10 PT)

Th fb ff ffb ffh ffi ffj ffk ffl fft fh fi fj fk fl ft Ťh ſb ſh ſi ſk ſl ſſ ſt ſſi ſll Ţh

TYPOGRAPHIC PERFORMANCE TEST FORM | 2015 © NATALIA LUMBY & ALYSSA KAYE ANDINO SYMBOLS !@#\$%^&*(),./\[]?{}<>|- $=_{+i}^{\circ} @ \pounds c I^{1/2} @ e^{\circ^2} @$ SMALL CAPS ABCDEFGHIJKLM NOPQRSTUVWXYZ ACCENTS áàâäå ÁÀÂÄÅ çç éèêë ÉÈÊË íìîï íìîï ññ óòôöõ óòôöõ úùûü ÚÙÛÜ ŸŸ WEIGHTS Regular Italic Bold **Bold Italic**

LEADING, WOHD SPACING (10 P1) Soluptatenti quickes paranquia nam que et quo dolor aute nobitibusam is quo conse nimaximus, quae. El iduci odit es autas que quo te niscis as porernatur? Ut quiaestem auta boritatem a voluta num faci voluptati ali cil millanditis esequasse nusania simped ut volorem facero evendaerchit aut quo illupta sectoresti scerum volum dit aut volorectur serum verum que velende mianisna latesen iniliore de por quate non repe latem racribus, quo iumque elessitia quis am fugiam acesciam, quae susandam evenis archi lipsunt experi officat usandebitiae odis est, ate dolupti aspera ipicipiciet eiur?

KERNING (METRIC) НОООНННОННООО KERNING (OPTICAL) НОООНННОННООО UPPERCASE LETTERS ABCDEFGHIJKLM

NOPQRSTUVWXYZ LOWERCASE LETTERS abcdefghijklmnopqrstuvwxyz

NUMBERS 1234567890

Figure 1: Typographic test form

The metrics the form uses are identified more explicitly in Table 1. These were selected on the basis of being identifiable on a test form by a designer. Given that body text and display text vary in their use, this list of attributes is also translated on the test form as a checklist, dependent on the use. Thus a designer can use the test form to qualify which typeface can be used for each purpose.

Measure	Information
Completeness of glyphs	The typeface includes:
	• Upper case characters
	• Lower case characters
	• Numbers
	• Accents
	Symbols
	• Ligatures
Size performance	The typeface is legible (i.e. the word can be read and understood
	easily) at both small and large sizes
Spacing	The typeface is legible when leading is unadjusted (set to auto).
	The word spacing promotes identifying separate words comfortably.
	Words with all capital as well as mixed case letters are well kerned
	metrically using a standard phrase (Willis et al., 2012)
Variety	The typeface includes a variety of weight and style alternates such as
	bold, italic, and etc.
Structure	The number of anchor points utilized to create the letterforms is
	limited to those necessary (only indicator evaluated digitally) (Shamir,
	1997)

Table 1: Test form metrics

Once created, the test form was output to both a soft and hard copy proof for inspection. Adobe Acrobat was used for the soft proof, while the EPSON 7900 output the hard copy proof via Esko Automation Engine 14.0. Lastly, the typeface was validated using a font management software tool (FontBook).

3. Results and Discussion

Firstly, in investigating the performance of free fonts, we must qualify the meaning of "free". Many fonts are "free for personal use" only. This "try before you buy" approach is also becoming popular within paid fonts, where the designer makes available one of the styles in a type family. All but one of the typefaces we analysed here are 100% free for both commercial and personal use.

Anecdotally new designers are encouraged to stay away from free typefaces. However, with type technology becoming more and more intuitive it is likely that these same designers are also creators of free typefaces themselves. In addition, we are seeing free typefaces appear in commercial work. Figure 2 shows two examples of free typefaces used commercially. In particular design templates have been very successful using free fonts as they can point clients do download typefaces without worrying about licensing and additional cost to their client.



Figure 2: Caviar Dreams on a book cover, and Bebas in a corporate brochure template (designed by TypoEdition)

In looking at visual trends we created word clouds of the key words used to identify the most popular fonts downloaded in 2014. Figure 3 identifies the words used to tag the most popular paid fonts on the Veer.com and fonts.com webpages.



Figure 3: Key words for most popular paid fonts

The free fonts analysed using the same technique revealed a very different trend picture. Here we can infer that designers primarily look to free typefaces for decorative applications.



Figure 4: Key words for most popular free fonts

In addition to looking at some key trends using download statistics as a metric for popularity, results of the test forms along with general font information were analysed. The top ten most downloaded free fonts available on dafont.com illustrate that while free fonts are improving in quality, they are still not preforming at the level of a paid typeface.

On the whole, free typefaces provide less variety and therefore flexibility. When comparing paid fonts to their free look-alikes we found that on average free faces have about half of the number of glyphs designed as part of the paid font. Stemming from missing glyphs were issues of missing symbols as well as ligatures. Further, a majority of the free typefaces only offered one weight or style. Where a paid typeface may have upwards of 6 styles offered (to as many as 27 in this sample). In cases where there were more options in the free fonts, the paid fonts were older—with the maturity of the font potentially being linked to the number of options existing.

The spacing of letters, words, and lines of type was also evaluated. Firstly, kerning pairs revealed that free fonts rely on optical kerning produced by the layout program in about 40% of the cases (Figure 5). Most of the kerning issues exist not as a result of the kerning tables used, but rather the design of the font itself. In the body text typefaces tested, free fonts performed well in terms of the default word spacing and leading. However, the display typefaces were not as successful. This is not necessarily an indicator of poor quality, as one should not use display type to set paragraphs.

KERNING (METRIC) HOOOHHHOHHOOO KERNING (OPTICAL) HOOOHHHOHHOOO

Figure 5: Example of kerning for a free typeface

Another indicator of quality we evaluated was the number of anchor points created when the typeface was converted to outlines. This measure was also used in a study by Shamir *et al.* (1997). Along with a lack of variety, this seemed to be a clear mark of difference between free and paid counterparts— particularly for display typefaces. The free font outlines used many more anchors. This in turn meant that they did not perform as well in very small (8pt) and very large (144pt) sizes. In small sizes the letters began to fill in, while in large sizes the sloppiness of the vectors became evident. Figure 6 shows a sample of a paid font and free font anchor comparison. Figure 7 is an example of type breaking down at a large size. This is interesting because as previously mentioned most free typefaces are display/titling and therefore should perform well at these larger sizes.

The quick The quick

The quick The quick

Figure 6: Comparison of font outlines between free fonts (top) and similar paid counterparts (bottom)



Figure 7: Sizing issues resulting in untidy outlines

It should be noted that while professionals may question the design quality of free fonts, from a technical perspective both the body text and display fonts would perform well in most cases. This could be the result of increasingly intuitive font technology. Using a validation check in Font Book revealed 23 minor warnings among 122 tested fonts. The warnings consisted of faulty kerning information as well as naming issues. These are issues tied to versioning as well as the operating

system being used (Mang, 2006). While all of the warnings were associated with the free fonts tested, none prevented the font from outputting outright.

4. Conclusions

Designers must understand that they need to be careful building a brand around a typeface that is incomplete. Particularly important with more easily identifiable display fonts, which cannot easily be swapped out later as the brand matures. At the same time the presence and availability of free fonts cannot be ignored. The free fonts tested in this study have cumulatively been downloaded over 10 million times. When considering that the price of each comparator typeface in this study averages about \$8—that is a cost savings of \$80 million.

From the perspective of current design trends, given the options available, it is likely that free typographic designs will be predominantly used for titling and decorative work. Here free fonts are likely to thrive because some of the obstacles discussed above are eliminated. For example most titles do not require special characters. Furthermore, because most titles span a single line, the need for leading is not common, kerning can be quickly adjusted, and the lack of style/weight options can be overcome with mixing in other free typefaces.

The sheer variety and playfulness of free fonts available are likely to be positive factors in their selection. Certainly from a trend perspective free fonts seem to be catering to titling/decorative work. Therefore, even though free fonts may not test well across the board using a complete test form tool, they may still have niche benefits. As the quality of free fonts continues to improve, it is likely that we will continue to see a greater mix of free and paid typefaces being used together to produce impactful designs.

Acknowledgments

I would like to acknowledge, with most sincere gratitude, my research assistant Alyssa Kaye Andino, for her hard work and unwavering enthusiasm for this project. Thank you as well to Gillian Mothersill for sharing with us her boundless typographic wisdom. This work could not have been completed without the support of the Faculty of Communication and Design, as well as the research office at Ryerson University.

References

20 Best Free Fonts of Year 2014. (2014, August 23). Retrieved February 15, 2015, from http://www.designscanyon.com/best-free-fonts/

Bringhurst, R. (2004). The elements of typographic style 3rd edition. Vancouver: Hartley & Marks.

Carter, M. (1982) What Is A Typeface? *The Printing Salesman's Herald Book 44*. New York: Champion Papers, Champion International Corporation

Childers, T. L., & Jass, J. (2002). All dressed up with something to say: Effects of typeface semantic associations on brand perceptions and consumer memory. *Journal of Consumer Psychology*, *12*(2), 93-106.

Dyer, I. C. (2014). An Examination of Typographic Standards and Their Relevance to Contemporary User-Centred Web and Application Design. In Cross-Cultural Design (pp. 140-149). Springer International Publishing.

Faisal, M. (2014). *45 Best Free Fonts for Designers*. Retrieved February 15, 2015, from http://graphicdesignjunction.com/2014/08/free-fonts-best-of-2014/

Faiola, A. (2000). Typography primer. GATFPress.

Hakki, I. (2013). The Effects Of Font Type Choosing On Visual Perception And Visual Communication. *Online Journal of Art & Design*, 1(3), 35.

Haley, A. (2013, February 15). *Type Trends: Hot Faces & Cool Type*. Retrieved February 14, 2015, from http://www.howdesign.com/design-creativity/fonts-typography/cool-type-2013/

Haley, A. (n.d.). Cold Cash for Hot Fonts. Retrieved February 15, 2015, from <u>http://www.commarts.com/columns/cold-cash-hot-fonts</u>

Heller, S. (2004, August 4). Crimes Against Typography. Retrieved February 15, 2015, from <u>http://www.aiga.org/crimes-against-typography/</u>

Heller, S. (2005, September 8). *Too Many Grads or Too Few Competencies? The Design School Dilemma*. Retrieved February 15, 2015, from <u>http://www.aiga.org/too-many-grads-or-too-few-competencies-the-design-school-dilemma/</u>

Henderson, P. W., Giese, J. L., & Cote, J. A. (2004). Impression management using typeface design. *Journal of Marketing*, 68(4), 60-72.

Jamra, M. (1993, November). Some elements of proportion and optical image support in a typeface. In *Visual and technical aspects of type* (pp. 47-55). Cambridge University Press.

Karow, P. (1994). Font technology (pp. 85-104). Springer Berlin Heidelberg.

Koch, B. E. (2012). Emotion in Typographic Design: An Empirical Examination. Visible Language, 46(3), 206.

Mang. (2006). ttf font on OSX - name table problem?. Retreived May 25, 2015 from <u>http://forum.high-logic.com/viewtopic.php?t=1414</u>

Marra, R. M., Moore, J. L., & Klimczak, A. K. (2004). Content analysis of online discussion forums: A comparative analysis of protocols. *Educational Technology Research and Development*, 52(2), 23-40.

Meissner, G. (2012, July 30). *How to navigate free fonts for your next design project*. Retrieved February 15, 2015, from <u>http://99designs.com/designer-blog/2012/07/30/typefaces-the-good-the-bad-and-the-legal/</u>

No Spec. (n.d.). Retrieved February 15, 2015, from http://www.rgd.ca/resources/no-spec

O'Donovan, P., Lībeks, J., Agarwala, A., & Hertzmann, A. (2014). Exploratory font selection using crowdsourced attributes. *ACM Transactions on Graphics (TOG)*, *33*(4), 92.

Paterson, D. G., & Tinker, M. A. (1931). Studies of typographical factors influencing speed of reading. *Journal of Applied Psychology*, 15(3), 241.

Shamir, A., & Rappoport, A. (1997). Quality enhancements of digital outline fonts. *Computers & Graphics*, 21(6), 713-725.

Shoaf, J. (2014, March 12). Taking A Second Look At Free Fonts. Retrieved February 15, 2015, from <u>http://www.smashingmagazine.com/2014/03/12/taking-a-second-look-at-free-fonts/</u>

Willis, N., Adams, V., Sorkin, E., Pagura, J., Martin, B., Skala, M., Brilliant, M. (2012). Design with FontForge. Retreived May 22, 2015 from <u>http://designwithfontforge.com/en-US/Spacing_Metrics_and_Kerning.html</u>

Innovative Combinations of Print & Digital – attitudes towards change in the European printing industry

Markéta Držková¹, Sanne Tiekstra², Paula Miranda³, Pedro Isaías³, Igor Karlović⁴, Rozália Szentgyörgyvölgyi⁵, Tadeja Muck⁶, Kaisa Vehmas⁷, Claire Gauzente⁸, Anu Seisto⁷

- ³ IADIS, Portugal, <u>pcrmiranda@gmail.com</u>, pedroisaias@gmail.com
 ⁴ Department of Graphic Engineering and Design, Faculty of Technical Sciences, University of Novi Sad, Serbia, karlovic@uns.ac.rs
- ⁵ Óbuda University, Faculty of Light Industry and Environmental Protection Engineering, Hungary, szentgyorgyvolgyi.rozsa@rkk.uni-obuda.hu
- ⁶ Faculty of Natural Sciences and Engineering, University of Ljubljana, Slovenia, tadeja.muck@ntf.uni-lj.si
- VTT Technical Research Centre, Finland, kaisa.vehmas@vtt.fi, anu.seisto@vtt.fi
- ⁸ LEMNA, University of Nantes, France, claire.gauzente@univ-nantes.fr

Short Abstract

The present study reports a European industry survey of the state and future of innovative printing. Main emphasis addresses how paper and board and graphics industry could benefit the most from the possibilities provided by combining print with digital and by printed functionality. Experiences and inputs from representatives of the industry were collected to provide viewpoints on how print media and packaging could be developed and used in the future and how the industry can utilize available knowledge in science for the benefit of their customers and the consumers.

First innovation adaption process and methodologies are reviewed and then the available empirical results are described and analyzed. It can be suggested that (1) European graphical companies see their future in innovative printing, (2) beside costs, lack of market demand is the most important factor in preventing companies to move to innovative printing, and (3) approach and vision of the industry is in general similar across Europe although Western European printing houses seem to be more active in the field of innovative printing, while innovations utilized today by printers in Eastern Europe mainly comprise effect and personalized printing, indicating a delay in implementation of available technologies.

Keywords: Innovation, Printing, Europe, Prospective study

1. Introduction and background

Innovation has long been recognized as a key element of economic development. Today there is a great need for understanding the state of the printing industry and providing further direction on this topic that entails both technological and strategic dimensions. To achieve this, various aspects of diffusion of innovation need to be considered and put into relation with current situation of innovative printing in Europe.

Academic literature about innovation is plentiful; many research contributions exist that characterize different aspects and originate from different disciplines, as summarized by Connolly, Gauzente and Dumoulin (2012). They state that in economic literature the focus has been 'technology push' (attempts to commercialize and increase diffusion of the innovation) versus 'demand pull' (user need for the technological innovation). Looking at different aspects of innovation, they discuss that uncertainty and perceived risk play an important role from the perspective of potential adopters. The benefit might not always be obvious, thus one cannot be sure that the innovation is preferable to the existing product. Nor is the consequence, increasing the perception of risk and uncertainty. In addition, resistance to change is a powerful factor that hinders the adoption of innovations. As far back as 1964,

¹ Department of Graphic Arts and Photophysics, Faculty of Chemical Technology, University of Pardubice, Czech Republic, marketa.drzkova@upce.cz

Bumaga BV, Netherlands, s.tiekstra@bumaga.nl

Bright observed that "Anyone introducing a technological innovation is implicitly or explicitly predicting acceptance and a rate of adoption. Yet a fact of technological history is that many innovations are subject to frustrating delays and deliberate resistances to adoption" (1964: 171).

2. Printing industry in Europe

Nowadays, the European printing industry has around 120,000 (mainly small sized) companies, employing around 750,000 people for a turnover of about €85 billion (Eurostat, 2015). On the other hand, according to Confederation of European Paper Industries (CEPI, 2015), its members produced 91 Mt paper and board in 2014. There are around 630 companies active in pulp, paper and board industry in Europe (excluding paper and board mills), generating a turnover of approximately €75 billion and more than 180,000 workplaces (CEPI, 2014).

While the paper and board industry is struggling with stagnation or very slow growth in some countries, many printers are currently facing overcapacity problems, price competition and replacement of print by digital (Intergraf, 2011) which they try to overcome by cutting costs, investment in new technologies and equipment. All Intergraf's respondents agreed that the main investment needs to be put in new knowledge. These new opportunities came from maturing of several important innovations which emerged along time, like digital printing, printing on demand, 3D printing, augmented reality, and printed electronics. Numerous aspects of the last one, important for developments in paper and board as well as printing industry, are discussed in literature (e.g. Bollström et al., 2014; Pettersson et al., 2014; Määttänen et al., 2010). Innovative technologies enable increased productivity, new markets, products and services and open up opportunities to integrate services along the value chain, and eventually even reduce costs (PMG, 2012).

According to Vehmas et al. (2011), printing houses still are not willing to move to completely new business areas due to the huge investments and R&D needed for old printing machines to produce totally new products and the risk is seen to be too high in new customer markets. Drivers for enormous change are missing and new business cannot therefore evolve, but even if giant leaps are not taken, many actors in the European printing industry had recognized the need to transfer within the current business area. Three approaches to survive were identified:

- Efficient web printing production via flexible production and effective materials usage
- Added value for the printed product
- Printed non-media products.

In all cases customer service, co-operation in print production chain and open communication are needed to succeed.

A report by Aistrup (2009) states that collaboration is important as companies cannot handle the cascade of complex knowledge and they should cooperate in complementary innovation networks to expand value rather than simply improving their existing value share. Currently many segments of the printed electronics are not profitable with mostly more than 10 years of development: as the profitable ones are identified electroluminescent displays, sensors and conductive inks (IDTechEX, 2015).

In this study, the goal was to promote discussion between industry and academia on the benefits that may arise from combining print and digital. Several examples exist where successful combinations have been applied e.g. through the use of image recognition, augmented reality or printed electronics to bring interactivity into fiber based products. Experiences and inputs are collected to provide viewpoints on how print media could be developed and used in the future, how industry can utilize the information for the benefit of their customers, and to find out the awareness of the possibilities for combining print with digital as well as the attitude of the industry towards these technologies.

3. Materials and Methods

To initiate this discussion, a questionnaire was set up to assess industry's opinion on innovative printing, referring to printed electronics, printed intelligence, printed functionalities, combining print with digital, etc. In each country (Czech Republic, Hungary, Netherlands, Portugal, Serbia, Slovenia, Slovakia, and United Kingdom), the composed questionnaire was distributed among the targeted companies in their local language in order to overcome any language barriers and to maximize understanding. Further, the English version reached respondents in several other Western European countries (Belgium, Germany, Italy, Norway, and Sweden); the origin of these responses is jointly designated as "Other" in following text as the number of responses per country was too low to analyze them separately.

3.1 Survey instrument – questionnaire

The questionnaire consisted of six questions, both open and closed. First question was meant to characterize the companies based on e.g. company size and printing technology. The second question inventoried the use of innovative printing techniques, after which questions three and four tried to identify the most important limitations to develop such printing solutions and the presence of competitive and financial advantage. Question five gave the opportunity for the industry to express their interest and opinion on cooperation with academia. Finally, the last question requested the outlook and opinion of the industry into the fastest growing markets in the area of innovative printing.

Intentionally, the questionnaire remained brief in order to increase responses and willingness of companies to participate.

Country	Online questionnaire	E-mail questionnaire	Phone conversation	Personal conversation
Czech Republic (CZ)	✓ (B, C)		\checkmark	
Hungary (HU)	✓ (A, C)			
Netherlands (NL)	✓			✓
Portugal (PT)	✓ (D)	✓ (D)		
Serbia (RS)	✓		✓	
Slovenia (SI)		✓		
Slovakia (SK)	✓			
United Kingdom (UK) and Others	\checkmark	\checkmark		

Table 1: Survey Instrument

A - Posted on a web portal, B - Link distributed by e-mail to general contact addresses,

C – Link distributed by e-mail addressed to company representatives,

D – With a previous phone contact with the company representative

Various ways were employed for gathering results (as summarized in Table 1) to optimize the effectiveness of the study and to receive the maximum amount of responses, especially in countries where the target group was relatively small. More detailed answers were possible in personal or phone conversation, in some occasions only used as a follow up. In all cases however, the questions remained the same.

3.2 Sample size and characteristics

The study aimed at a well spread sample size across the European countries. To date, over 190 companies participated; both Western and Eastern European countries (Figure 1a), and bigger as well as small enterprises (Figure 1b). Predominantly, the sample was random, although in some cases it was known beforehand that the printers are working on innovative printing.

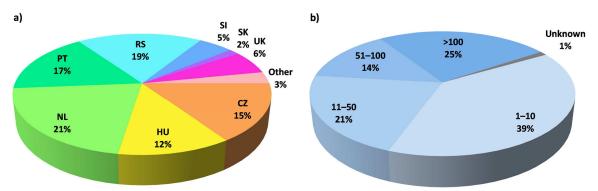


Figure 1. Respondents' country of origin (a) and number of employees amongst the respondents (b)

In terms of the distribution of the respondents throughout the different countries, the current results show that the majority of the respondents come from the Netherlands, Serbia, Portugal, the Czech Republic and Hungary. The cleavage between the countries representation seems to be, at first instance, result of different schedules and instruments for administering the surveys. In addition, since the graphical industry is not the same in each country, it is rather impossible to target the same amount of responses in each country. Finally, some interviewees just seemed more active and willing to answer. For all these reasons, the aim was to have as many responses as possible per country. To date, the total amount of responses considered as sufficient for quantitative analysis was not reached and is in fact expected to be impossible to be reached per individual country, because the authors contacted actual industry. Nevertheless, monitoring the survey progress suggests that it would give very similar results, as the patterns in current results for almost 200 respondents are very close to those obtained with half of this number. It must be concluded that the survey results are a fair representation of the printing industry throughout Europe. However, the data collection that started in 2014 is still ongoing and will be extended beyond what is presented in this paper.

With regards to the size of the companies, so far in this research, the respondents come mainly from small companies with 1–10 employees (39%). Nonetheless the other categories of companies' sizes are also sufficiently represented. The respondents also provided a diverse representation of industry types, ranging from general printing over packaging and security printing to digital media or paper production and converting.

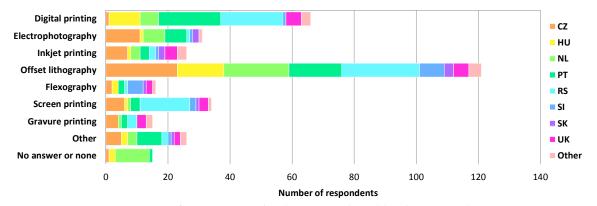


Figure 2. Printing technologies employed by the respondents

Figure 2 shows the variety of printing technologies that the participants employ. This clearly identifies offset lithography as the most popular printing technique along with the digital printing in general and electrophotography or inkjet printing in particular (in total listed by 123 respondents). From the main printing technologies, flexography and gravure printing are shown as the techniques that the participants use the least.

4. Results and Discussion

The results indicate that all respondents (no matter the extent to which they are aware of new technologies) would welcome more info on this subject. Obviously, printers are open towards cooperation with academia (see Figure 3), for example by local meetings to moderately discuss relevant topics. The majority of the respondents are interested in training and short courses, would like to participate in research partnerships and believe that the academia plays a valuable part in terms of specific measurements.

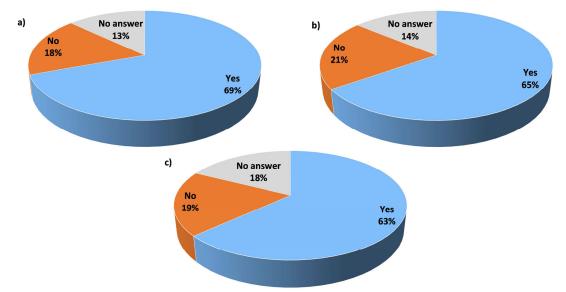


Figure 3. Respondents' attitude towards cooperation with academia – interested in training seminars and short courses (a), in research partnership (b), and in measurements (c)

During the past years printing industry has crossed structural changes, e.g. consolidation and closing overcapacity. Still, printing houses have a strong faith on prevalence of printed media products, but they also understood that some changes need to be done to survive (Vehmas et al., 2011). Consequently, based on available results (Figure 4), 40 % of the respondents offer innovative printing (as defined in 3. Materials and Methods), while the half did not yet do so. According to this study, printing houses in Western Europe seem to be more active in this field, while innovations utilized today by printers in Eastern Europe mainly comprise effect and personalized printing. This indicates a delay in implementing technologies related to printed functionalities and combining print with digital. However, the limitations of these innovations perceived in surveyed countries do not copy this scheme and their distribution is more similar in all countries.

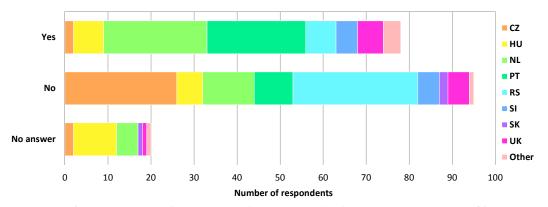


Figure 4. Responses to the question 'Are you involved in innovative printing?'

Most mentioned limitations (Figure 5) are the lack of market demand and costs related to new equipment, new skill sets or training of employees etc. The lack of market demand was stressed by many respondents as the key factor. In this context, the costs on customer side were mentioned as well as previously mentioned by Vehmas et al. (2011). The responses of the participants showed that in the great majority of the cases, their customers are exerting little or no pressure for innovative printing. About one third of the respondents did answer that the specialties are beneficial to the company's turnover (Figure 6).

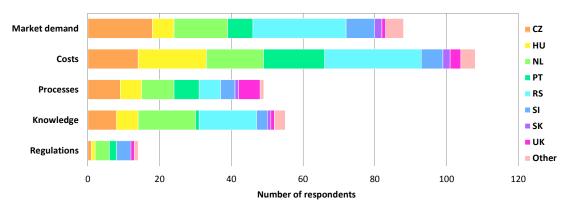


Figure 5. Limitations to use innovative printing as perceived by the respondents

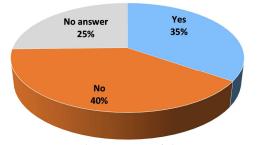


Figure 6. Distribution of the answers to the question if the companies gained competitive advantage and increased margins due to innovative technologies

As for the manner of receiving responses, a combination of different methods works best. Even though personal conversation is subjective, it gives a bit more detail to the answers than an online questionnaire.

5. Conclusions

This study does in a sense show the 'old truths', discussed in the introduction, as still valid in today's printing. The combination of lack of market demand with the perception that innovative printing is expensive and complex may be at the origin of the shy investment of certain industries in this area. On the other hand, nearly 200 industrial respondents took the time to reply to the questionnaires, which shows their commitment to the subject and innovation in general. In contrast to often presented opinion that the markets in question are open mostly for specialized and agile companies outside printing sector or only to the strongest but at the same time flexible printers, it was shown that innovations more or less related to printed electronics and combining print with digital are considered also by some traditional printers belonging to SMEs. Despite all the limitations associated with innovative printing, one of the main conclusions is the fact that companies are manifesting their interest in education in this area, as well as their willingness to take part in research partnerships. This predisposition towards innovative printing, might, in the long term, work in favor of this area.

Furthermore, even though all countries differ in the progress they made in innovation or technology, the approach and vision of the industry in general is similar across Europe. New technology is not the bottleneck. The challenge is to fit new technology to future requirements and business concepts. R&D and cooperation between different partners is needed to reach future goals. Since most of the respondents use paper and board as a substrate, this willingness to innovate also creates new opportunities for the European paper and board industry and help sustain the future of the graphical industry overall.

This activity driven by participants of COST Action FP1104 will continue to strengthen the cooperation between printing industry and academia and to gather in depth feedback on the survey results. Within COST Action FP1405, focusing towards active and intelligent packaging, industry will be continuously motivated to invest and develop. This will also enable the researchers to revisit the current studies, get more in-depth feedback on drivers for change, and focus on patterns, relations or discrepancies among the results.

Acknowledgments

This work is done in scope of COST Action FP1104, New possibilities for print media and packaging – combining print with digital, for which COST Association is gratefully acknowledged. Janet Preston (Imerys Minerals Ltd., United Kingdom), Beatrice Klose (Intergraf), John Charnock (Print Research International Ltd., United Kingdom) and Tim Claypole (WCPC, United Kingdom) are thanked for cooperation on the survey. Regina Connolly (Dublin City University, Ireland) is acknowledged for her input on innovation theories. The authors give special thanks to all industry representatives who took part in the survey.

References

Aistrup Consulting, (2009). *Linking into the Value Chain for the Printed Electronics Industry*. [online]. http://www.aistrupconsulting.com/resources/linkingintothevaluechain_printedelectronics.pdf

Bollström, R., Pettersson, F., Dolietis, P., Preston, J., Österbacka, R. and Toivakka M., 2014. Impact of humidity on functionality of on-paper printed electronics. *Nanotechnology*, 25(9), 094003

Bright, J.R., 1964. Research, development and technological innovation, Illinois: Richard Irwin Inc.

CEPI, 2014. *Key Statistics 2013 European Pulp and Paper Industry*. [online] http://www.cepi.org system files/public/documents/publications/statistics/2014/Final%20Key%20statistics%202 013.pdf

CEPI, 2015. *Preliminary Statistics 2014*. [online] http://www.cepi.org/system/files/public/documents/publications/statistics/2015/Preliminary%20Final%202014.pdf

Connolly, R., Gauzente, C. and Dumoulin, R., 2012. Adopting IT in public healthcare sector : an institutional research agenda. In: Nunes, M. B., Isaías, P. and Powell, P., eds. *Proceedings of the IADIS International Conference Information Systems*. http://www.iadisportal.org/information-systems-2012-proceedings

IDTechEX, 2015. Printed, Organic & Flexible Electronics: Forecasts, Players & Opportunities 2015-2025

Eurostat, 2015. Annual detailed enterprise statistics for industry (NACE Rev. 2, B-E), Most recent data: 2013 [online] http://ec.europa.eu/eurostat/web/products-datasets/-/sbs_na_ind_r2 [Accessed 24 April 2015]

Intergraf, 2011. *The Future of the European printing industry-In our own hands, What the industry says*. [online] http://www.intergraf.eu/images/SociallyResponsible_Study_ENG.pdf [Accessed 13 February 2015]

Määttänen, A., Ihalainen, P., Bollström, R., Toivakka, M. and Peltonen, J., 2010. Wetting and print quality study of an inkjet-printed poly(3-hexylthiophene) on pigment coated papers. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 367(1–3), pp. 76–84

Pettersson, F., Österbacka, R., Koskela, J., Kilpelä, A., Remonen, T., Zhang, Y., Inkinen, S., Wilén, C.-E., Bollström, R., Toivakka, M., Määttänen, A., Ihalainen, P. and Peltonen, J., 2014. Ion-modulated transistors on paper using phase-separated semiconductor/insulator blends. *MRS Communications*, 4(2), pp. 51–55

PMG, 2012. *Print Media in Times of Change, A Key Contributor to EU 2020.* [online] http://www.intergraf.eu/images/PMG_WEB.pdf

Vehmas, K., Kariniemi, M., Linna, H., Jokiaho, K. and Torniainen, E., 2011. *Future of European Printing Industry*. Espoo: VTT publication. 9 p.

42nd International Research Conference, 06 to 09 September 2015



Session 1B

Printing process

Monday, 7 September 2015 11:05 – 12:45

Chair: Gorazd Golob

Ink-Water Balance during Emulsification and its Relation to Substrate Transfer and Optical Properties of Prints

Carl-Mikael Tåg¹, Pertti Silfsten², Cathy Ridgway³, Kai-Erik Peiponen², Patrick A.C. Gane^{3, 4}

¹Stora Enso Support Centre Mönchengladbach, DE-41600, Mönchengladbach, Germany

²Institute of Photonics, University of Eastern Finland, Joensuu Campus, P. O. Box 111, FI-80101 Joensuu, Finland

³Omya International AG, CH-4665 Oftringen, Switzerland

⁴School of Chemical Technology, Forest Products Technology, Aalto University, 00076 Aalto, Finland

Short Abstract

An ink rheology testing technique (TackOscope), incorporating the possibility to apply an aqueous liquid (print fountain solution) to mix in an oil-based ink to create an emulsion in a twin roll nip, was used to provide information relating to the tack property during emulsification. By expressing this as a function of the emulsion component ratio, it was possible to study the influence on the optical properties of the final print. Internal cohesion of the ink-fountain solution emulsion is recorded as film split force between the two rollers during fountain solution titration and evaporation, and defines the intrinsic tack. Prints were made on plastic films, with a defined amount of ink with fountain solution addition loaded on the print application roller. Resultant print properties included, for example, printed gloss, which was seen to decrease with increasing fountain solution amount. In practice this can be reasonably strongly correlated to an increase in fountain solution consumption on the press. This indicates that the emulsion properties can be primarily expressed in terms of instantaneous tack of the ink-fountain solution film, and this technique can be used to monitor the role of the ink-water balance in the printing nip in controlling ink transfer dynamics, amount and final print result. The tack parameter was correlated over all conditions of emulsification. The ink used showed a continuous tack increase over time. However, superposed on this trend, addition of intermediate amounts of fountain solution was shown to decrease the tack of the emulsion monotonically.

Keywords: Emulsification, print gloss, printing, ink transfer, roughness, ink tack

1. Introduction

The aim of this study is to determine how the inks having varying content of fountain solution, parameterised in respect to tack, influence the optical properties of prints. This enables the boundary limits to be accommodated within a realistic range of tack. In turn, the recording of tack in this way itself can be used to monitor actual fount content, and so provide in principle a method to analyse the role of emulsification and emulsion stability on the press.

To the authors' knowledge there is no device existing, where the emulsified ink can be printed on a substrate in laboratory scale. The simple reason for this is that a restricted sample volume spread in the form of thin films on application rollers in non-equilibrium isolation exposed to the ambient environment changes in composition more rapidly than the experiment can be performed. Nonetheless, it is possible to measure the tack force for an ink over a range of fountain solution content incorporated as an emulsion, tracking the role of evaporation. This enables the limitations in correlating tack directly with the abovementioned end print properties to be accommodated within the range of application of tack, itself enabling the definition of actual fount content, and so providing the ability to understand the role of emulsification and emulsion stability on the print outcome. Thus, since the ink tack is not directly correlated with print properties, due to the evaporation limitations in transfer, rather the influence of the procedure to record ink tack as a function of added fountain

solution is analysed, and in turn accomplishes the second aim of the study. In which case, the apparent fount amount in the ink is related to the running/spraying time applied on the device, and monitored from the ink tack. The same ink-fountain solution emulsions were then printed on a plastic film without intermediate transfer using the same device, whereafter the optical properties were determined. Using the methodology described above, it is possible to define the role of "emulsification degree" on the optical properties of the prints.

2. Materials and Methods

The heatset ink used was a low tack variant (Premoking 6000 supplied by Flint Group), and the fountain solution contained 5 % of isopropylalcohol and 4 % of a fount additive.

The TackOscope device is based on a set of contacting rotating rollers. This method comprises a system similar to that of an ink distribution chain on a printing press, with an additional feature for applying fountain solution in a controlled manner by using a precise ultrasonic spray dampening. The device is built on the platform of a standard tack meter. The test result is presented as a "fount consumption curve" for a given ink/fount combination.

The printed samples were prepared in laboratory scale by application of an ink-fountain solution emulsion onto a plastic film with a nip contact method applied in the same device (TackOscope) as the emulsions were prepared, using a novel simultaneous twin substrate method. The ink distribution device was run so that an increasing amount of fountain solution was added to the ink prior to printing, thus obtaining prints with different fountain solution content. The monitoring of tack at different fount addition levels showed changes in ink-fount behaviour. Due to evaporation from the thin film distribution, the quantitative amount of fountain solution was not known, but the tack was recordable and therefore could in principle be back-related to a pre-scaled relative fountain solution amount. A deeper look into this will be conducted in a separate study. After printing, the prints were air dried under ambient conditions.

A quantity of 4 ml printing ink was placed on the TackOscope distribution rollers. The transfer roller was made of brass and the impression cylinder was from rubber. The TackOscope was set at a speed of 50 m.min⁻¹ and held at 30 °C during ink distribution. During the first 30 s, the ink becomes evenly distributed on the roller surfaces, after which the actual measurement is started and the speed increased to 200 m.min⁻¹. Depending on the desired amount of fount to be added, the machine was run for a corresponding time to apply the chosen volume from the spraying unit, which delivered fount at the rate of 10 µls⁻¹. The used plastic film was a Soennecken 5504, supplied for black and white copying and laser printers. The thickness was 0.1 mm. Two plastic films were put back to back one on top of the other through the TackOscope nip, thus enabling one side printing whilst maintaining a balance contact with both roller surfaces. After reaching the desired fount concentration point, ignoring evaporation, the contacting films were passed through the nip immediately to transfer the pre-formed ink-fount emulsion onto the film and allowed to air dry under ambient conditions. It can be assumed that the emulsion droplet size varies somewhat due to pressure gradients during levelling of the emulsion on the film. Any analysis of the heterogeneity in the dried print due to emulsion droplet size was not conducted. A schematic picture of the procedure is shown in Figure 1, where the adoption of two sheets of substrate, placed back to back, enables two one-side prints to be made, each contacting the respective upper or lower inked roller only.

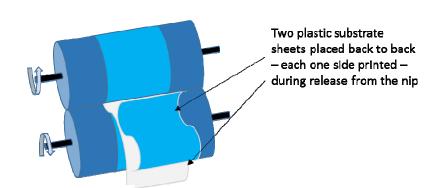


Figure 1: Procedure of printing the ink-fount emulsion.

When dry, the optical and the structural properties of the films were measured. The ink film thickness was determined from scanning electron microscopy images taken in cross-section. Since the sequencing was maintained the same for all trial points, and the exposure to evaporation was always the same, allowing us to claim that the series of different fountain solution contents follow a linear trend with the amount of solution added.

The gloss maps were measured using a *Diffractive Optical Element Glossmeter* μ DOG 2D scanning glossmeter [Juuti, *et al.*, 2007; Juuti, *et al.*, 2008]. The light source is a semiconductor laser (= 5 mW), with a wavelength of 635 nm, and the detector is a photodiode. The angle of incidence for this device is 0°, i.e. normal to the surface. The focus beam size of the laser is 30 µm diameter at the 1/*e*-level of the maximum irradiance of the light beam. The samples were measured by scanning 5 x 5 mm² area with a 5 µm interval resolution. As the calibration of the device is performed with a black glass gloss standard, the mean values calculated from the gloss maps represent the gloss of the samples. As all the samples have relatively high transmittance with the measurement wavelength, particularly given the geometry of normal incidence, the measured gloss value is also affected by the back surface reflectance of the sample.

The *Lightness* measurements from the prints were conducted with a Perkin-Elmer Lambda 18 spectrophotometer with a 150 mm integrating sphere. The spectra show only the diffuse scattered light component meaning the specular component is excluded. The lightness parameter L^* is calculated separately for each spectrum from the transmission value. This has a linear correlation to the lightness differences observable by the human eye. The light source in the calculations was a standard light corresponding to a 500 W tungsten light with a light temperature of 2 850 K, having no interactional effect on the calculated results.

The surface roughness was determined with *Confocal Light Microscopy*. This is an optical imaging technique designed to increase optical resolution of a light micrograph by using point illumination from a laser and a position adjustable spatial pinhole to eliminate out-of-focus light. A stack of micrographs at different height-levels enables the reconstruction of three-dimensional (3D) surface structures z(x,y). Additionally, the 3D-reconstruction allows the calculation of micro-roughness by the following formula:

$$R_{q} = \sqrt{\frac{1}{MN} \sum_{m=1}^{M} \sum_{n=1}^{N} \left(z(x_{m}, y_{n}) - \langle z \rangle \right)^{2}}$$
[1]

where (M, N) define the limits of the data collection point pairs (m, n) over the xy plane.

3. Results and Discussion

Figure 2 presents the development of tack with time with a constant dosage rate of fountain solution. It can be seen that the tack first of all increases under shear in the nip due to the progressively increasing film thickness with the ink viscosity dominating the mix. However, at the point where the fount content within the ink destroys its cohesive structure a collapse is seen where the tack is clearly reduced. For this test series, at 60 s of fount spraying, the tack is levelling out, and the critical point, referring to tack drop, is seen at 85 s, corresponding to a fount delivery of 1 ml in the ink (25 wt-% of the original ink amount). This fount delivery, however, is not the absolute amount as the evaporation was not taken into account.

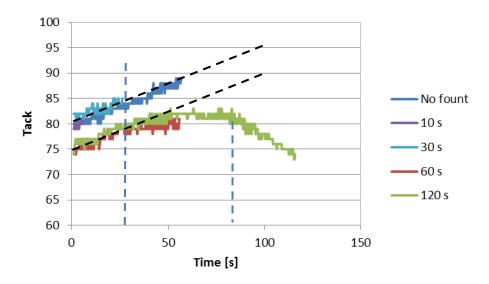


Figure 2 Ink tack as a function of fountain solution dosage time at constant rate: evaporation effects not included.

It is clear that the tack measure in Figure 2 gives similar-shaped curves for all addition conditions over a given time range, in the sense that tack first rises due to the intrinsic aging of the ink under shear and then decreases to a minimum during fountain solution addition due to emulsified droplets lowering ink film cohesion, and any non-emulsified surface water that might be present creating a weak boundary layer.

It should be noted that the use in this study of impermeable, non-absorbing plastic film acts to increase the negative role of fountain solution. It must be assumed, therefore, that the application onto absorbent substrates, such as uncoated and coated paper and board, acts to allow fountain solution to be separately absorbed or permeated under pressure into the substrate.

3.1 Surface roughness of the printed ink films

Figure 3 shows that an increase in emulsification degree affects the surface of the prints, a higher loading increasing surface roughness. The fountain solution most likely creates cavities in the splitting ink film resulting in a rougher surface on drying, i.e. phase separation. The formation of cavities in the ink film is a prelude for splitting of the film in the nip. However, an excess amount of fount might cause trapped fount in the ink resulting in air pockets when dried, thus making the surface rougher. As well, an increased amount of fount within the ink reduces the viscosity of the applied ink-fount emulsion, also affecting the penetration of ink into the substrate if porous (which is not the case in this instance), and surface levelling – a dominant factor in this case.

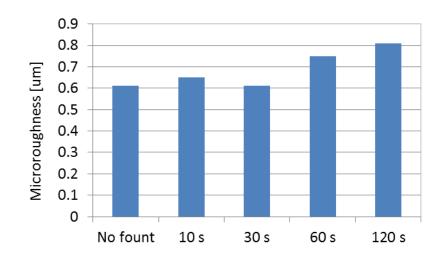


Figure 3. Surface micro-roughness of prints after varying fount spraying time, quoted as time in seconds [s] of fountain solution dosing at constant rate.

3.2 Gloss measurements

Gloss maps were measured using the diffractive optical element based glossmeter (Myller *et al.*, 2003). From the gloss maps, an average gloss value was calculated. It has been shown that if the ink film thickness is above 2 μ m, it can be assumed that there is nearly no backside reflectance of the plastic film affecting the results [Niskanen *et al.*, 2007]. The μ DOG gloss values were scaled so that a black glass plate gives the value of 100. Figure 4 presents the variation in gloss over the measured area including transmittance and sample reverse side reflectance, which is assumed to have a constant boundary roughness.

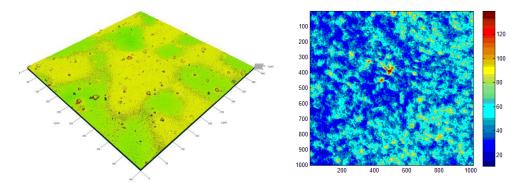


Figure 4. Gloss maps. The gloss is colour coded ranging from blue at low level to red at high.

Figure 5 shows that the gloss level is decreasing with higher emulsification degree. Especially the sample with overdose of fountain solution leading to emulsion inversion from water-in-ink to ink-in-water changes the light reflectance properties from the applied film, which upon drying resembles that of water on hydrophobe reticulation, drastically reducing gloss. Most likely the resulting cavities increase the diffuse reflection thus also lowering the specular gloss at high fount loading.

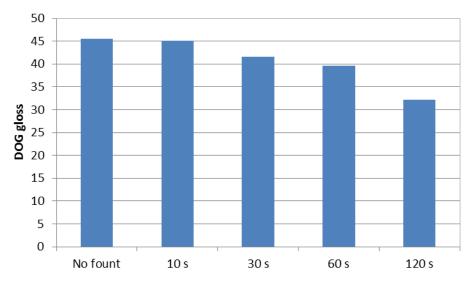


Figure 5. Gloss level (gloss units) of ink films with increasing dosing time in seconds [s] of added fount.

3.3 Surface roughness versus optical properties of the studied prints

Gloss maps and colour coordinates as measured by the DOG and spectral camera, respectively, were correlated with the structural properties, using confocal laser microscopy (CLSM) to define surface topographical roughness. The best correlations found were DOG gloss against CLSM roughness with an R^2 of 0.83. Tracing the gloss values in Figure 5 as a function of ink-fount emulsification dosing time, it can be seen that the gloss is as expected inversely proportional to roughness (Figure 6).

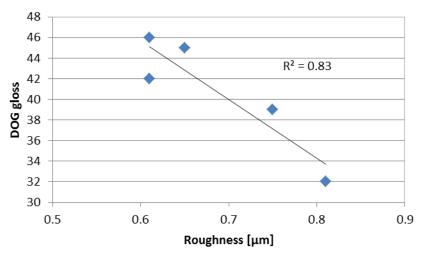


Figure 6. DOG gloss level against ink film roughness $[\mu m]$.

The lightness parameter L^* is calculated separately for each spectrum. The image contrast defined as ΔL^* is modelled to have a linear correlation to the lightness differences observable by the human eye i.e. the more intense the image on a given substrate, the greater is the effective print density. The lightness parameter (L^*) of the ink films is presented in Figure 7. It can be seen that the lightness level of the ink emulsion films varies with emulsification level. It reaches a minimum at an emulsification level of dosing at constant rate equivalent to 4 % on original ink (ignoring evaporation) and increases again with higher fount content.

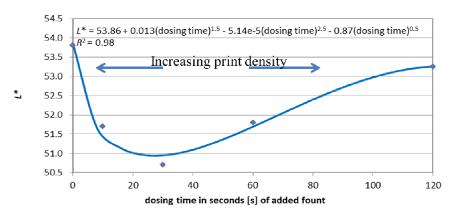


Figure 7. Lightness level of ink films as a function of increasing amount of added fount (ignoring evaporation).

The ink film thickness for the different prints was determined with scanning electron microscopy and a picture of the cross-section as seen by the electron microscope is presented in Figure 8.

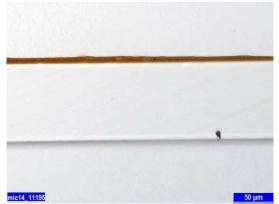


Figure 8: Cross-section of the ink film as viewed in the electron microscope from which the ink thicknesses were derived: an ink layer without fount is presented here.

The correlation between lightness and mean ink film thickness using the fitting equation is shown below in Figure 9 - an exponential (equivalent to a log function). Colour functions are a logarithmic fit of how much "colour" one has in a print. The correlation level for a simple logarithmic fit reaches an R^2 of 0.92.

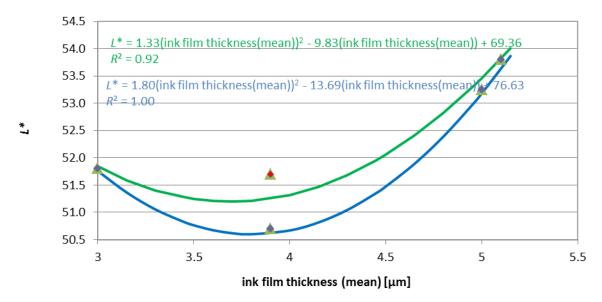


Figure 9. Lightness against ink film thickness.

Perception of gloss from micro-rough surfaces is known to be dependent on the geometry of illumination and observation [Gate and Leaity, 1991; Elton, 2009; Oksman *et al.*, 2008]. It has been previously shown that surface roughness scale, both in terms of amplitude and lateral distribution, affects the observed gloss [Gane *et al.*, 2012]. In the same study it was also shown that the parameters are also dependent on how the gloss and roughness are measured, namely the influence of incident angle.

The results show that the transferred ink amount varies depending on the emulsification degree/composition, and this affects the end print properties. The results in this study indicate the difference between the case where an emulsion is formed at low to medium fount addition, resulting in ever-decreasing ink amount as the volume of ink in the nip reduces as a function of added fountain solution, and the case when phase separation between the ink and fountain solution can occur at the point of emulsion inversion. Since the total volume in the nip under lubrication conditions remains constant, a replacement of ink volume by fountain solution, the emulsion inverts, i.e. ink-in-water rather than water-in-ink, and the ink and aqueous phase under the extensional flow entering the nip can separate. The nip entrance then acts effectively to eliminate the fountain solution from the ink, such that the ink volume in the nip returns almost to the level when no fountain solution was added. Under this condition of phase separation, the quality of the print returns to the high level seen at the zero fount level. The mechanism is shown schematically in Figure 10.

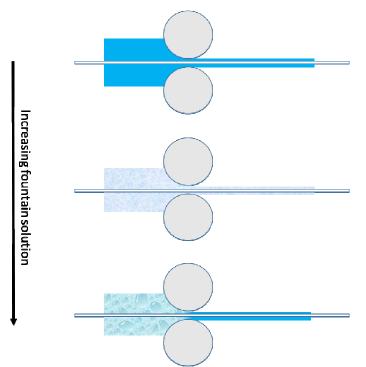


Figure 10. Schematic representation of the change in delivered ink volume as a function of the emulsion component ratio. In the case of excess fountain solution, the emulsion inversion results in expulsion of fountain solution at the entrance to the nip, such that the ink transfer returns to that of close to zero fount addition.

The results show that the optical properties of the ink film changes with the emulsion composition resulting in changes in film formation. If a high fount level is reached within the ink, it causes phase separation, the transfer in the nip creating a weak boundary split, squeezing fount out and transferring ink "only" [Tåg, *et al.*, 2012]. With this high loading, an unstable emulsion is created, resulting in print defects, such as ink repellence.

4. Conclusions

The study shows the role of emulsification degree on the optical properties of prints. With the novel measurement procedure described in this work, we were able to transfer an emulsified ink to a plastic film under constant nip conditions.

We may conclude that:

- 1. A novel dynamic contact procedure could be developed to see the influence of ink-fount emulsification level on optical properties of prints when the substrate is passed through the same nip as is used for the emulsion production. The print surface was analysed using optical measurement tools.
- 2. The optical properties of the prints can be optimised when running with optimal emulsification degree, thus being able to improve the perception quality of products.
- 3. The measurement could provide a relatively easy way to follow the dynamics at different press stations, which could be used as a monitoring tool to predict and control the offset ink-fountain solution process properties.
- 4. The results suggest that a correction factor for evaporation is needed to describe the relationships between dosage time, roughness and gloss a conclusion derived from the curved relationship between the studied parameters.

References

Elton, N.J., Optical measurement of microroughness of pigment coatings on rough substrates, Measurement science and technology, 20, 025303, 2009

Gane, P.A.C., Silfsten, P., Tåg, C.-M., Pääkkönen, P., Hiltunen, J., Kuivalainen, K., Oksman, A. and Peiponen, K.-E., *Isolating contributions to gloss from mechanical and optical roughness, thin layer refractive index and wavelength filtering as a function of illumination and geometry of incidence*, JPMTR, 2, 81-95, 2012.

Gate, L.F. and Leaity, K., *New aspects on the gloss of coated paper*, TAPPI Coating Conference Proceedings, 473-478, 1991.

Juuti, M. Kalima, V., Pakkanen, T. T. and Peiponen, K.-E., A novel method to measure and analyse delta gloss by diffractive glossmeter, Meac. Sci. Technol., 18, L5-L8, 2007

Juuti, M., Koivula, H., Toivakka, M. and Peiponen, K.-E., A diffractive gloss meter for local gloss measurements of papers and prints, Tappi Journal, 27-32, 4, 2008.

Myller K., Peiponen K-E. and Silvennoinen R., *Two-dimensional map of gloss of plastics measured by diffractive-element-based glossmeter*, Opt. Eng. 42, 11, 3194-3197, 2003.

Niskanen, I., Räty, J. and K.-E., Peiponen, *Complex refractive index of turbid liquids*, Optics letters, 32, 7, 2007. Oksman, A., Juuti, M. and Peiponen, K.-E., *Statistical parameters and analysis of local contrast gloss*, Opt Express, 16, 16, 12415-12422, 2008.

Tåg, C.-M., Toiviainen, M., Juuti, M. and Gane, P.A.C., Online Detection of Moisture in Heatset Printing: liquid transfer to printed and non-image areas, Nordic Pulp and Paper Research Journal, 27, 1, 112-121, 2012

A Memory Effect in Sheet-Fed-Offset Printing

Wolfgang Fuchs, Michael Dauer, Ulrich Hirn, Wolfgang Bauer

Graz University of Technology, Institute for Paper, Pulp and Fibre Technology, Austria

E-mail: wolfgang.fuchs@tugraz.at, ulrich.him@tugraz.at

Short Abstract

We are presenting a method to quantify the similarity of local mottling patterns in consecutively printed copies in sheet fed offset printing. It employs image registration to extract precisely defined image regions in a series of printed copies. The similarity of these image regions is calculated using point wise correlation. By analysing the similarity in the mottling patterns in printing runs over several thousand copies we demonstrate that there are mottling patterns that are time stable and in fixed positions of the print. We call this repeated occurrence of mottling patterns over hundreds of printed sheets a 'memory effect' in offset printing. Between 20% and 50% of the print mottle in two print trials consisted of such location and time stable patterns. It can be speculated that this time stable mottle is related to printing press parameters (e.g. rubber blanket, printing form....), whereas the remaining, non-stationary mottle is related to local variations of the printed paper or stochastic variations in the printing process.

Keywords: print mottle, image analysis, pattern, offset printing

1. Introduction and Background

Mottling is defined as an undesired unevenness in perceived print colour, print density or print gloss. An even printed image is assumed to be generated due to a perfect interaction of the three main components involved during the printing process, namely the printing press, the printing ink and the substrate (i.e. paper). Print mottle is the result of an imperfect interaction between any of the three components which leads to uneven transmission or absorption of the printing ink and thus to print mottle.

Depending on the phenomena causing the imperfect interaction, there are three common types of mottle back-trap mottle, water interference mottle and ink-trap mottle (Sadovnikov *et al.*, 2008). Besides this definition, Fahlcrantz divided print mottle into two components: a systematic and a stochastic component (Fahlcrantz, 2005). The stochastic component of print mottle describes the randomly distributed noise in the final print. This random pattern is often accompanied by a systematic component. The systematic mottle component is perceived as a structured pattern, it can be caused by vibrations of the printing machine (Krzyzkowski and Pyryev, 2011) or by periodic structural patterns in the paper like wire marks (MacGregor and Conners, 1987).

In this study we are investigating a new type of mottle. We analyse the time stability of stochastic mottling patterns which occur repeatedly over a longer series of printed sheets. We call this a 'memory effect' in print mottle, because some mottle structures seem to be remembered over time. As an example please refer to Figure 4 where two consecutively printed sheets show identical mottle features *in the exact same location* of the printed colour field. The key point is, that a time stable mottle pattern at fixed positions occurring in consecutively printed sheets indicates that the printing press is also involved in the development of the print mottle. It can be speculated that this type of mottle might result from some local modifications of the rubber blanket that systematically changes the ink transfer in certain positions. Although a stochastic variation during the printing process cannot be ruled out, such as random ink-surface adhesion failure (Alm *et al.*, 2015), we do not consider this in the current discussion. Our work also does not focus on finding the reasons for such location and time stable mottle patterns but introduces a method capable to capture and quantify this 'memory effect', i.e. the similarity between the mottling patterns in consecutively printed sheets.

2. Materials and Methods

2.1 Paper Samples

The examined paper samples were commercial glossy wood free coated (WFC) paper grades. The samples are divided into three groups: a standard WFC paper (135 g/m², coat weight per side 31 g/m²) and two reference WFC samples (115 g/m², coat weight each side 24 g/m²). The first reference sample was referred to be of good print quality (WFC+) and the second reference sample was referred to be of poor print quality (WFC-).

2.2 Printing Machine and Printing Sequence

Two print trials were performed within this study. All paper samples were printed on a 6 colour *Heidelberg SM XL 8* sheet fed offset press with a printing speed of 8 000 sheets/h. The printing plates used were AGFA Amigo. The ink used for the print trial was a NOVAVIT[®] Supreme Bio and the rubber blanket was a Continental[®] SP Evolution. The colour sequence was key black (K), cyan (C), magenta (M), yellow (Y), pantone blue (P) and last black (B).

2.2.1 Print trial A

In this trial only one type of WFC paper (i.e. the standard WFC paper) was printed. First 1 000 sheets of standard WFC paper (135 g/m²) were printed with 6 colours. The printing machine was stopped and the last printing unit (B) was lifted off. Afterwards 100 sheets were printed with 5 colours. The same procedure was performed for the remaining printing units. As a result, a stack of 1 500 sheets consisting of 100 sheets (K), 100 (K+C), 100 (K+C+M), 100 (K+C+M+Y), 100 (K+C+M+Y+P) and 1 000 with all six colours was obtained. Figure 1 shows the obtained stack and the numbers underneath the colour represent the amount of back-traps of one colour.

After printing the paper sheets were scanned and the mottle patterns were analysed. From the first 1000 sheets (printed with all 6 colours, i.e. the bottom part of the stack in Figure 1) we selected, 3 sheets every 50^{th} sheet (1,2,3,51,52,53,101,102,103 etc...). From the sheets printed with 1 to 5 colours (Figure 1, upper part of the stack) we selected 3 sheets every 15^{th} sheet (1,2,3,15,16,17,30,31,32, etc...). From each selected sheet five different colour fields were scanned: 40% tone value C, 100% C, 80% K, 80% B and the mixed colour field 100% C/60% M. We used a flatbed scanner (*Epson Perfection 4990*[®]) at 1 200 dpi (i.e. 21.17 µm/pixel) resolution.



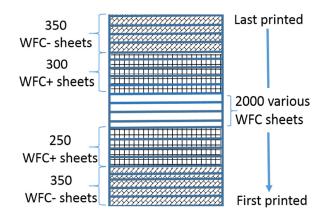


Figure 1. **Print trial A.** The stack of printed sheets obtained from the trial. The numbers below the colours represent the amount of back-traps of one colour.

Figure 2. The stack of sheets obtained from print trial B.

2.2.2 Print trial B

Here one WFC paper with good print quality (WFC+) and one paper with poor print quality (WFC-) was printed on the same printing machine (*Heidelberg SM XL 8*). These papers were printed with 6 colours at 8 000 sheets/h. First 350 sheets of WFC- and then 250 WFC+ sheets were printed. Then 2 000 sheets of various other WFC papers (not examined) were printed. Finally, again 300 WFC+ sheets and 350 WFC- sheets were printed (see Figure 2).

After printing the paper sheets were scanned and the mottle patterns were analysed. From each printed WFC- and WFC+ stack the 10 first printed sheets and 10 last printed sheets were selected (e.g. for the first printed WFC- stack: 1,2, ... 9,10,341,342, ... 349,350 and for the first printed WFC+ stack: 1,2, ... 9,10,241,242, ... 249,250). From each selected sheet the same colour fields were scanned and examined as for print trial A.

2.3. Image Analysis

An example of the print form containing all colour fields used for this study is shown in Figure 3(a). Figure 3(b) depicts an 80% K colour field which shows the size of the finally examined area ($50x50 \text{ mm}^2$; red square).



(a) Test print form.

(b) Colour field 80% K from the print test form with extracted image region.

Figure 3. (a) The test print form. (b) From each sheet a defined part from a colour field is extracted, here the 80% K colour field.

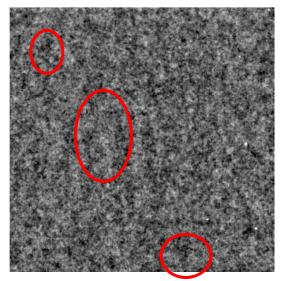
For image analysis, first the exact same region of a scanned colour field is extracted from consecutive sheets using image registration. Image registration is a procedure where exactly the same region in different images is extracted and brought to congruence. The images are registered by applying a shape preserving coordinate transform where the edge points of the colour fields were used as registration marks (Hirn *et al.*, 2008).

After registration, a set of images showing exactly the same regions in the colour fields for all printed sheets is available. Figure 3(b) shows one image of a registered 80% K colour field. From these images, a sub-image is extracted from every sheet resulting in a set of sub-images, which is then used for analysis. The final size of the analysed sub-images is 50x50 mm² (i.e. the size of the square in Figure 3(b)).

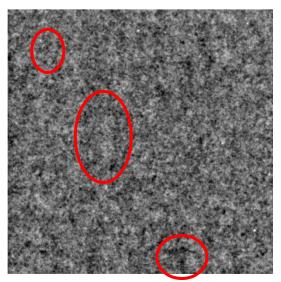
After extraction, the sub-images of all samples were descreened and then post processed in two ways. On the one hand, spectral filtering (i.e. a FFT pass band filter) was performed by a pass band filter in the wavelength band of 1-16 mm. The reason for spectral filtering is that the human eye visually perceives non-uniformities differently at different length scales. The region of interest for visual perception of print mottle according to Johansson is located in the wavelength band of 1-16 mm (Johansson, 1993). This type of filtering produces images where only the structures most relevant for print mottle are preserved.

On the other hand, the images were rescaled to a pixel size of 250 μ m. That is the size of structures, which the human eye can resolve under good illumination at a viewing distance of 30 cm (Olzak and Thomas, 1986). Hence, at a pixel size of 250 μ m *all* structures perceived by the human eye are preserved.

The final result of scanning, filtering and rescaling can be seen in Figure 4. It shows two 80% K fields from consecutively printed sheets which were spectral filtered in the relevant region for print mottle, the images were extracted from the print as indicated in Figure 3(b).



(a) Extracted image region from sheet #1000



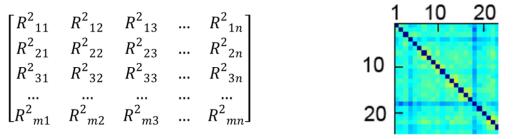
(b) Extracted image region from sheet #1001

Figure 4. Example of two contrast enhanced images extracted from two 80% K regions of consecutively printed sheets (print test A). The images were spectral filtered (wavelength band 1-16 mm). Some mottling features occurring in exactly the same location in both prints are highlighted by ellipses (size of images: 50x50 mm²).

In order to evaluate the location and time stability of the mottle pattern we quantify the similarity between the extracted and spectral filtered images. We apply point wise correlation of the registered images (Hirn *et al.*, 2008) where the coefficient of determination (R^2) is the resulting measure of similarity between the images. The coefficient of determination explains how much of the variance in one data set is explained by another (or several other) data set(s) (Neter *et al.*, 1996). A value of $R^2 = 1$ between two images indicates that the mottle pattern in these prints is exactly the same, a value of $R^2 = 0$ indicates that there is no similarity at all. Analysis of R^2 was performed for both types of images, i.e. the pass band filtered and the rescaled images.

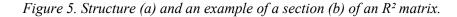
The coefficients of determination between the images are displayed in R^2 matrices for each colour field. The structure of the matrix is shown in Figure 5(a). The indices are in accordance with the order of the selected stack of printed sheets from bottom (first printed) to top (last printed). For example in print trial A, indices 1 to 3 represent the images of the first three printed sheets in the stack. The consecutive three indices (i.e. 4-6) represent the 51st to 53rd printed sheet, because 50 sheets are

skipped in between (see description of print trial A. As a consequence, the last three indices (i.e. n-2, n-1 and n) represent the last three printed sheets (i.e. the top of the stack in Figure 1). The square matrix (m = n) is symmetric since R^2_{12} and R^2_{21} are identical because the similarity is equal in both directions. An example of a section of an R^2 matrix is displayed in Figure 5(b). The main diagonal represents R^2 values between one and the same image (e.g. R^2_{11}). As this is the similarity between one and the same image, R^2 is 1 for the main diagonal.



(a) Structure of the R^2 matrix

(b) Example of the upper left part of an R^2 matrix.



For print trial B the structure of the R^2 matrix is similar. Since sampling was a little different compared to print trial A, the first 10 indices (n = 1-10) represent the images of the first 10 printed sheets of WFC- (i.e. at the bottom of the whole stack in Figure 2, sheet 1-10) and the next 10 indices (n = 11-20) the images of the 10 last printed sheets of WFC- (i.e. at the top of the bottom WFC- stack in Figure 2, sheet 341-350). This allocation applies also for the other WFC+ and WFC- samples of the stack obtained from print trial B.

The variance (i.e. squared standard deviation) of each image was also calculated and depicted on top of the R^2 matrices (Figure 6 to Figure 10) and is a measure for the print unevenness in the image. A lower variance indicates lower print unevenness, whereas a higher variance indicates a higher print unevenness. Thus, not only the similarity between the print mottle patterns in the images can be quantified, but also the development of print unevenness over time is captured.

3. Results and Discussion

3.1 Print trial A

In this print trial the printing units were lifted off one after the other. In Figure 6(a) and (b) the R^2 matrices for the colour field 80% K are depicted. Figure 6(a) shows the matrix where all images were spectrally filtered within the wavelength band 1-16 mm, whereas the images in (b) were rescaled to a pixel size of 250 µm. Above the R^2 matrices the variance of each image is plotted. Below the matrix, the printing units which were active are shown. The sequence is the same as the order in Figure 1 (bottom to top implies first printed to the last printed). Thus, KCMYPB means that all printing units were active and K means only the K unit was active.

Figure 6 reveals that the print mottle of one sheet is related to the print mottle of other sheets. It shows further that the print mottle is persistent for a rather long period of printed sheets. The correlation drops after a printing unit has been lifted, especially after lifting B and Y. However, when restarting printing after lifting off, the similarity of the print mottle again rises, which means that after a brief interruption due to the lifting of a printing unit some part of the earlier mottling structure re-appears in the print. We termed this systematic re-occurrence of mottling patterns over a large amount of printed sheets 'memory effect'.

The correlation between two images is higher when they are located close to each other, i.e. when they have been printed shortly one after another. In the R^2 matrices, R^2 values of images printed close to each other in time are located in the plot region surrounding the main diagonal. The highest

correlations were found in the R^2 matrix with the rescaled images (Figure 6(b)). There, R^2 is close to 0.50. R^2 is lower in the region of interest for print mottle (1-16 mm wavelength band, Figure 6(a)). However, even after 1 000 sheets the R^2 between the print mottle of the first and the last sheet is still close to 0.35. It seems, that there is an inherent print mottle which stabilises after restarting and is continuously re-occurring.

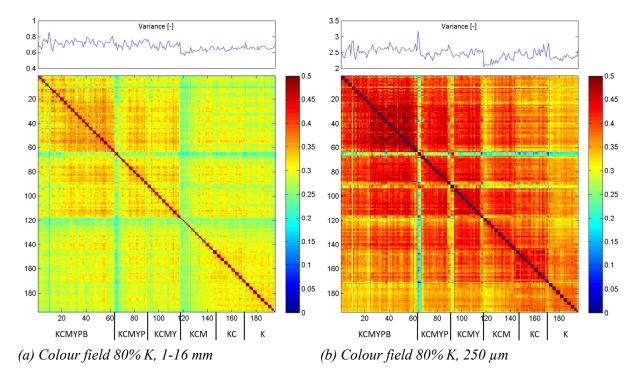


Figure 6. R² matrices of colour field 80% K. In (a) images were spectrally filtered (wavelength band 1-16 mm) and in (b) the images were rescaled to a pixel size of 250 µm.

Furthermore, Figure 6 shows that the variance (i.e. print unevenness) decreases after lifting of the printing units, especially after lifting B and Y. This agrees with the common notion that print unevenness decreases with decreasing number of back-traps.

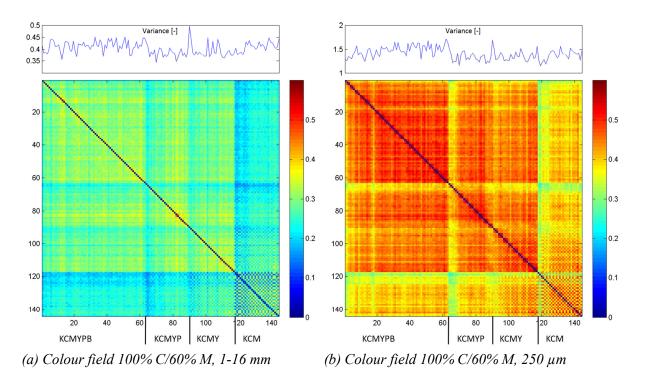


Figure 7. R² matrices of colour field 100%C/60% M. In (a) images were spectral filtered (wavelength band 1-16 mm) and in (b) the images were rescaled to a pixel size of 250 µm.

Figure 7 shows similar results for the mixed colour field 100% C/60% M. R^2 is stable for a rather long period of time. It is up to 0.55 when looking at the rescaled images (Figure 7(b)). For the regions of interest for print mottle (Figure 7(a), 1-16 mm wavelength band), R^2 is up to 0.40. The correlation decreases after lifting the B, P and Y printing units and increases again with further printing. When printing KCM, an alternating pattern seems to occur which is shown by the alternating R^2 in the KCM part of the matrix in Figure 7(a) and (b). This means that the similar print mottle does not occur on every consecutive sheet, but on every second or third sheet, suggesting that the phenomenon is either unstable or related to a patterning on the original paper tambour.

The print unevenness decreases when lifting B, but increases when lifting Y. Usually print unevenness is expected to decrease with decreasing amounts of back-traps, but in Figure 7(a) the variance has the same level when comparing KCM and KCMYPB which is assumed to be related to the alternating R^2 in the KCM part of the matrix.

The results for 40% C and 80% B reveal findings similar to the 80% K and 100% C/60% M fields. In contrast, however, the R^2 matrices of the full tone 100% C present different results (see Figure 8(a) and (b)). There is little to no correlation (only up to 0.15) between the images. Furthermore, the low correlation is only stable for about 3 printed sheets in a row. This suggests, that the time stable and re-occurring print mottle is not a problem for full tone printing. It is yet unclear why this is the case.

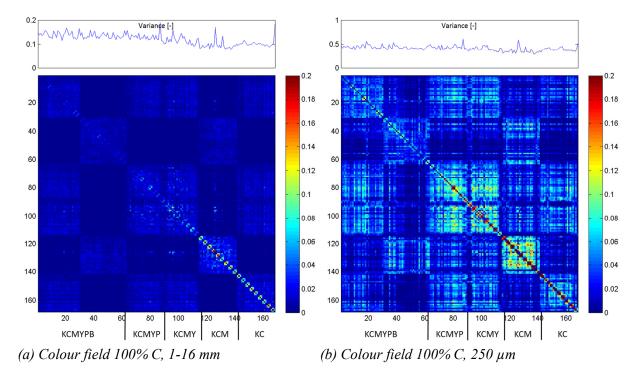


Figure 8. R² matrices of colour field 100%C. In (a) images were spectral filtered (wavelength band 1-16 mm) and in (b) the images were rescaled to a pixel size of 250 µm.

3.2 Print Trial B

In print trial B stacks of WFC+ and WFC- samples were distributed in an entire stack (compare Figure 2). Below the matrices in Figure 9(a) and (b) the order of the distributed samples in the printed stack is described. Firstly the R^2 of the 10 first and 10 last printed WFC- samples, immediately afterwards R^2 of the WFC+ samples and secondly the R^2 of the WFC+ and WFC- samples again. As shown in Figure 2, there were approximately 2 000 sheets of various WFC grades (Figure 9(a) and (b) between number 40 and 41) between the two WFC+ stacks.

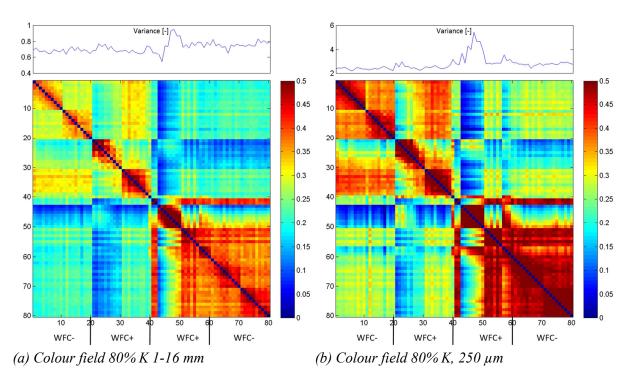


Figure 9. R² matrices of colour field 80%K. In (a) images were spectral filtered (wavelength band 1-16 mm) and in (b) the images were rescaled to a pixel size of 250 µm.

The print mottle pattern for the first 10 WFC- sheets (see Figure 9(a) and (b), numbers 1-10) show values of an R^2 up to 0.45 and similarly for the last 10 sheets printed in the first stack (see Figure 9(a) and (b), numbers 11-20). Since there were 350 WFC- sheets printed, this tells us that R^2 decreases from the beginning to the end of the stack which can be seen in the R^2 matrix where the images of numbers 11-20 are correlated to numbers 1-10. These findings are similar to those of the second stack (WFC+). Within 10 sheets the correlation is higher, and subsequently decreasing when reaching the end of the stack. However, there is also a correlation between the print mottle observed in the WFC- and WFC+ images (up to 0.37).

At the beginning of the second WFC+ stack there seems to appear a print defect, but disappearing afterwards. The 3^{rd} to 10^{th} image (in the second WFC+ stack) are highly correlated to each other, but not to the rest of the stack. It can also be seen in the variance of the images. The variance is higher as compared to the rest of the images. After disappearance of the print defect, the R^2 between the 10 last images of the WFC+ stack and the images of the WFC- stack is very stable. There is even a correlation between the sheets printed at the beginning and after 3 000 sheets which is shown by the upper right part of the R^2 matrices (Figure 9(a): 0.20, and Figure 9(b): 0.25). R^2 is higher for the rescaled filtered images (Figure 9(b), 250 µm) and is a little lower in the relevant wavelength for print mottle (Figure 9(a), wavelength band 1-16 mm).

Figure 10 shows the R^2 matrices for 100% C/60% M. For the first stack of WFC- and WFC+ the correlation is higher for the images of the sheets printed close to each other in time. R^2 is lower for the images which are printed with hundreds of sheets lying in between, but is stable. These R^2 matrices prove that another print defect shows up in the second WFC+ stack. The variance in Figure 10(a) and (b) is higher where the print defect occurs. However, the print defect disappears, because the images printed afterwards correlate with the first printed ones again. It is apparent from Figure 10 that R^2 is higher than 0.50 for the rescaled images (Figure 10(b), 250 µm) and up to 0.50 for the print mottle wavelength band (Figure 10(a), wavelength band 1-16 mm).

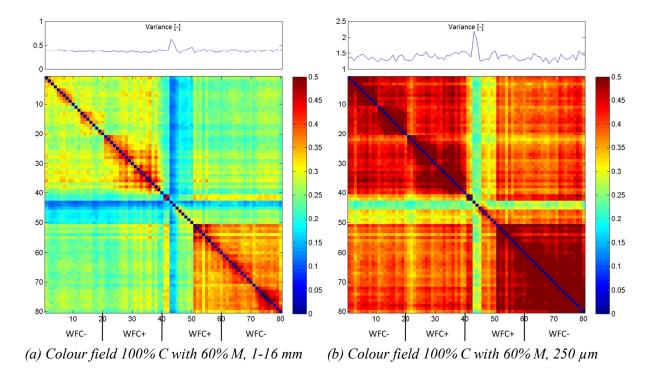


Figure 10. R^2 matrices of colour field 100%C with 60% M. In (a) images were spectral filtered (wavelength band 1-16 mm) and in (b) the images were rescaled to a pixel size of 250 μ m.

The results for the other colour fields (80% B and 40% C) are similar to those presented above. However, like in the previous print test A, the 100% C field shows little to no correlation (compare Figure 8). These findings suggest that the print mottle of these images is not only time stable and re-occurring within one paper grade, but is also stable over various paper grades. It is yet unclear where this time stable and re-occurring effect ('memory effect') is generated.

4. Conclusions

We have introduced a method that quantifies the degree of similarity between the mottling patterns in consecutively printed sheets. The similarity between the prints was measured using point wise correlations, it was collected and displayed in R^2 matrices. The similarity of the print mottle patterns in five different colour fields was investigated. The images were examined in the relevant region for print mottle (i.e. wavelength band 1-16 mm) and in the region where all structures are perceived by the human eye (>250 μ m).

The results show that there exists a re-occurring print mottle which can be found over a large number of printed sheets. We observed this print mottle memory effect over more than 3 000 copies, even when different paper grades were printed. In one printing trial the R^2 for a 100% C/60% M colour field was up to $R^2 = 0.50$ in the wavelength band which corresponds best with the human perception of print mottle. That means that up to 50% of the mottling structure visible in this colour field is a re-occurring mottling pattern. In another trial, the correlations between the different images in the 100% C/60% M colour field was considerably lower with $R^2 = 0.30$.

Our method detects re-occurring mottle patterns appearing at fixed positions, thus it can be speculated that this mottle is related to modifications of the rubber blanket in the printing press. The remaining mottle should be related either to variations in local paper properties that lead to local variations in ink transfer and light absorption or assigned to stochastic interactions in the process.

Acknowledgments

The authors gratefully acknowledge financial support of the Austrian Research Promotion Agency (FFG). Furthermore, the authors thank Sappi Austria Productions GmbH for their help to run the print trials.

References

Alm, H., Ström, G., Schoelkopf, J., and Gane, P. (2015). Ink-lift-off during offset printing: A novel mechanism behind ink-paper coating adhesion failure. *Journal of Adhesion Science and Technology*, 29(5): pp. 370–391.

Fahlcrantz, C. (2005). On the Evaluation of Print Mottle. PhD thesis, KTH Stockholm.

Hirn, U., Lechthaler, M., and Bauer, W. (2008). Registration and point wise correlation of local paper properties. *Nordic Pulp and Paper Research Journal*, 23(4): pp. 374–381.

Johansson, P. (1993). Print mottle evaluation by band-pass image analysis. In *Advances in Printing Science and Technology*, IARIGAI, pp. 403.

Krzyzkowski, J. and Pyryev, Y. (2011). Analysis of vibration of printing unit of offset printing press. *Challenges of Modern Technology*, 2(2): pp. 31–34.

MacGregor, M. A. and Conners, T. E. (1987). Image analysis of an LWC paper reveals wire mark in the print density variations. *Tappi Journal*, 70(9): pp. 95–99.

Neter, J., Kutner, M., Nachtsheim, C., and Wasserman, W. (1996). *Applied Linear Statistical Models*. Irwin, Chicago, 4th edition.

Olzak, L. A. and Thomas, J. (1986). Seeing Spatial Patterns. John Wiley & Sons.

Sadovnikov, A., Lensu, L., and Kälviäinen, H. (2008). On estimation of perceived mottling prior to printing. *Proceedings of SPIE - The International Society for Optical Engineering*, Vol. 6808.

A new Concept to simulate commercial Print Trials on Lab Scale Part 1: introduction of a lab printing device

Dietmar Wolf¹, Dirk Kisters¹, Patrick Gane^{1,2}

¹Omya International AG, CH-4665 Oftringen, Switzerland

²School of Chemical Technology, Department of Forest Products Technology, Aalto University, FI-00076 Aalto; Helsinki, Finland

E-mail: dietmar.wolf@omya.com; dirk.kisters@omya.com, patrick.gane@omya.com

Short Abstract

The print quality on a wide range of substrates, e.g. graphical papers, board, packaging etc., is one of the most important parameters to guarantee the high quality appearance of goods in the market and consumer acceptance. The realistic evaluation of the print performance during the development / optimization of formulations is a key factor in accomplishing this target of a high quality end product. Although a variety of laboratory tests have been carefully developed in the past to simulate commercial printing, including rotogravure, flexography, offset, and more recently digital imaging, it has to be concluded that all these methods are either lacking in their overall applicability or are so strongly limited toward troubleshooting when a given well-known print problem is observed in practice, that they are unsuitable for print quality prediction purposes, rendering the ever-present need for expensive commercial print trials. For this reason, the construction of a reel-to-reel laboratory printing press platform has been undertaken at the Omya International AG facilities, and is coming on-line for practitioners, developers and the like, providing the organization and evaluation of commercially realistic print trials to support all kinds of development activities. The application is also extended to converting properties (partly) and runnability. The setup of the machine is presented in this report, and an example correlation to commercial print trial results in flexographic and rotogravure printing will be shown and discussed.

Keywords: Flexographic Printing, Rotogravure, Lab Methods, Correlation Commercial / Lab Printing,

1. Introduction and background

The print quality of all kinds of substrates is one of the most important parameters when targeting a high quality appearance of printed products in the market and resulting consumer acceptance. The evaluation of the print quality during the development / optimization of formulations is a key factor in accomplishing a high quality end product across markets as diverse as graphical papers, board, packaging etc. Although a variety of lab tests exist, they are designed primarily to provide troubleshooting tools in cases where print problems have become manifest in commercial applications. A limited number of examples of such tests can be found in (Klein, 2009; Nierhoff, 2009; Hasse, 2009; Jutila et al., 2012), and methods such as Prüfbau and Deltack (Dr.-Ing. H. Dürner GmbH), Labra-Test (Norbert Schläfli Maschinen, Switzerland), Helio-Test (IGT Testing Systems, The Netherlands) and ISIT (SeGan Ltd., UK), to mention just a few, are exceedingly useful tools for lab studies. The existing laboratory methods are valuable in studying basic properties of ink-substrate interactions, such as dry picking properties, tack build and the like, and may lead to important information on print controlling properties, but a prediction of results from commercial print trials regarding overall print quality, colour gamut, converting and runnability properties is limited and difficult to achieve a priori. Therefore, formulating a correlation from laboratory to commercial print trial proves difficult with such specific tools, and when the task in hand is to predict overall print quality, the general conclusion is that they are on the whole unsuitable with regard to the predictability of commercial print trials.

The simulation of typical commercial print phenomena requires the inclusion of:

- reel-to-reel printing dynamics
- simulation at realistic speeds under equilibrium conditions
- reproducing drying processes on commercial printing machines
- use of commercially available printing plates
- suitability for a broad range of substrates (e.g. packaging materials)
- application of commercial inks
- evaluating runnability and converting performance parameters, which have become key factors -
- providing dimensionally equivalent electrostatic print assist (ESA) for rotogravure evaluation

amongst others.

Due also to the fact that commercial print trials are expensive, time-consuming and very inflexible in terms of experimental design, the Forest Products Technology Laboratory at Omya International AG began in 2011 to evaluate the possibility of constructing a reel-to-reel lab printing press to simulate commercial print trials with regard to print quality, converting properties (partly) and runnability, and this development process has subsequently been exemplified in the work of Ridgway *et al.* (2013), in which a technique was developed to reproduce tail edge pick in offset using a modification of the rotational function of the ink-surface interaction testing device, ISIT.

In the current paper the setup of an in-line print unit machine will be presented, which brings the initial concept designs into practice, and an example correlation to commercial print trial results in flexographic printing will be shown and discussed. The equipment will be available for use to developers, customers etc. for the evaluation of commercially realistic print trials to support a wide range of development activities.

2. Demands on the system

During the development of a suitable lab printing press the following requirements had to be met:

- flexible setup for different printing technologies
- flexible handling of a broad range of substrates to print on
- usage of a wide range of printing plates
- usage of an extensive range of commercial inks for the different printing technologies (e.g. water-based, solvent-based, UV-curing inks etc.)
- flexible drying setup
- flexible adjustment of machine speed at a reasonable level (to match commercial printing speeds)
- printing width has to match the existing in-house machinery (e.g. lab coating system, lab calendering)

3. Machine Setup

The machine is described here in respect of its general design and then more specifically in respect to the units installed.

3.1 General Design

The basic setup of the press is shown in Figures 1 and 2. The working principle of the machine is reelto-reel. Therefore, an unwind/rewind station is implemented. Both the speed and the web tension can be adjusted by means of a remote control panel. The maximum speed of the machine is 100 m.min⁻¹ at a nominal printing width of 150 mm. Due to the adjustable web tension, a variety of different substrates can be used for printing, e.g. LWC-paper, transparent films, board, liner, corrugated board etc. (Ridgway *et al.*, 2013).

To be close to the market reality a web edge guide control is installed. The flexibility of the machine is preconceived by the provision of stainless steel mounting plates on the horizontal axis (direction of the web). All printing-relevant modules, like printing units, dryers, web guides etc., can be mounted on these plates and therefore can be installed and removed easily. Due to this preconceived flexibility along the web horizontal axis, it is easy to adjust the distance between the printing units and/or the dryers, and in combination with the adjustable speed of the machine, it is therefore possible to simulate the conditions in commercial printing presses in respect to such parameters as distance between print units etc. With the current setup, two in-line printing units, two hot-air dryers and two web guides can be installed. For the simulation of runnability parameters, such as smearing, a system of flexible deflection rollers can be suitably mounted.

The hot-air dryer works at adjustable temperatures up to 140°C and is sufficient for all expected applications.

A system for the adjustment of the side and the circumference register, indispensable parameters to achieve reasonable two-colour printouts, is installed.

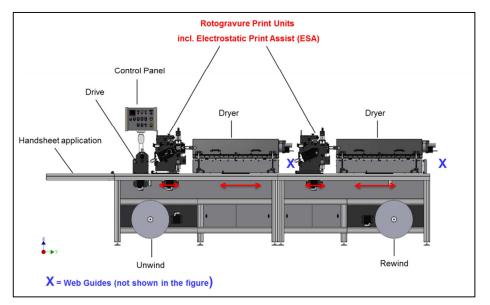
In addition to the reel-to-reel setup, a simple manual feed is also integrated to enable printing on sheets.



Figure 1: testacolor tfm 157-2 lab printing press (Norbert Schläfli Maschinen, Switzerland)

3.2 Rotogravure

Although rotogravure printing is a decreasing market for graphical papers, and is becoming at least partly replaced by offset, the demand for rotogravure in the packaging segment is still increasing. Furthermore, the price pressure in the graphical paper segment leads to a need for cost-optimized papers, which have to be competitive with regard to print quality and runnability. This provides an opportunity for the development of grades containing high amounts of calcium carbonate. Achieving this is not straightforward as traditional calcium carbonate slurry pigments are renowned for their negative impact on rotogravure mottling, especially on application of ESA. The need to have a realistic reel-to-reel ESA controlled rotogravure printing platform is therefore essential.



The rotogravure setup, which is integrated in the lab press, is shown schematically in Figure 2.

Figure 2: schematic rotogravure setup on the lab printing press

The design consists of two rotogravure printing units with both inter unit and post printing drying stations (hot air). The printing process is, therefore, a two colour wet-on-dry process. Two web guides (not shown in Figure 2) are installed after each dryer to ensure an accurate control of the register during printing. The printing cylinders are commercially available engraved copper, chromium-plated cylinders. The gravure parameters represent standards from a German print shop as applied to highly mineral filled supercalendered (SC) papers, and are given in Table 1. The width of each cylinder is 150 mm, the circumference is 314.32 mm. Figure 3 shows the geometry of the ink cells. The standard ink applied is toluene-based (Siegwerk Druckfarben AG & Co. KGaA).

Gravure parameters	Magenta	Cyan
screen / lines cm ⁻¹	70	70
angle / °	0	2
cross diagonal / µm	150	100
cell walls / µm	30	33
cell depth / µm	20	15

	Table 1:	gravure	cylinder	parameters
--	----------	---------	----------	------------

An important and uniquely designed feature of the rotogravure system is the integrated electrostatic print assist (ESA). The ESA is fully adjustable from 0 - 100 %, and comparable to industrial print assist systems. Due to the fact that in commercial rotogravure presses an ESA is nowadays standard and used to improve the print quality significantly, the integration of the ESA in the lab printing press is seen as a key success factor to ensure a close simulation of commercial rotogravure print trials (Martorana *et al.*, 2006; Gravure Education Foundation & Gravure Association of America, 1991; Ollech, 1993).

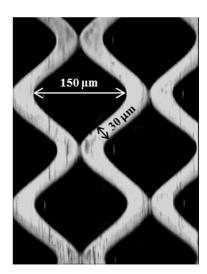


Figure 3: surface image solid tone of magenta cylinder -screening 70/0 lines cm⁻¹

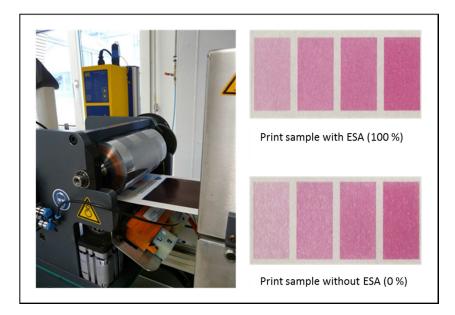


Figure 4: engraved cylinder, ESA unit and resulting test prints

3.4 Flexography

Flexography is the principally used printing method in the packaging sector, e.g. printing of corrugated board (post-print). Very often it is found in use for printing on liner, which will be laminated later by the corrugator (pre-print). The standard flexographic setup for graphical papers, shown previously in Figure 1, is a two colour wet-on-wet process, equipped with two print units and a hot-air dryer at the end of the press only. A detailed drawing of the print units is shown in Figure 5. For printing on board, liner and standard coated liner, the quality-defined process is two-colour wet-on-dry, and for this setup a second hot-air-drying station (see rotogravure setup) is used. However, increasingly for products such as "direct to display shelf" boxes, e.g. wine boxes, a wet-on-wet flexo process is adopted, placing high demands on the substrate absorbency characteristics (Ridgway *et al.*, 2013).

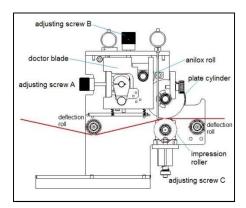


Figure 5: lab flexo printing unit (Norbert Schläfli Maschinen)

A defined ink transfer is achieved by using an anilox roller and a chambered doctor blade, which lies negatively engaged on rotation of the anilox. The negative angle of the doctor blade ensures an optimized filling of the anilox cells and is nowadays standard in the industry. The ink chambers are filled by an ink circulation system using tube pumps (not shown in Figure 5). Due to the fact that printing on different substrates needs specific anilox rollers, the printing units can be equipped as follows:

- screen: 120 lines cm⁻¹, volume: 15.2 cm³m⁻²
- screen: 260 lines cm⁻¹, volume: 4.0 cm³m⁻²
- screen: 340 lines cm⁻¹, volume: 4.8 cm³m⁻²

The screen angle for all mentioned anilox rollers is 60°. Both water and solvent based inks can be used, with standard inks in current use being water-based from Huber (Michael Huber München GmbH). For an optimum ink coverage on different substrates, various printing plates are available. In general, the choice of plate follows the rule: for rough surfaces thick and soft plates, for smooth surfaces thin and hard plates should be applied. For this reason, plate cylinders with undercuts of 1.14 mm and 2.54 mm are used. The plates can be manufactured using the newest screening technologies, such as HD-Flexo, and are delivered by the main suppliers, including Flint, DuPont, Asahi or Kodak.

4. Results and Discussion: example print trials

Two example trials are reported, first a brief illustration of a rotogravure application, and secondly a more detailed discussion of a flexographic trial.

4.1 Example of a rotogravure lab-trial

Table 2 shows a typical evaluation for a rotogravure lab print trial, where different ESA-settings were applied to a range of trial LWC-papers, which contained 100 % CaCO₃ and a varying amount and type of dispersant. All papers were printed with the setup described above and were evaluated by means of the Verity IA software from Prüfbau (mottling), an image analysing system, and the Software Gravure Quality Analyse, a software tool designed for measurement of missing dots, developed by Prof. A. Weichmann, Stuttgart Media University, Germany. This software is in the beta phase of development and will be evaluated at Omya. Each mottle value comprises mottle and grain indexes for cyan half-tone patterns, whereby low values represent good results.

Samples		1 Sat			2 Sat			3 Sat	
ESA Settings / %	0	30	100	0	30	100	0	30	100
Mottling of 50% Cyan	30.6	41.05	35.54	57.21	64.09	135.25	33.27	42.06	68.17
Missing Dots / Ncm ⁻²	4 806	39	62	7 269	54	48	8 813	176	102

Table 2: correlation of ESA performance regarding missing dots and mottling

In the past LWC-grades containing high amounts of traditional CaCO₃ showed high missing dot values when ESA is not used, but unfortunately when ESA is applied to correct this a strong deterioration in large scale mottling appears [11]. From Table 2 it becomes apparent that the installed ESA-module can be used successfully to reduce the number of missing dots significantly. Already at an ESA-level of 30 % the missing dots are reduced to the detectable limit of the applied method/software. The higher the ESA applied, the traditional problem of increased mottling appears. As can be seen, this is most strong in the case of the trial paper 2 Sat, and this coating represents the traditional case. The coating 3 Sat is less sensitive, and the case 1 Sat is virtually immune to mottling and represents a new development in carbonate coating.

4.2 Example of a flexographic print trial

Here we exemplify a flexographic print trial comparing a commercial print with the laboratory printing press results.

4.2.1 Test form and print trial setup

The commercial print trials were carried out at the DFTA in Stuttgart on an F&K Flexpress 6S/8 – an 8 colour central impression machine. As shown in Table 3, most of the print parameters were comparable for both commercial and laboratory printing (same plate material, screening technology, plate thickness, ink etc.). A few parameters (e.g. speed) were given by the setup of the presses and not changeable.

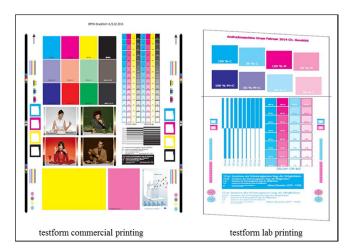


Figure 6: laboratory and commercial test form used for the print trials (flexographic printing)

Application parameter	Commercial printing press	Laboratory printing press
machine	F&K Flexpress 6S/8 (8 colour impression)	testacolor tfm 157-2 lab printing press
operation mode	"wet-on-dry", 40°C intermediate / 60°C end drying	"wet-on-dry", 115°C intermediate and end drying
speed /m.min ⁻¹	250	100
ink	Huber colour series Hydro X	Huber colour series Hydro X
ink viscosity / s	21-22 (viscosity cup 4 mm, DIN53211)	21-22 (viscosity cup 4 mm, DIN53211)
colour application	Y-M-C-B	M-C
cliché	nyloflex [®] ART Digital, 1.14 mm	nyloflex [®] ART Digital, 1.14 mm
tape	Lohmann DuploFLEX [®] FOL 020, 0.2 mm	Lohmann DuploFLEX [®] FOL 020, 0.2 mm
anilox roll	340 lines cm ⁻¹ ; dip volume 4.8 cm ³ m ⁻²	340 lines cm ⁻¹ ; dip volume 4.8 cm ³ m ⁻²
printing plate	Flint group 60 lines cm ⁻¹	Flint group 60 lines cm ⁻¹
print gloss, optical density measurement image	100 % magenta, 100 % cyan	100 % magenta, 100 % cyan
mottle measurement image	100 % cyan	100 % cyan

Table 3: summary of the laboratory and commercial print trial setup (flexographic printing)

4.2.2 Substrates used in flexo trial

A two-ply 140 gm⁻² liner was taken out of the production line before coating. As the base control for the coating trials, the fibre testliner in this uncoated condition was used. The ground ply is made out of ordinary recovered paper grades, the top-ply out of high recovered grades, at times also fresh fibres are used. The top-ply was surface sized with starch and a copolymer. In addition an optical brightening agent was added into the top ply.

For the flexographic printing trials the testliner was coated. The coating trials were done within the scope of a Master's thesis (Goodrick, 2014) on a pilot coater with either a filmpress (2 CaCO₃ containing formulations, coat weight ~4 gm⁻²) or a rod coating (4 CaCO₃ containing formulations, coat weight ~10 gm⁻²). Furthermore, 2 double coated (precoat rod ~9 gm⁻², topcoat blade ~8 gm⁻²) samples were produced for the print trials. All samples were calendared before printing by means of a pilot-supercalender (115 kNm⁻¹, 30°C, 1 nip). The applied coating formulations are representative standard coating formulations for liner products in the market.

4.2.3 Trial evaluation

For the evaluation and correlation of the print trials carried out on the commercial and the lab printing press, the following parameters were measured on the 100 % cyan and magenta fields:

- print gloss Tappi 75° (Lehmann LGDL-05.3-Labor, ISO 8254-1)
- optical print density OD (Techkon, Spectrodens, ISO 5-3/4)
- mottle (Verity I/A from prüfbau Dr.-Ing. H. Dürner GmbH)

From Figures 7-9 it becomes apparent that all three measured parameters show an excellent correlation between the commercial and the laboratory printed samples.

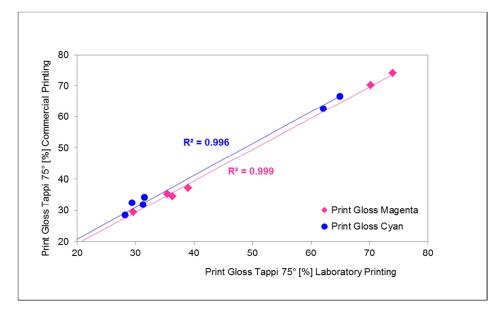


Figure 7: linear correlation print gloss commercial and laboratory flexo printing

The print gloss values for both cyan and magenta show a very good correlation. It is important to emphasize that the print gloss shows a direct comparability without any offset between the commercial and the laboratory press. Therefore, a prediction of resulting print gloss based on lab printing trials should be reliably reproduced.

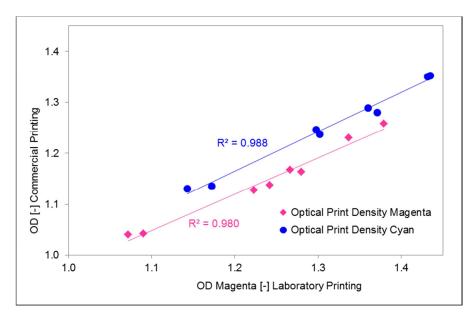


Figure 8: linear correlation between optical print density from commercial and laboratory flexo printing

The measured values for the optical print densities show also a good correlation for each colour. It becomes apparent that, in contrast to the print gloss, there is an observable offset divergence between the colours from the commercial and the laboratory print trials. The lab prints show in general a higher optical density for magenta than for cyan, and for both inks higher than the commercial prints. Since

the instrumentation used in the optical density measurement is identical, there must be a press-related or ink difference. A possible explanation for this phenomenon is the printing speed - a higher printing speed leads to a shorter contact time between the plate and the substrate and results in slightly reduced ink transfer in the commercial printing process (Johnson, 2008). It is also known that the compressionrelaxation behaviour of the printing plates has an influence on the ink transfer, and therefore on the resulting optical densities, but this effect is difficult to study in isolation. Furthermore, it is recognised that the anilox rollers, even if they have nominally the same parameters, do not necessarily have the same ink-release behaviour. A variation between the different production lots cannot be excluded. Ascertaining the true cause for the offset in optical density will necessarily be part of future work.

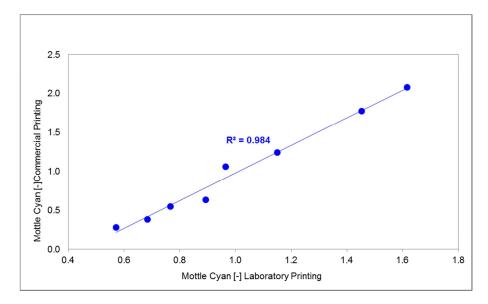


Figure 9: linear correlation between mottle from commercial and laboratory flexo printing (exemplified for cyan)

Also for the mottle (Figure 9) a good general correlation was found, although skewing function in respect to absolute values is observed. The commercial printouts show in general a slightly greater range in mottle value, as measured by the software, i.e. at low mottle values, the commercial test shows slightly lower values, whereas at higher mottle the levels it shows slightly higher values. A reasonable explanation is difficult to provide, but the dynamics of the printing plates under different speeds could once again be the root cause. Also this phenomenon will be part of additional investigations in the future.

4.3 Impact of different flexo printing plate materials on ink trapping

Besides the usage of the lab printing machine as a tool to simulate commercial print trials it is also possible to simulate different experimental setups on this machine. As an example, we studied the effect of different types of flexo printing plates on the ink trapping – a very important and decisive quality parameter in commercial flexographic printing.

The trial was conducted with two different plates from the Flint Group – ACE 1, 70 mm (hard plate) and ART 2.54 mm (soft plate). The difference in the hardness is 10 shore A. These plates include a full tone area and screened surfaces of 98 %, 95 %, 92 %, 90 % and 88 % tone value in the layout. An anilox-roller with a volume of 15.4 cm³m⁻² and a screening of 120 lines cm⁻¹ was used for providing the ink transfer, and the ink viscosity was measured with a 4 mm cup (according to DIN 53211). 100 m min⁻¹ was the chosen machine speed to print on a doubled coated Liner made for flexo postprint.

Table 4 shows the findings of the print trial. The print densities of various levels were measured for each plate with a Techkon Spectro Dens. In Figure 10, example values are shown for Magenta.

	Optical print of	lensity of Cyan	Optical print density of Magenta		
Screen value in %	Flint ACE 1,7 mm	Flint ART 2,54 mm	Flint ACE 1,7 mm	Flint ART 2,54 mm	
100	1.60	1.66	1.68	1.77	
98	1.60	1.66	1.67	1.76	
95	1.57	1.64	1.64	1.75	
92	1.56	1.63	1.61	1.74	
90	1.52	1.61	1.57	1.70	
88	1.48	1.57	1.51	1.62	

Table 4: optical print densities of Cyan and Magenta (ACE Flint plates)

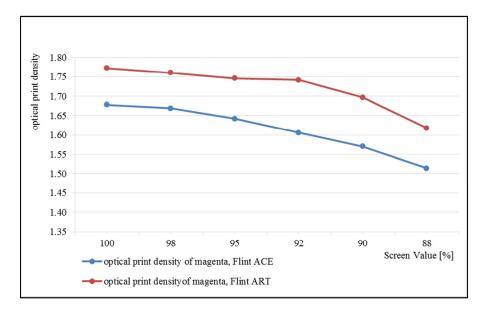


Figure 10: comparison optical print densities for different printing plates

The results demonstrate that through a harder printing plate and a modified prepress repro the ink trapping from the anilox to the printing plate and finally to the substrate can be adjusted. In spite of a reduced tonal value of 5 % in both colours, the density of the colours is nearly the same. In addition, the density difference caused by the screening value is very slight.

These findings provide a good example of how the lab printing machine is also very suitable for studying a variety of important print parameters, which are difficult to simulate by means of existing laboratory print methods.

5. Summary and Outlook

A modular reel-to-reel laboratory printing press is presented as a suitable device for simulating commercial print trials, and closes the existing gap between lab methods and commercial printing, allowing predictions of the resulting print quality of commercial prints to be made. Examples of trials in both rotogravure and flexography illustrate the potential of the device.

For the tested pilot-coated liner samples, it was clearly shown that the print quality of the laboratory flexo printing press is closely comparable to commercial presses. For the print gloss, the optical print densities and the mottle, good correlations were observed. Further work is needed to explain the small offsets between commercial and laboratory printing, but it is quite likely that the different speed of the presses plays an important role, together with factors such as plate pressure or the ink release behaviour of the anilox rollers.

Besides the flexo system, the machine can be equipped with rotogravure printing units – the validation of this printing technology (comparison to commercial presses) has already started and will be ongoing.

In the future additional print units, including high speed inkjet and offset will be implemented and validated.

References

Goodrick Ch., Master Thesis: "Comparability and practicability of laboratory scale to pilot scale endlessly coating and printing", Munich University of Applied Sciences, July 2014

Gravure Education Foundation & Gravure Association of America, "Gravure, Process and Technology" 1991 (ISBN 1-880290-00-6), pp. 324-338

Hasse A., Offset- und Flexodruckeignung von gestrichenem Liner-Karton, 20. Dresdner APV Tagung, 2009, Dresden

Johnson J. Dissertation: "Aspects of flexographic print quality and relationship to some printing parameters", Karlstad University – Faculty of Technology Science Chemical Engineering, 2008, pp. 44-55

Jutila, E., Ridgway, C.J., Gane, P.A.C. "Developing a laboratory simulation of tail edge pick", Advances in Printing and Media Technology, Vol. XXXIX, 2012, Ed. Nils Enlund and Mladen Lovrecek, (ISSN 225-6067, ISBN 978-3-9812704-5-7), 195-205.

Klein R., 2009, "*Praxis- und Labordruckergebnisse von Naturtiefdruckpapieren im Vergleich"*, Wochenblatt der Papierfabrikation 3-4/2009 pp. 120-128

Kogler W. United States Patent Patent: "Pigment misture for the paper industry consisting of calcium carbonate , dolomit or mixtures theref and talc-kaoline mixture", Patent Number: 5,120,365 Date of Patent: Jun. 9, 1992, pp. 2

Martorana E., Ziegler H., a Campo F.W., Jühe H.H., "Ursachen von Missing Dots im Tiefdruck", Wochenblatt der Papierfabrikation" 134, 11-12/2006, pp. 690-698

Nierhoff S., Möglichkeiten und Grenzen des Laborprobedrucker zur Mottling-Vorhersage, apr Magazin 12/2009, pp. 24-26

Ollech B., "Tiefdruck: Grundlagen und Verfahrensschritte der modernen Tiefdrucktechnik" 1993 (ISBN 3-87641-181-5), pp.341-347

Ridgway, C.J., Weihs, J., Grossmann, O., Hunziker, P., Gane, P.A.C. (2013): "Designing nanotechnology coatings for replacement of fibrous white top liner", Nordic Pulp and Paper Research Journal, 28(4), 2013, 560-572

Evaluation of Sensors for Inline Viscosity Measurement in Gravure Printing

Henrik Knauber, Peter Schöffler, Thomas Sprinzing, Armin Weichmann

Hochschule der Medien, Institut für Angewandte Forschung (IAF), Nobelstr. 10, 70569 Stuttgart, Germany

E-mail: hk037@hdm-stuttgart.de; ps092@HdM-Stuttgart.de; sprinzing@hdm-stuttgart.de; weichmann@hdm-stuttgart.de

Short Abstract

For the stabilisation of printing quality, gravure printing machines are normally equipped with viscosity measurement systems. Recently two newly developed viscosity measuring systems, a microelectromechanical tuning fork sensor and an acoustic wave sensor, were introduced to the market. Those systems compete with the traditional rotary viscometer and the dropping body measurement. A system comparison of the different systems were implemented and performed at a Rotomec MW 60 rotogravure press. The aim was to find a system for inline viscosity measuring of printing inks, which is as accurately as possible and does need minimal cleaning effort. Four experiments were conducted to evaluate the different viscosity measuring instruments, namely, measurement accuracy of the solvent concentration, measurement capability, temperature behaviour in ink and influencing factors of viscosity measurement in the printing process. The results show that the acoustic wave sensor and the rotary viscometer are suitable for the viscosity measurement.

Keywords: Inline viscosity measurement, viscosity sensors, tuning fork sensor, acoustic wave sensor, rotary viscosimeter

1. Introduction and background

For the stabilisation of printing quality, gravure printing machines are normally equipped with viscosity measurement systems. Recently, two newly developed sensor systems were introduced to the market, a micro-electromechanical tuning fork sensor and an acoustic wave sensor. Those systems compete with the traditional rotary viscometer and the dropping body measurement. With regard to the technical equipment of the machine, a system comparison of the different measurement systems were implemented at a Rotomec MW 60 rotogravure press.

For the examination of the different sensors, four tests have been designed, precision, behaviour during the printing process, temperature behaviour in ink, and the behaviour to external disturbing factors. Based on these tests recommendations were derived.

Viscosity is expressed in different measurement units. Most commonly used are the kinematic viscosity v and the dynamic viscosity η . The kinematic viscosity arises from dynamic viscosity by dividing it through the specific density of the sample. In gravure printing efflux cups are most common and therefore with these viscosity is measured in (efflux cup) seconds.

Acoustic wave sensors measure the product of dynamic viscosity and the specific density of the measurement sample. The measurement of an acoustic wave sensor is based on the acoustic impedance

$$Z = (\omega \cdot \rho \cdot \eta)^{1/2} \qquad [1]$$

where

 ω represents the angular frequency $\omega = 2 \cdot \pi \cdot f$,

 ρ the specific density and η the dynamic viscosity (Praxis Sensorik, 2008).

To measure the viscosity, the resonator, an electrostrictive quartz crystal plate, is put in contact with the fluid. The resonator is driven by the electrode at the bottom surface (Figure 1) and moves sinusoidally perpendicular to the sensor surface with the frequency ω and the amplitude U. The frequency is defined by the construction of the sensor, whereas the amplitude depends on the strength of the applied electrical signal. A certain layer of the fluid is hydrodynamically coupled with the sensor surface, its thickness dependent on the viscosity.

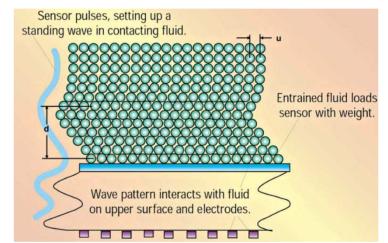


Figure 1: Cross section of acoustic wave sensor (Special Bahnelektronik, 2007)

The penetration depth (d) of the wave into the fluid is dependent on the frequency, the viscosity and the specific density of the fluid:

$$d = \left(\frac{2 \cdot \eta}{\omega \cdot \rho}\right)^{\frac{1}{2}}$$
[2]

The acoustic viscosity is measured through the power consumption of the quartz resonator that emits ultrasonic waves into the measurement fluid to a certain depth (d). The measured number (referred by the vendor as "acoustic viscosity") is the product of the specific density and the dynamic viscosity and has the unit $g/cm^3 \cdot mPa \cdot s$ (Special Bahnelektronik, 2007).

$$\eta_A = \frac{Z^2}{\omega} = \eta \cdot \rho \qquad [3]$$

The micro-electromechanical tuning fork sensor delivers specific density, viscosity and temperature, thus it measures kinematic and dynamic viscosity simultaneously. The sensor can be seen as a tuning fork with a flat profile at its end, which is electrically stimulated to an elliptical oscillation within a fluid with its resonant frequency. The surrounding medium impacts the resonant frequency of the tuning fork. As the fork profile is different in the two oscillation directions, the detuning in this two directions provides two distinct values, therefore enabling the calculation of both, dynamic viscosity and specific density.

A rotary viscometer served as a reference instrument, which was placed directly into the ink tank. As the name suggests the sensor has a cylindrical measuring body which rotates around its axis. With increasing viscosity of the measured fluid the torque resistance at the measuring body increases. As the torque of the motor is held constant, the rotational speed decreases. The decreasing is captured by the evaluation unit and transformed to dynamic viscosity.

2. Materials and methods

2.1 Experiments

Four experiments were conducted to evaluate the different viscosity measuring instruments.

2.1.1 Experiment 1: Measurement accuracy of the solvent concentration

Reproducibility of the viscosity measurements was tested with different inks representing the whole process range of solvent concentrations. The data obtained from this experiment were used to map the individual measurements to $L^{\%}$ (solvent content of the ink).

2.1.2 Experiment 2: Equipment capability study

The equipment capability study is a method to evaluate the accuracy and reliability of different measurement systems. This study allows researchers to analyse and determine the best measurement system for their respective areas of application. The commonly used method resulted from decades of experience within the automotive industry. Three analytical procedures, as shown in Table 1, are foreseen:

Procedure	Purpose	Parameter
Procedure 1	Systematic error and repeatability	C_{g}, C_{gk}, t -Test,
		confidence intervals
Procedure 2	Repeatability, reproducibility (with	%R&R, ndc
	operator influence)	
Procedure 3	Repeatability, reproducibility	%R&R, ndc
	(without operator influence)	

Table 1: Equipment capability study

Procedure 1 is normally used by the manufacturer of the measurement system to prove the suitability and capability. Procedure 2 and 3 are deployed by the user to confirm suitability on-site.

2.1.2.1 Procedure 1

The two most important quality parameters for the measurement equipment are the C_g and C_{gk} -Value. Procedure one uses these values to determine the parameters within its application. With this procedure measurement series are made at a calibration standard and out of these series (at least 25 values) the arithmetic average and standard deviation are calculated. After that the C_g und C_{gk} -Value, in combination with the specified characteristic tolerances, can be calculated. A measurement system is defined capable if the C_g und C_{gk} -Value are greater than 1.3.

2.1.2.2 Procedure 2

Procedure 2 is also known as %R&R or as the GR&R-Study (Gage repeatability & Reproducibility). After defining the amount of test objects (n) and number of testers (k), whereby the test objects should cover the process range, the number of repeats (r) are determined, where $n \cdot r \cdot k > 30$ (Dietrich E, Schulze A., 1998). An analysis of variation (ANOVA) is made and values obtained are repeatability (EV), reproducibility (AV) and part deviation (PV). EV is a value for the influence of the measurement equipment, whereas the AV-value is a measure of the user's influence.

The R&R-value is calculated by

$$R\&R = \sqrt{EV^2 + AV^2} \quad [4]$$

For a particular case the single values (EV, AV, PV, R&R) are set into relation with the benchmark RF. RF can be the seen as the tolerance (T) of the process or the total deviation (TV) out of the process deviation. If the process deviation is unknown, the TV-value arises out of the following relation:

$$RF = \sqrt{EV^2 + AV^2} \qquad [5]$$

The relative values %EV, %AV, %PV, %R&R result by dividing EV, AV, PV, R&R by RF.

The evaluation of the measurement results can be made according to the Ford guideline EU 1880:

%R&R ≤ 20 % measurement system is capable %R&R ≤ 30 % measurement system is partly capable %R&R ≥ 30 % measurement system is not capable

2.1.2.3 Procedure 3

In this survey procedure 3 is used. Procedure 3 is a derivation of procedure 2, where the influence of the user is omitted. This is possible if the user has no influence on the results, like in automated measurements or inline systems. In this case $n \cdot r > 20$ applies, with n the number of test objects and r the number of repeats. If there are no different test objects the amount of repeats has to be increased. As already described the ANOVA is used for the evaluation of the system, where the reproducibility (AV) equals zero. The R&R-value corresponds in that case to the repeatability (EV).

2.1.3 Experiment 3: temperature behaviour in ink

The change in viscosity was measured as a function of temperature. The aim was to examine the behaviour of the different measuring instruments when handling ink which varies in temperature, e.g. cold ink at start up or heated up ink during running the printing process.

2.1.4 Experiment 4: Influencing factors of the viscosity measurement in the printing process

Typical influencing parameters to the measurement process, printing speed, flow rate of ink within the inking system and micro foaming extent were varied on press. The aim of the experiment was to determine the viscosity measuring instrument with the best resilience within the process environment.

2.2 Inks and solvents

Rotogravure printing inks are organic solvent or water-based inks of low viscosity with a dynamic viscosity η ranging from 10 to 200 mPa·s. To achieve the correct viscosity, the inks are mixed with solvent, which affects the pigment concentration and optical density of the colour.

As solvent evaporates out of the ink during printing, the viscosity increases. Additionally the behaviour of the ink changes in several respects, including colour strength (pigment concentration), fluid behaviour during cell filling and doctoring, and the behaviour within the printing nip and drying section. For this paper conventional process colours with toluene (publication rotogravure printing) and ethanol / ethyl acetate (packaging gravure printing) in different concentration levels were used. Additionally, a white gravure ink was mixed with varying concentrations of ethyl acetate.

2.3 Experimental setup

The viscosity measuring instruments were installed in the ink circulation of a Rotomec MW60 gravure printing press, which is driven by a double acting pneumatic pump. Figure 2 illustrates the individual components of the general experimental setup.

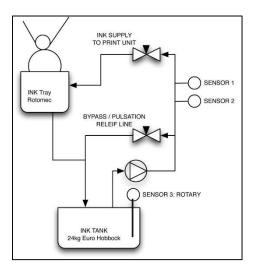


Figure 2: Experimental setup. Sensor 1: Tuning fork; Sensor 2: Acoustic wave, 3: Rotary

2.4 Reference system: solvent concentration

The meaning of the values of the different viscosity measuring instruments differ according to the different measuring principles (see section 1) and not all deliver dynamic viscosity. Therefore the correlation between the different systems was based on a reference system solvent concentration: L% in %, the quotient of the amount of solvent additionally poured into the basic ink, delivered by the ink manufacturer, to the amount of the basic ink itself. As the viscosity is indirectly proportional to this number, the transformation [6] was used to match the values of the devices to L%, with characteristic constants *a* and *b* for each device and ink type.

$$y = \frac{a}{x} + b \qquad [6]$$

x : solvent concentration L% in %,

y : value of viscosity measurement device.

3. Results and Discussion

3.1 Experiment 1: Measurement accuracy of the solvent concentration

All measurement values were converted through [6] to the solvent content and given as L%. Figure 3 shows these L% values of the devices compared to the actual solvent concentration L%.

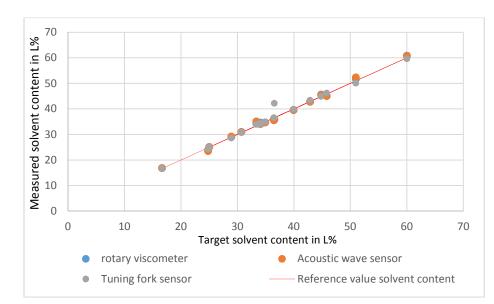


Figure 3: Target-actual comparison of the three sensors for the different dilutions.

All instruments can reproduce the different dilutions levels with good precision with the exception of one outlier, presumably a faulty measurement. The acoustic wave sensor has the lowest measured standard deviation of ± 0.07 L% of all dilutions, the tuning fork sensor the highest with ± 0.09 L%, the rotary viscometer is in between: ± 0.08 L%.

3.2 Experiment 2: Equipment capability study

The testing fluid for the evaluation was Black NC 133 from Siegwerk with 150 % extender and ethanol as solvent with a solvent content of 36.44 L%. The tolerances were defined to +/- one second (3 mm DIN cup), which is a common number in order to avoid variations in print. For a solvent content of 36.44 % one cup second (3 mm DIN cup) corresponds to a solvent content of 2 %. This leads to a tolerance of 4 L%.

	Rotaryviscometerin L%	Acoustic wave sensor in L%	Tuning fork sensor in L%
Basic size in L%	36.44	36.44	36.44
Tolerance in L%	4	4	4
Mean in L% Standard deviation in L%	36.107 0.124	35.53791378 0.049	36.00670819 0.698
Cg	1.607	4.00	0.286
C _{gk}	0.243	-5.09	-0.029
T _{min/Cg} in L%	3.309	1.32	18.59
T _{min/Cgk} in L%	-0.085	-7.76	14.18
%EV	17.5	5.76	71.61
%R&R	17.5	5.76	71.61

Table 2: Results for the equipment capability study procedure 1 and 3

The equipment capability study showed that the rotary viscometer and the acoustic wave sensor are capable with a C_g 1.61 and 4.00 respectively. The tuning fork sensor is with C_g 0.28 far below the critical value of 1.33 and therefore is not capable. For the tuning fork sensor at the best a tolerance of

18.59 L% can be achieved. That means that variations of ± -4.5 flow cup seconds (3 mm cup) are possible. A reason for the poor results of the tuning fork sensor could be the experimental setup, where the cross section of the tube, the sensor was integrated in, was too low and the measurements were affected by the pulsation of the pump.

The results of procedure 3 confirm the results of procedure 1 and show that the acoustic wave senor has a very good repeatability (%EV = 5.76). The rotary viscometer with %EV of 17.5 is also below the limit of 20 % and therefore is capable.

3.3 Experiment 3: Temperature behaviour in ink

The black ink was stored in a closed tank for 24 hours at 7°C. During the measuring operation the ink container was heated from 12° C to 36° C. The measured values of the three sensors are shown in Figure 4.

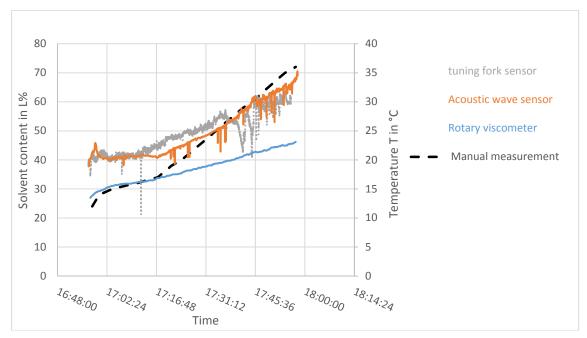


Figure 4: Temperature behaviour in ink (higher L% mean lower viscosity)

The viscosity change by increasing the ink temperature is accurately reproduced by the sensors. However the slope of the increase is significantly lower than the slope measured with an efflux cup (manual measurement). This can be explained by density changes, which occur in parallel to the lowering of the viscosity and which are not registered by the efflux cup. As all sensors measure either kinematic viscosity or the acoustic impedance acc. to [1], they are sensible to density changes too and have to be calibrated separately to compensate for temperature changes.

3.4 Experiment 4: Influencing factors of the viscosity measurement in the printing process

Firstly the effect of printing speed was investigated. The diving of the cylinder with the empty cells into the ink brings many small air bubbles into the ink and creates micro foaming. The higher the speed, the bigger this effect is.

The following tables (Table 3, Table 4 and Table 5) show the sequence of the test procedures.

Table 3: Process description printing speed

Influence Nr.	Time	Process Description	
1	11:32	Rotary viscometer not surrounded with ink	

2	11:44	Added ink
3	12:10	Printing speed: 180 m/min
4	12:15	Rotary viscometer not surrounded with ink
5	12:30	Added ink
6	12:35	Reduced printing speed: 60 m/min
7	12:54	Increased printing speed: 240 m/min
8	12:57	Reduced printing speed: 60 m/min

Additionally the pumping creates foaming too. This effect was investigated in a second measurement series by varying the pressure to the pneumatic pump therefore modifying the ink flow and additionally the temporal pressure profile within the tubes. The higher the flow rate, the more foam is created. Additionally, as the tuning fork sensor and the acoustic wave sensor are located within the pumping tube line, the flow through or along the sensor was expected to affect the measurement.

Influence Nr.	Time	Process Description
9	13:05	Bypass: off
10	13:08	Air throttling: open
11	13:11	Pump pressure: 3.5 bar / Bypass: closed / Air throttling: open
12	13:14	Pump pressure: 3.5 bar / Bypass: closed / Air throttling: open / Ink throttling: slightly open
13	13:18	Pump pressure: 3.5 bar / Bypass: closed / Air throttling: open / Ink throttling: half open
14	13:21	Pump pressure: 3.5 bar / Bypass: closed / Air throttling: open / Ink throttling: fully open
15	13:23	Pump pressure: 2.2 bar / Bypass: closed / Air throttling: open / Ink throttling: fully open
16	13:26	Pump pressure: 2.2 bar / Bypass: open / Air throttling: open / Ink throttling: fully open
17	13:29	Standard-setting as 13:05 clock

Often a stirring rod is used to ensure a stable dispersion of the ink, especially with special effect pigments (metallic, irodine) or with white. This again results in enhanced foaming. Additionally the rotary sensor could be affected by the ink flow generated in the tank. Therefore a third measuring series was set up with a stirring rod.

Influence Nr.	Time	Process Description
18	14:18	Printing speed: 300 m/min (without substrate)
19	14:24	Activation of stirrer
20	14:39	Aspiration of foam from the ink surface
21	14:42	Without ink tray
22	14:46	Aspiration of foam from the ink surface

Table 5: Process description micro foam

3.4.1 Results: rotary viscometer

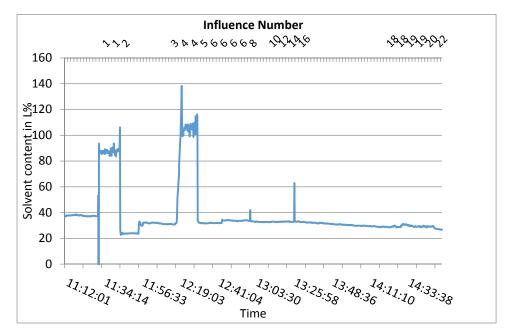


Figure 5: Measurements of rotary viscometer through measurement series

In the period 11:30 - 11:45 and 12:20 - 12:30 the measurements are incorrect, because the rotary viscometer was not surrounded with ink. The addition of solvent at 11:53 is correctly reproduced by the rotation viscometer. The graph shows the ongoing test procedure in a relative smooth graph.

Between 12:35 to 13:00 the printing speed was changed and between 13:05 and 13:30 parameters of the pump stroke varied. To the end of the experiment, the influence of micro foam was checked. The curve slightly descends, as the solvent evaporates slowly. The rotary viscometer can be described as highly resistant to the tested influencing factors.

3.4.2 Results: Acoustic wave sensor

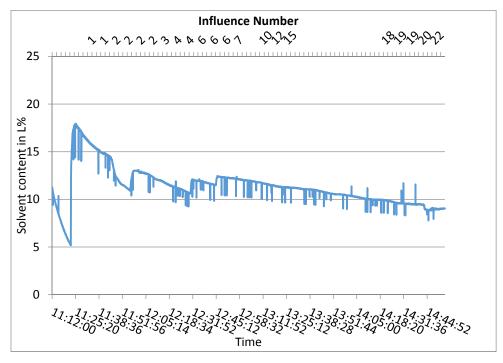
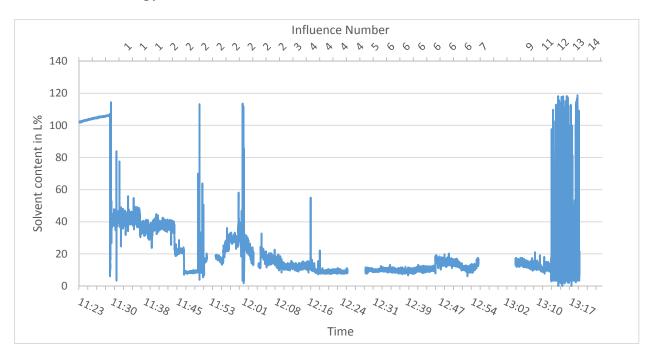


Figure 6: Measurements of acoustic wave sensor through measurement series

The addition of solvent at 11:53 is correctly reproduced by the acoustic wave sensor. Nevertheless the values decrease after reaching a local maximum more than expected, exaggerating the effect of the solvent evaporation.



3.4.3 Results: Tuning fork sensor

Figure 7: Measurements of tuning fork sensor through measurement series

The tuning fork sensor is susceptible to pump strokes and micro foam and produces a lot of incorrect measurement values. During the variation of the pump settings measurement errors resulted when the ink throttle was opened. The reason may be that the diameter of the tube, where the sensor unit sits in, has a diameter which is too small, therefore causing high flow speeds and high pressure peaks, which disturb the measurement. Consequently, a change in the ink flow has a negative effect on the value acquisition by the tuning fork system.

3.4.3 Discussion

The evaluation of the three investigated measuring sensors showed that the tuning fork sensor is least tolerant to the pump strokes and to the micro-foaming. Pneumatic pumps create a pulsing ink flow, therefore strongly varying fluid velocities and pressures. The measurement is performed through the elliptical oscillations of a rod positioned within the tube for the ink transport from bucket to ink pan, perpendicular to the vector of ink flow. Varying pressures might impact on the rod during pulse cycles therefore generating erroneous values. A special shunt with homogenized and slow ink flow might solve this problem, however adding to the effort on installation and cleaning. Additionally it can be assumed, that micro bubbles, which come in proximity to the oscillating rod change the behaviour of the fluid significantly, as they change the mean density and, as relative big "particles" within the fluid, the mean mobility of the fluid. As this is the number to be measured, these micro bubbles influence every measurement system. Nevertheless the tuning fork principle seems to be most sensitive to this. The acoustic wave sensor is difficult to classify because it is unclear why the measured values amplify the decrease of viscosity due to solvent evaporation. As this sensor measures the "acoustic viscosity", which is the product of dynamic viscosity and density and both parameters increase with lower solvent content, it could be assumed that the overall signal is more sensitive compared to measuring only dynamic viscosity. In any case, the density and its change have to be taken into account when calibrating and transforming the measured values from this measuring principle to one which measures the dynamic viscosity. Unfortunately we did not measure the density with a separate measuring device, so we cannot confirm this assumption yet. (The tuning fork sensor delivers values for the specific density too, but the quality of these values was uncertain to the same extend as the values for the dynamic viscosity itself.)

The rotary viscometer performed as expected over the entire experimental period. If it is ensured that the sensor is always completely surrounded with ink, print speed and pump strokes do not influence the measurement and micro foam does only to a small extent.

4. Conclusion

The tests showed that basically all sensors are capable of gathering viscosity. The different levels of dilution are captured accurately by all measurement systems. However the acoustic wave sensor delivers the smallest measurement variations over all levels of dilution. This result is verified in the equipment capability study, where the acoustic wave sensor proved to be the most capable of the three sensors. The tuning fork sensor could not prove its capability in the test set up used.

As the sensors use different measuring principles, they respond differently to temperature variations of the ink, the coupled viscosity and density changes and have to be calibrated to correct for this effect.

The tuning fork sensor reacts very sensible to pump strokes of the pneumatic pump and to micro foam and therefore is without serious change of the setup used not suitable for an inline viscosity measurement system. The acoustic wave sensor is much more insensitive to these effects. Nevertheless, before applying this sensor it has to be investigated if the measuring principle (product of dynamic viscosity and specific density) sufficiently explains the measured stronger decline compared to the rotary viscometer when solvent evaporates out of the ink. The rotary viscometer is capable and represents the current state of the art in the printing industry. However, it was the aim to find a system, which is accurate, minimize cleaning effort and can be installed on the press with minimal handling issues. In this context the acoustic wave sensor is to be preferred.

References

Dietrich E, Schulze A., 1998. *Statistische Verfahren zur Qualifikation von Messmitteln, Maschinen und Prozessen*. 3rd ed. München, Wien, 1998

Praxis Sensorik, 2008. Schnelle Messungen der Viskosität. Elektro AUTOMATION; September 2008:84-7.

Special Bahnelektronik, 2007: Messung der Ölviskosität. Elektronik Industrie 2007:72-4.

The Statement and Investigation of the Problem of Separation of a Paper Sheet from the Offset Cylinder after Printing.

Rostislav Moginov, Alexander Vorozhtsov

The Moscow State University of Graphic Arts (MGUP), St. Pryanishnikova 2A, Moscow, Russia, 127550

E-mail: kvd21@mail.ru; a vorozhtsov@bk.ru

Short Abstract

The problem of determining the ratio between the speed of printing and thickness of ink film remaining on the printed sheet is an important element in determining quality of the publication. We consider the condition conduct separation process layer of viscoplastic deformation of the liquid at the quasi-equilibrium (without discontinuity layer). The separation of paper from printing cylinder to paper surface acts tensile stress that occurs in the decomposition of the paint layer. You observe the condition that in place of deformation (flow) come gap layer and dispersion. The task includes the definition of the separation force F_T is applied to the sheet printed material. Breakout force F_T – this force that is applied valves printing cylinder. Force F_T applied to the sheet at the moment separation ink from the printing cylinder, affects the accuracy sheet feeding during multicolor offset printing. The resulting mathematical model allows calculating the force, which is necessary for the printing cylinder valves during the removal of the sheet.

In this paper analytical calculation of the breakout force of the sheet which should be attached to the printing cylinder valves, so that the sheet completely off from the surface offset cylinder. Knowing the breakout force, you can calculate the number of capture valves on a printing cylinder of a sheet fed printing presses. The urgency of the task is consistent with the variety of printed materials, including on non-absorbent surfaces. Such variety of printed materials can lead to changes in structural parameters and sheet fed settings printing equipment.

Keywords: ink, offset printing, sheet separation, rheological behavior, paper, misting.

1. Introduction

The problem of determination the ratio between the speed of printing and ink film thickness remaining on the sheet is an important element in specification the quality of the publication. Kinetics of delaminating (splitting of ink layer) in the separation of the sheet to determine the relationship between the paper sheet tension and the angle of its capture represented in the work (Mueller. P. 1988). Evaluation the criterion speed separation of the layers of ink between two plane plates considered in (Tikhonov V.P., Gulyaev S.A., Semenyuta S.S. 1993), (Yakhnin E.D. 1987), (Claypole T., Vlachopoulos G., Bould D. 2010), (Laurette Vieille-Grosjean, Alice Vermeulin 2012), (Ruemer Klein, Patrick Cunningham, Gerd Meder, Martina Miletic 2012). Relying on previous research in this article, we consider the condition of the separation process of the layer of viscoplastic fluid with a quasiequilibrium deformation (without discontinuity layer) and the condition of the gap, and, therefore, conditions for the stable of the printing process.

The processes of ink layers separation in offset printing are considered in the fundamental work (McPhee J., 1998). But in this work not considered the dynamics of the separation ink layer in the transition to the printed material, and there is no calculation of the breakout force of the printed sheet from the blanket surface (McPhee J., (1998).

The effect of temperature parameters on the separation (misting) of the ink layer is considered in the works (Claypole T., Vlachopoulos G., Bould D. 2010), (Savarmand S., Bousfield D., Durand R., Warren R. 2010). However, in these studies does not contain an analytical description of this process.

The theoretical positions of the works (Claypole T., Beynon D., Hamblyn S. 2008), (Klaulitz T., Dorsam E. 2008) focuses on the properties of the printed surface but not to the process of separation of the ink layer, which is reflected in the analytical descriptions presented in these works and research results.

In (Dube M., Drolet F., Bloch J.-F., Daneault C., Mangin P. 2007), (Ozaki Y., Fujisawa N., Kawamura E. 2008) was used other research methods when the examina- experiments. Obtained research results in these studies and their practical application were examined under the other point angle of view in comparison with offered in this article method.

2. Materials and Methods

In sheet fed offset printing machines offset cylinder interact with an ink and paper during the print. In the area of separation of paper from the blanket cylinder to the paper surface acts tensile stresses which occur during cleavage of the ink layer (Claypole T., Vlachopoulos G., Bould D., 2010).

Consider the process of separating the fluid layer of viscoplastic deformation in quasi-equilibrium, provided that such a change of deformation (flow) will break the layer and dispersion.

Denote certain conditions: considering the time of separation ink from the blanket cylinder, we believe that m1 quantity of ink on the material and m2 quantity of ink on the blanket are equal (m1 = m2). The thicknesses of the ink layer on the blanket and on the paper are also equal and have values 1.5 μ m (microns). Frictional forces is neglected, as well as centrifugal force. Cohesive force is significantly lower adhesion force, that mean Fk << Fa.

The task is to determine the force F_T , which you need to exert to the sheet of paper to transfer the ink (50% ink layer) from the blanket cylinder to the printing material.

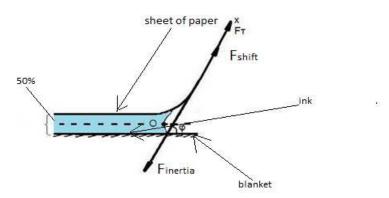


Figure 1 Diagram of the separation process the sheet of paper from the blanket cylinder

2.1 Solution of the problem

According to Newton's second law:

$$F_{\rm T} + F_{\rm SHIFT} + F_{\rm INERTIA} = 0$$
^[1]

$$F_{\rm T} = F_{\rm INERTIA} - F_{\rm SHIFT}$$
[2]

Find the force F_{INERTIA}, using the equation Meshcherskij (Drong V.I. and others 2005):

$$F_{\text{INERTIA}} = \frac{dm}{dx} V x$$
 [3]

Where: $Vx - speed of the separated particle, \frac{dm}{dx} = 1$ (by the hypothesis).

To find Vx consider one-dimensional problem (see Figure 2):

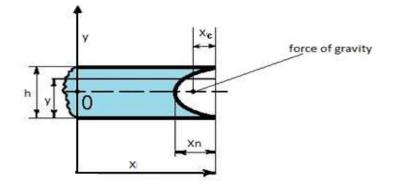


Figure 2 Model of deformation of a liquid between the surfaces when they are moving apart

We estimate the criterial speed of separation layer of viscoplastic fluid situated between two surfaces. Below this speed deformation fluid occurs so that the shape of the surface formed at each moment to adopt the equilibrium moving apart succeeds view. In this model (Figure 2) fluid volume V_0 is constant. Wetted perimeter defined by a radius x_0 and also remains constant. Moving apart the plates is carried out in the Y direction at a constant velocity Vy.

The volume of fluid located between the surfaces, which moving apart, in a general form can be represented as:

$$V_0 = \pi X_0^2 Y - 2\pi (X_0 - X_C) \times S_{ABC}$$
[4]

Where: Y – the distance between the surfaces; X_C – coordinate of the center of gravity of the cross section S_{ABC}

Since X_0 exceeds Y_0 by several orders of magnitude, the profile of the fluid meniscus, during the surface moving apart, will have the shape of a parabola:

$$S_{ABC} = \frac{2}{3}XY$$
[5]

Where: X_n – coordinate of the vertex of the parabola (Vygodsky M.J. 2010).

Coordinate of the center of gravity of the cross section of a parabolic will be:

$$X_{C} = \frac{2}{5}X$$
[6]

Substituting the values of S_{ABC} and X_C in equation [4], we obtain:

$$V_0 = \pi X_0^2 Y - \frac{4}{3}\pi X_0 \times X \times Y + \frac{4}{3}\pi \times \frac{2}{5}X \times X \times Y = \pi X_0^2 Y - \frac{4}{3}\pi X_0 \times X \times Y + \frac{8}{15}\pi X^2 \times Y$$
[7]

Differentiating equation [7] with respect to time and using the notation:

$$\frac{\mathrm{d}y}{\mathrm{d}t} = V_{\mathrm{y}}; \ \frac{\mathrm{d}x}{\mathrm{d}t} = V_{\mathrm{x}}; \ \frac{\mathrm{d}V_{\mathrm{0}}}{\mathrm{d}t} = 0$$
^[8]

Obtain:

$$0 = \pi X_0^2 \frac{dy}{dt} - \frac{4}{3}\pi X_0 \frac{d}{dt} (X \times Y) + \frac{8}{15}\pi \frac{d}{dt} (X^2 Y)$$
[9]

By transforming equation [9] we get:

$$0 = \pi X_0^2 \frac{dy}{dt} - \frac{4}{3}\pi X_0 \left[X \frac{dy}{dt} + Y \frac{dx}{dt} \right] + \frac{8}{15}\pi \left[2X \frac{dx}{dt} \times Y + X^2 \frac{dy}{dt} \right]$$
^[10]

In view of [8] we get:

$$0 = \pi X_0^2 V_y - \frac{4}{3} \pi X_0 [X V_y + Y V_x] + \frac{8}{15} \pi [2X \times Y \times V_x + X^2 V_y]$$
[11]

From [11] we find Vy depending on Vx, i.e. we have:

$$\pi \left[X_0^2 - \frac{4}{3} X_0 \times X + \frac{8}{15} X^2 \right] Y = \pi Y \left[\frac{4}{3} X_0 - \frac{8}{15} Q_x \right] V_x$$
^[12]

From [12] we obtain:

$$V_{y} = \left(\frac{Y\left[\frac{4}{3}X_{0} - \frac{16}{15}X\right]}{\left[X_{0}^{2} - \frac{4}{3}X_{0} \times X + \frac{8}{15}X^{2}\right]}\right) \times V_{x}$$
^[13]

Expression [13] makes it possible to estimate the distance between the plates Y = YMAX at which one closing lines of the free surface (vertex of the parabola) will be in the center of the fluid layer (point O). Since in this case $X = X_0$ from equation [7] we have:

$$Y_{max} = 5\pi \frac{V_0}{X_0^2}$$
[14]

Since the volume of fluid between the plates is not changed and is equal to the volume of moving apart plates, then substituting the value V_0 of the equation [8] into equation [14] we obtain:

$$Y_{\max} = 5Y_0$$
^[15]

The maximum stress at the interface between the newly formed fluids occurs in areas with the smallest radius of curvature. The radius of curvature will be situated in the middle layer, i.e. the vertex of the parabola. The parabolas radius at its vertex is calculated by the formula:

$$R = \frac{Y^2}{8X}$$
[16]

Surface tension at a given point in accordance with Laplace's formula must be equal to:

[18]

[26]

$$P_{\rm L} = \frac{\sigma}{R} = \frac{8\sigma X}{Y^2}$$
[17]

Where: P_L – Laplace pressure, and σ – surface tension of the ink (fluid) on the border with the air. In steady quasi-equilibrium deformation we have:

$$P_L = S_M$$

Where: S_M – limiting the possible tension in the fluid without breaking its continuity.

$$S_{\rm M} = \frac{8\sigma X}{Y^2}$$
^[19]

During the deformation of fluid in any direction the fluid stresses are proportional to the product of its viscosity η relative to the rate of deformation:

$$S_{M} = \frac{V_{y}}{\Delta Y} \times \eta$$
^[20]

Using equations [18] and [19] we have:

$$\frac{V_y}{\Delta Y} \times \eta = \frac{8\sigma X}{Y^2}$$
^[21]

Since the plates are moving apart at a constant rate, then at the time of the separation rate does not change. Substituting in the expression [13] $X = X_0$ and $Y = Y_{MAX} = 5Y_0$, we get:

$$V_{y} = 1,28 \frac{\sigma}{\eta} \left(\frac{X_{0}}{Y_{0}} \right)$$
^[22]

As follows from the above, at the speeds moves apart the surfaces less than or equal speeds obtained from the expression [22] fluid layer deformation is going quasi-equilibrium. I.e. fluid layer gradually becomes thinner thread is pulled in and breaks up. Therefore, the value of Vy from equation [22] should be considered as critical, above which the separation of fluid between the plates will be in violation of the continuity in its entirety. The separating speed Vx of the ink's layer is determined from formula [13]:

$$V_{x} = V_{y} \left(\frac{X_{0}^{2} - \frac{4}{3}X_{0}X + \frac{8}{15}X^{2}}{Y\left[\frac{4}{3}X_{0} - \frac{16}{15}X\right]} \right)$$
[23]

Accept the initial conditions X = 0 and $Y = Y_0$, then equation [23] takes the form:

$$V_{x} = V_{y} \frac{X_{0}^{2}}{\frac{4}{3}X_{0} \times Y_{0}} = \frac{3}{4} \frac{X_{0}}{Y_{0}} \times V_{y}$$
^[24]

Substituting Vy from [22] to [23] we get:

$$Vx = \frac{3}{4}1,28\frac{\sigma}{\eta}\frac{X_0}{Y_0} = 0,96\frac{\sigma}{\eta}\left(\frac{X_0}{Y_0}\right)^2$$
[25]

Nature of F_{SHIFT} paper sheet passing through a printing nip in a plane offset printing machine discussed above (see Figure 1). The differential equation describing the state of the paper in contact with fluid is described by the equation in the [1].

$$\frac{\mathrm{d}^5 \mathrm{y}}{\mathrm{dx}^5} - \frac{\mathrm{G}}{\mathrm{V}\eta} \frac{\mathrm{d}^4 \mathrm{y}}{\mathrm{dx}^4} + \frac{\mathrm{G}}{\mathrm{EIy}_0} \frac{\mathrm{dy}}{\mathrm{dx}} = 0$$
^[26]

Referring to Figure 2, at which point b is the point of maximum stress S_M , that is, the point at which a fluid cavitations pores begin to appear, we get:

$$S_{\rm M} = M_0(p^2 + q^2) + 2pP\sin\phi$$
 [27]

Where: M_0 – moment point bending sheet arising in the splitting of the ink layer; p and q – numerical coefficients obtained by solving equation [26]; P – shear strength.

Knowing the maximum voltage S_M, in which the ink starting to appear cavitations pores, we find P:

$$S_{\rm M} = \frac{V_{\rm y}}{\Delta Y} \times \eta = 0.32 \frac{X_0}{Y_0^2}$$
^[28]

$$\Delta Y = Y_{max} - Y_0 = 5Y_0 - y_0 = 4Y_0$$
^[29]

By means of program MathCAD calculate the parameters M₀, p and q:

$$p = 6,204 \times 10^4;$$
 $q = \sqrt{2,428 \times 10^{-3}};$ $M_0 = 0,001$

From equation (27) we calculate the value of P is equal to:

$$P = \frac{S_{\rm M} - M_0(p^2 + q^2)}{2p\sin\phi} = 2.8 \times 10^2$$
^[30]

Thus:

$$F_{SHIFT} = P \sin \phi$$
[31]

Then, an angle equal to $\varphi = 45^{\circ}$; $\sin \varphi = \sin 45^{\circ} = \frac{\sqrt{2}}{2}$; $F_{SHIFT} = 1,98N$

The numerical calculation produces the following values: $Y_0 = 3 \ \mu m;$ $X_0 = 8;$ 6; 4 mm; $\frac{\sigma}{n} = 0.95 \times 10^{-3}$

By the equation [25] we obtain the following values $F_{INERTIA}$: $F_{INERTIA} = 6,5 \text{ N}$ $X_0 = 8 \text{ mm}$ $F_{INERTIA} = 4,6 \text{ N}$ $X_0 = 6 \text{ mm}$ $F_{INERTIA} = 3,3 \text{ N}$ $X_0 = 4 \text{ mm}$

Then we get from the equation [2] we obtain the following values F_T : $F_T = 6,5 - 1,98 = 4,52$ $X_0 = 8 \text{ mm}$ $F_T = 4,6 - 1,98 = 2,62$ $X_0 = 6 \text{ mm}$ $F_T = 3,3 - 1,98 = 1,32$ $X_0 = 4 \text{ mm}$

3. Results and Discussion

Choosing ink was determined on the basis of a low molecular weight known and constant pigment carbon black used in the manufacture of black ink. Black ink produced – UF «Sicura Plast 4770» of «Sieqwerk». Viscosity – 20.5 - 42.5 Pa × s.

Printed material, physical and mechanical properties are: thickness -0,035 mm; the weight -32.0 g/m^2 ; tensile strength in the longitudinal direction.

Place of experiment – laboratory of the Department "Technology of Printing Production", The Moscow State University of Graphic Arts Moscow (Russia), printing "Almaz-Press", Moscow (Russia), "Goznak", Borisov (Belarus).

In the experiment, – modeling of the separation sheet from the blanket cylinder, the following results were obtained and presented in Table 1 and as the graphs on Figure 3.

	h, µm (microns)	F _T , N	V, m / c
Printing material	1,7859	5,42	7,40
		3,27	5,25
		3,02	5,00
		1,32	3,30
		0,17	2,15

Table 1 Experimental data

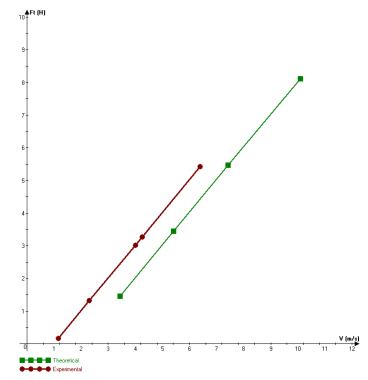


Figure 3 Graphic dependence of the detachment force on the speed

In Figure 3, the deviations of the experimental results from the calculated theoretical represent 12 percent, which indicates a high degree of confidence in the results of the study.

The graphs show that the power of the printed sheet separation increases with the speed of printing and the dependence itself is linear.

4. Conclusions

1). The use of non-absorbent materials for printing is growing every day. Print on polymeric materials are always complicated because of the worsening problem of misalignment colors in colorful prints and dot gain, which is caused by stretching of the material web between inking units of printing presses sections, and the problem of adhesion of the ink to the surface of the polymer film.

2). On the basis of theoretical and experimental studies solved the problem of quantifying the detachment force of the printed sheet from the blanket cylinder in the printing nip zone.

3). The results will allow for an analytical level, to produce calculations for stabilization of the offset printing process.

4). The results will allow to accelerate the process of standardization and certification for a broader range of new printed materials, including synthetic non-absorbent flap surface, in the framework of ISO 12647-2.

References

Claypole T., Beynon D., Hamblyn S. (2008), "Ink transfer during flexographic printing to paper", IARIGAI vol. XXXV, pages 181-188

Claypole T., Vlachopoulos G., Bould D., (2010), "Ink misting formation in roller trains", IARIGAI vol. XXXVII, pages 227-234

Drong V.I. and others (2005), "Course in theoretical mechanics", Moscow MGTU, p.736

Dube M., Drolet F., Bloch J.-F., Daneault C., Mangin P. (2007), "Tack, ink transfer and ink distribution on paper", IARIGAI vol. XXXIV, pages 23-30

Klaulitz T., Dorsam E. (2008), "Highly accurate material characterization of paper for the simulation of the printing processes", IARIGAI vol. XXXV, pages 263-270

Laurette Vieille-Grosjean, Alice Vermeulin, (2012) "Development of a comparative tool to predict ink misting" Advances in printing and media technology – printing techniques, pages 35-42

McPhee J., (1998), "Fundamentals of lithographic printing", Volume 1: Mechanism of printing

Mueller. P. (1988), "Offset printing", Moscow Kniga, p.208

Ozaki Y., Fujisawa N., Kawamura E. (2008), "Evaluation of the ink transfers of prints using low-angle shaving system", IARIGAI vol. XXXV, pages 189-194

Ruemer Klein, Patrick Cunningham, Gerd Meder, Martina Miletic (2012), "On developing methods for the identification of relevant paper properties", JPMTR 1, pages 25-39

Savarmand S., Bousfield D., Durand R., Warren R. (2010), "A laboratory method to characterize and predict ink misting", IARIGAI vol. XXXVII, pages 235-241

Tikhonov V.P., Gulyaev S.A., Semenyuta S.S. (1993), "About break layer of viscoplastic fluid between the two surfaces", Colloid Journal, v.55 № 4, p.104 – 109

Vygodsky M.J. (2010), "Handbook of mathematics", Moscow AST, Atsrel, p.1055

Yakhnin E.D. (1987), "Deformation, destruction and redistribution of fluid between the layers of the surfaces of solids and spreading the success of colloid chemistry", FAN Tashkent Uzbek SSR, p.110

42nd International Research Conference, 06 to 09 September 2015



Session 2A

Printed functionality 1

Monday, 7 September 2015 14:00 – 15:40

Chair: Wolfgang Faigle

All Inkjet-Printed transparent Piezoelectric Polymer Actuators for Microfluidic Lab-on-a-Chip Systems

Oliver Pabst^{1,2}, Maximilian Reif², Erik Beckert², Ramona Eberhardt², Andreas Tünnermann^{1,2}

¹ Institute of Applied Physics, Abbe Center of Photonics (ACP), Friedrich Schiller University Jena, Max-Wien-Platz 1, D-07743 Jena, Germany

² Fraunhofer Institute for Applied Optics and Precision Engineering (IOF), Albert-Einstein-Str. 7, D-07745 Jena, Germany

E-mail: oliver.pabst@iof.fraunhofer.de

Short Abstract

Fully inkjet-printed, transparent piezoelectric polymer actuators for pumping tasks in microfluidic labon-a-chip systems (LOC) are presented. The actuators are employed as the active element of a chipintegrated pump for LOC systems. Target applications of the devices are disposable LOC systems for point-of-care analysis tasks. Due to the single-use character, cost-effective manufacturing is essential. The actuators are created by inkjet printing a stack of two PEDOT:PSS electrodes and a piezoelectric P(VDF-TrFE) film on a passive polymer substrate. Maximum process temperatures of 130 °C are employed, which is compatible with typical substrate materials used in LOC systems. Different from most current micropump approaches, the direct printing approach does not require an additional hybrid joining step to mount the actuator on the pump membrane. Based on previous work on piezoelectric polymer actuators with silver electrodes, a modified process flow is introduced to realize a fully inkjetprinted and transparent actuator setup. The optical and ferroelectric behavior of the actuators as well as the performance in a micropump demonstrator is investigated. Compared to previous work, the demonstrator exhibits higher pump rates and backpressure and furthermore allows optical detection through the device. The manufacturing approach presented here represents a cost-effective alternative to conventional, lithography-based processes and can be combined with further printed functionalities in the future.

Keywords: piezoelectric polymers, P(VDF-TrFE), inkjet printing, lab-on-a-chip systems, micropump

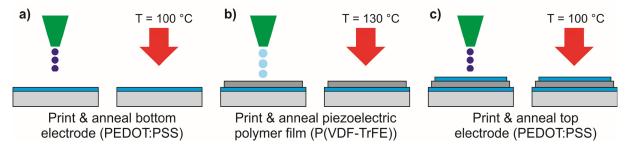
1. Introduction and Background

Microfluidic lab-on-a-chip systems (LOC) are systems that can be applied in life science applications for point-of-care analyses. The systems integrate chemical or biological reactions on compact chips that would typically require large and bulky equipment when running conventional lab analyses. Reactions can be performed using only small sample volumes. Typical applications of LOC systems include blood tests and water or food quality tests (Gervais et al., 2011). In order to avoid crosscontamination and to ensure sterile analysis conditions, the use of disposable, single-use systems is often desired. Therefore, cost-effective manufacturing approaches are required. Polymer replication processes based on injection molding or hot embossing are well-developed and are used for disposable LOC systems (Becker and Gärtner, 2008). Typical chip layouts include fluid channels, reaction chambers as well as mixers or other structures. The replicated polymer chips are sealed by a polymer cover foil. Despite the low manufacturing cost of the chip itself, additional functionalities like temperature control, fluid transport and optical detection are typically placed off-chip, which makes the systems more bulky. For direct function integration on the cover foil, the use of conventional, lithography-based manufacturing is economically not feasible for disposable devices. The use of digital printing techniques represents a promising alternative for cost-effective on-chip function integration. Drop-on-demand inkjet techniques can be used to deposit solutions or dispersions of functional materials in an additive, mask-less and non-contact way. Processing can be performed at low temperatures and is thus compatible with polymer materials that are used in LOC systems. Using inkjet printing, different functionalities have been demonstrated, including organic sources, RFID antennas, organic photodiodes and different sensor types (Korvink et al., 2012).

Controlled fluid distribution by pumping is a key element in most LOC systems (Zhang et al., 2007). Different manufacturing approaches for miniaturized pumps that are suitable for LOC systems have been reported (Iverson and Garimella, 2008). A common principle for such pumps are membrane pumps which generate a directed flow due to a volume change that is induced by a deflected membrane in combination with valves (Olsson, 1998). The actuators that generate the volume change are, however, in most cases manufactured separately and mounted on the pump membrane in a separate joining step. Most commonly, lead zirconate titanate (PZT) is used as actuator material. In this contribution, fully inkjet-printed piezoelectric polymer actuators based on P(VDF-TrFE) (poly(vinylidene fluoride-co-trifluoroethylene)) copolymers are presented. The actuators are printed directly on the cover foil of a microfluidic chip and thus do not require a separate joining step. P(VDF-TrFE) is suitable for solution-based deposition and has been used for printed sensor networks (Zirkl et al., 2011) or non-volatile memory elements (Mao et al., 2010). Recently, we reported fully printed P(VDF-TrFE) actuators with silver electrodes and demonstrated a working micropump setup as a proof-of-concept (Pabst et al., 2013, Pabst et al., 2014).

Different from PZT actuators, piezoelectric polymers can be processed at low temperatures that are compatible with polymer substrates and thus enable the direct integration on polymer-based microfluidic chips e.g. by printing processes. This is the most important benefit of employing piezoelectric polymers instead of PZT. P(VDF-TrFE) exhibits a lower piezoelectric d_{31} coefficient of approximately 10 pm V⁻¹ compared to 110 pm V⁻¹ for PZT (Seo and Zou, 1995). Furthermore, it requires larger driving fields of approximately 40 V μ m⁻¹ instead of 2 V μ m⁻¹ for PZT (Li and Cheng, 2009). However, due to the lower Young's modulus of P(VDF-TrFE) of 2 GPa compared to PZT with approximately 80 GPa, large deflections can be generated by piezoelectric polymer actuators when using polymer substrates.

In this work, electrically conductive PEDOT:PSS (poly(3,4-ethylenedioxythiophene)) polymers are used as an electrode material instead of silver. This leads to transparent actuators that enable optical detection through the actuator in LOC applications. Furthermore, the low Young's modulus of the polymer electrodes compared to silver leads to larger deflections of the actuators and thus higher pump rates.



2. Materials and Methods

Figure 1: Process flow employed for manufacturing transparent piezoelectric polymer actuators. A three-layer setup of a piezoelectric P(VDF-TrFE) film between two PEDOT:PSS electrodes is printed and annealed subsequently on top of a passive PET substrate.

Figure 1 shows the basic process flow that was used to print transparent actuators. Poly(ethylene terephthalate) films (PET, Goodfellow GmbH, Germany, 125 μ m thickness) were used as substrates. Bottom electrodes were printed using commercially available dispersions of electrically conductive PEDOT:PSS (Heraeus Precious Metals GmbH & Co. KG, Germany, Clevios P Jet HCv2) and a compact inkjet deposition system (Omnijet 100, Unijet Co., Ltd., South Korea) (a). Printing was performed using cartridges with 10 pL nominal droplet volume (DMC-11610, Dimatix-Fujifilm Inc., USA). After printing, the films were annealed at a temperature of 100 °C for one hour to remove remaining solvents. In a second step (b), an approximately 9 μ m thick P(VDF-TrFE) film was printed

using a 2 wt% solution of P(VDF-TrFE) (VDF:TrFE ratio 70:30 wt%, Solvay Specialty Polymers Italy S.p.A.) in cyclopentanone. Printheads with a droplet volume of 80 pL were used for printing the P(VDF-TrFE) film (Dimatix Nova 256/80). After printing, thermal annealing was performed at 130 °C for 24 hours to increase the crystallinity of the film and to remove remaining solvents. On top of the piezoelectric film, a PEDOT:PSS top electrode was printed and annealed similar to the bottom electrode (c).

Figure 2 shows photographs of fully processed actuators. It can be seen that the actuators are transparent but show a certain degree of light scattering (a). In order to realize membranes that are fixed around their perimeter, the actuators are fixed on aluminum mounts (b). Prior to characterization, the actuators were poled electrically at room temperature by applying a DC voltage of 600 V across the electrodes for one minute.

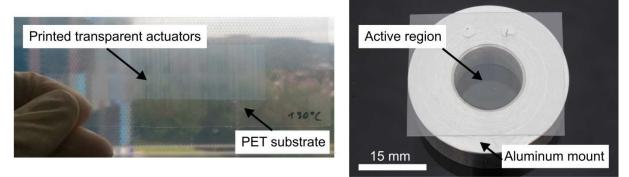


Figure 2: Photographs of printed transparent P(VDF-TrFE) actuators. PET substrate with printed actuator structures (left) and circular actuator fixed on an aluminum mount for characterization (right).

3. Results and Discussion

The as-prepared actuators were characterized concerning their optical properties, their ferroelectric behavior and their performance in a micropump demonstrator. Figure 3 shows transmission and reflection spectra that were recorded from the actuator setup of PET substrate and printed films in a wavelength range from 300 nm to 1200 nm. The measurement was performed in a Lambda 950 UV/VIS Spectrometer (Perkin Elmer Inc., USA) at an incident angle of 6°.

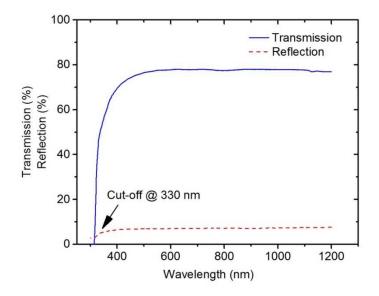


Figure 3: Transmission and reflection spectra measured from printed actuator on a PET substrate.

Below a cut-off wavelength of 300 nm, the actuators are non-transparent. With increasing wavelength the transmission increases and reaches a plateau of more than 75% for wavelengths between 460 nm and 1200 nm. A maximum reflection of 7.6% was measured, which indicates absorption and scattering of approximately 15.5%. The transmission and reflection of the PET substrate without any printed layers were measured to be 88% and 10.6%, respectively. It can be stated that the optical properties of the printed actuator setup allow an optical detection through the device due to its high transmission in general and the almost constant transmission between 500 nm and 1200 nm, which is a typical range for the evaluation of on-chip reactions.

The ferroelectric behavior of the actuators was characterized by measuring ferroelectric hysteresis loops using a commercial characterization setup (EasyCheck 300, Aixacct GmbH, Germany) and a high-voltage amplifier (2220CE, Trek Inc., USA). Measurements were repeated three times at a frequency of 0.05 Hz. Figure 4 shows measurement curves that were recorded when using maximum voltages of 400 V and 800 V, respectively. A driving voltage of 800 V corresponds to an electric field of approximately 89 kV mm⁻¹ and leads to distinct hysteresis behavior with polarisation saturation for large voltages and a remanent polarization P_{rem} of 4.4 μ C cm⁻², which is in a similar range than observed in previous investigations with P(VDF-TrFE) actuators with silver electrodes (Pabst et al., 2014).

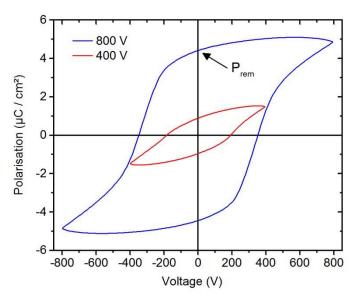


Figure 4: Examples of ferroelectric hysteresis loops measured from one printed actuator.

For the application in a micropump, circular actuators were prepared and combined with a metallic pump substrate. Figure 5 shows actuator elements on a PET substrate (a) as well as an assembled pump demonstrator (b). In this stage of the experiments, a milled pump substrate made from an aluminum alloy was used. However, the design is suitable for future manufacturing by polymer replication processes.

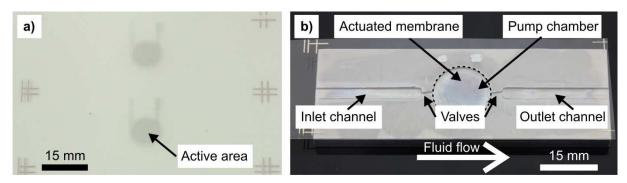


Figure 5: (a) PET substrate with printed actuator elements. (b) Assembled pump demonstrator, consisting of a metallic pump substrate and an actuated PET substrate that seals the chip.

The pump substrate includes a pump chamber with a diameter of 15 mm, inlet and outlet channels and passive diffuser valves. The design of the valves is based on typical geometries for passive valves (Olsson, 1998). The PET substrate seals the channels and includes a membrane actuator. When a cyclic voltage is applied, a volume change is induced under the deflected membrane, which leads to a directed fluid flow in combination with the valves. The performance of the micropump demonstrator was evaluated based on pump rate and backpressure measurements. The actuators were driven with quasi-static rectangular driving voltages of up to 800 V at a frequency of 30 Hz. Isopropanol was used as a pumping medium. The pump rate was calculated based on image processing from videos that were recorded during operation of the pump. The backpressure was estimated by placing the demonstrator setup on a rotation stage, setting different angles and calculating the angle-dependent normal force component. The results are displayed in Figure 6. An approximately linear dependency of pump rate on backpressure is found with maximum values for pump rate and backpressure of 135 μ L min⁻¹ and 78 Pa, respectively.

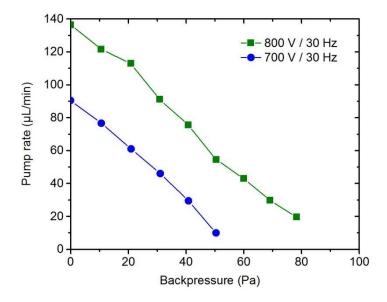


Figure 6: Measurement of pump rate vs. backpressure for different electric driving conditions.

Compared to previous results with an identical design of the pump chamber but an actuator with silver electrodes, significantly larger pump rates (135 μ L min⁻¹ instead of 105 μ L min⁻¹) and backpressures (78 Pa instead of 42 Pa) were obtained when using identical driving conditions. This is attributed to the low Young's modulus of the polymer electrodes (1 GPa, Greco et al., 2011), which hinders the actuator movement less than in the case of silver electrodes. In general, the pump rates are suitable for LOC applications (Zhang et al., 2007) and show the potential of the suggested manufacturing approach.

4. Conclusions

In this contribution, fully inkjet-printed, transparent piezoelectric polymer actuators were presented. Target applications of the devices are chip-integrated membrane pumps for disposable, polymer-based lab-on-a-chip systems. Different from most current micropump approaches, the direct printing approach does not require an additional joining step to mount the actuator on the pump membrane. Based on previous work on piezoelectric polymer actuators with silver electrodes, a modified process flow was introduced to realize a fully inkjet-printed and transparent actuator setup. PEDOT:PSS was used as an electrode material. This conductive polymer exhibits a lower Young's modulus than metallic electrodes and thus leads to higher actuator deflection compared to elements with silver electrodes. This was confirmed by liquid flow rate measurements in a pump demonstrator. Pump rates of up to $135 \,\mu L \,min^{-1}$ were realized, which is suitable for applications in LOC systems. The transmission spectrum of the devices was analyzed and found to be suitable for optical detection through the printed layer stack. In the future it is planned to combine the actuators with further printed on-chip functionalities like heating elements or optical excitation and detection to realize more complex yet cost-effective LOC systems.

Acknowledgments

The work presented in this paper was funded by the German Federal Ministry of Education and Research (BMBF) within the joint research project "Komplexer Optofluidchip" (FKZ 03IPT609A) and by the German Research Foundation (DFG) within the priority program "Active Micro-optics" (SPP 1337). The authors thank Johannes Jobst (previously Fraunhofer IOF) for performing transmission and reflection measurements.

References

Becker, H. and Gärtner, C., 2008. Polymer microfabrication technologies for microfluidic systems. *Anal. Bioanal. Chem.* 390, pp. 89–111

Carpi, F., Smela, E., Li, Z. and Cheng., Z., 2009. Piezoelectric and electrostrictive polymer actuators: Fundamentals, in: (Eds.), *Biomedical Applications of Electroactive Polymer Actuators*, John Wiley & Sons, Ltd, pp. 317–334

Gervais, L., de Rooij, N. and Delamarche, E., 2011. Microfluidic chips for point-of-care immunodiagnostics. *Adv. Mater.* 23(24), pp. H151–H176

Greco, F., Zucca, A., Taccola, S., Menciassi, A., Fujie, T., Haniuda, H., Takeoka, S., Dario, P. and Mattoli, V., 2011. Ultra-thin conductive free-standing PEDOT/PSS nanofilms. *Soft Matter* 7, pp. 10642–10650

Iverson, B. and Garimella, S., 2008. Recent advances in microscale pumping technologies: a review and evaluation. *Microfluid*. *Nanofluid*. 5(2), pp. 145–174

Korvink, J. G., Smith, P. J. and Shin, D.-Y. (eds), 2012. Inkjet-Based Micromanufacturing. Wiley-VCH Verlag GmbH & Co. KGaA

Mao, D., Quevedo-Lopez, M., Stiegler, H., Gnade, B. and Alshareef, H., 2010. Optimization of poly(vinylidene fluoride-trifluoroethylene) films as non-volatile memory for flexible electronics. *Org. Electron.* 11(5), pp. 925–932

Olsson, A., 1998. Valve-less Diffuser Micropumps, PhD thesis, Royal Institute of Technology Stockholm, Sweden

Pabst, O., Hölzer, S., Beckert, E., Perelaer, J., Schubert, U. S., Eberhardt, R. and Tünnermann, A., 2014. Inkjet printed micropump actuator based on piezoelectric polymers: Device performance and morphology studies. *Org. Electron*. 15(11), pp. 3306–3315

Pabst, O., Perelaer, J., Beckert, E., Schubert, U. S., Eberhardt, R. and Tünnermann, A., 2013. All inkjet-printed piezoelectric polymer actuators: Characterization and applications for micropumps in lab-on-a-chip systems. *Org. Electron.* 14(12), pp. 3423–3429

Seo, I. and Zou, D., 1995. Electromechanical applications, in: H. S. Nalwa (Ed.), *Ferroelectric polymers: chemistry, physics, and applications*, Marcel Dekker, Inc., pp. 699–734

Zhang, C., Xing, D. and Li, Y., 2007. Micropumps, microvalves, and micromixers within PCR microfluidic chips: Advances and trends. *Biotechnol. Adv.* 25(5), pp. 483–514

Zirkl, M., Sawatdee, A., Helbig, U., Krause, M., Scheipl, G., Kraker, E., Ersman, P. A., Nilsson, D., Platt, D., Bodö, P., Bauer, S., Domann, G. and Stadlober, B., 2011. An all-printed ferroelectric active matrix sensor network based on only five functional materials forming a touchless control interface. *Adv. Mater.* 23(18), pp. 1–6

Inkjet-Printed Reaction Arrays on Pigment Coated Substrates

Risto Koivunen¹, Eveliina Jutila¹, Roger Bollström², Patrick Gane^{1,2}

¹ Aalto University, School of Chemical Technology, Department of Forest Products Technology, PL 16400, 00076 Aalto, Finland
 ² Omya International AG, Baslerstrasse 42, CH-4665 Oftringen, Switzerland

E-mail: risto.koivunen@aalto.fi; eveliina.jutila@aalto.fi; roger.bollstroem@omya.com; patrick.gane@omya.com

Corresponding author: Risto Koivunen, Tel.: +358 50 4061259, Fax: +358 9 47124259.

Short Abstract

Paperfluidic devices are microfluidic devices fabricated from naturally porous material. This study investigates the hydrophobic patterning by functional inkjet printing of custom-designed coatings for application in paperfluidic devices. The patterning is exemplified by reaction arrays in the form of test cells in which aqueous reactants can be contained. Coatings consisted of porous functionalised calcium carbonate (FCC) pigment, which was combined with binders to provide cohesion and adhesion. The inks for patterning contained polystyrene (PS) or alkyl ketene dimer (AKD) in solvent. With PS ink, hydrophobic barriers could be produced on all tested pigment coatings, although generally requiring printing of multiple layers. With AKD ink, hydrophobic patterns could be produced on coatings with inorganic sodium silicate as sole binder. Barriers could be produced with fewer printed layers of AKD ink compared with PS ink. The hydrophobising agent amount needed to form a waterproof barrier depended on the properties of the coatings. Compared to cellulose filter papers, larger amounts of hydrophobising agent are required for hydrophobic patterning of these pigment coatings. This may be explained by the higher specific surface area and the very hydrophilic nature of the FCC pigment coating.

Keywords: paperfluidic, functional printing, functional pigment coating, functionalised calcium carbonate, hydrophobic ink.

1. Introduction and background

Paperfluidic devices are microfluidic devices fabricated out of paper or paper-like porous material. The porous nature allows the device to absorb and transport liquids by capillary action. In design, the devices can range from simple test station arrays resembling microzone well plates (Carrilho, *et al.*, 2009) to multi-step devices featuring channel networks. Paperfluidic devices provide a promising platform for cheap, simple and easily disposable diagnostic tools. Possible applications for such diagnostics include medical assays, such as detection of blood type (Li, *et al.*, 2012) or sickle cell disease (Yang, *et al.*, 2013), monitoring of nitrite levels in saliva (Klasner, *et al.*, 2010) or measuring glucose levels in urine (Cassano and Fan, 2013). Other possible applications include detection of bacteria in food (Jokerst, *et al.*, 2012) or water (Hossain, *et al.*, 2012), as well as detection of heavy metals (Feng, *et al.*, 2013) or reactive phosphates (Jayawardane, Kelvie and Kolev, 2012) in water. Paperfluidic devices are also expected to provide a platform for pharmaceutical research (Dhen, Kuo and Cheng, 2015).

Liquid transport in paperfluidic devices is propelled by capillary pressure in the hydrophilic pore matrix, with hydrophobic barriers applied to limit and direct the flow. These barriers are fabricated by treating the pore surfaces on selected areas of the substrate with hydrophobic material. Microfabrication methods, such as photolithography (Martinez, *et al.*, 2007), and various printing methods have been demonstrated as viable means for patterning such barriers. The most common printing method for hydrophobic patterns has been inkjet with a variety of hydrophobic materials, such as hot-melt wax (Carrilho, Martinez and Whitesides, 2009), solvent-diluted polydimethylsiloxane

(Määttänen, et al., 2011) and acrylic polymer in solvent-based solution (Apilux, et al., 2013). Key properties of a hydrophobising ink are the ability to penetrate the full depth of the porous layer and the ability to transform pore surface chemistry from hydrophilic to hydrophobic.

To date, most studies on paperfluidic devices have employed commercial cellulose-based chromatography papers (Martinez, *et al.*, 2007) or filter papers (Li, *et al.*, 2010), or custom-made laboratory handsheets (Böhm, *et al.*, 2014), as base substrates. Another successful cellulose-derived substrate is nitrocellulose membrane (Apilux, *et al.*, 2013). However, pigment paper coatings as alternative base substrates have received no significant interest. Commercial coated papers are not designed with paperfluidic applications in mind, though they have been hydrophobically patterned for application as reaction arrays (Määttänen, *et al.*, 2011). Custom pigment coatings, isolated from base paper by a barrier coating (Bollström, *et al.*, 2013), as shown in Figure 1, can provide new paperfluidic substrates.

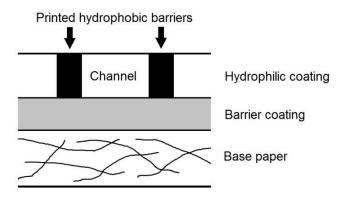


Figure 1. Schematic cross-section of a multilayer coated paper with printed hydrophobic pattern.

This study investigates hydrophobic patterning of novel custom pigmented coatings, designed for paperfluidic applications. Such a substrate design provides numerous advantages compared to plain cellulose paper. Firstly, the spatial dimensions of pigment particles are smaller than those of cellulose fibres, providing potentially a better printing resolution. Secondly, the thickness of the functional coating layer can be varied freely. Thirdly, if a barrier layer or an impermeable substrate is employed, the underside of the device is not open to the environment, allowing it to be placed on a surface while containing liquid. Base paper is also isolated from liquids in the top-coating by the barrier layer. Fourthly, new surface chemistry can be provided by pigments, possibly helping with binding of reagents or separation of components in the sample.

In the present work, hydrophobic patterning of novel specialised coatings, consisting of functionalised calcium carbonate (FCC) and various binders, is carried out by inkjet printing of custom-made inks consisting of polystyrene (PS) or paper sizing agent alkyl ketene dimer (AKD) dissolved in organic solvent. These inks have been shown to be effective for hydrophobic patterning of filter papers (Koivunen, Jutila and Gane, 2015). AKD-based inkjet inks have been applied in hydrophobic patterning also by other authors, either dissolved into organic solvent (Li, *et al.*, 2010) or as an aqueous emulsion (Wang, *et al.*, 2014). PS-based inks have been printed with flexography (Olkkonen, Lehtinen and Erho, 2011) and screen printing (Sameenoi, *et al.*, 2014). The main focus of the work reported is on determining print settings required for successful hydrophobisation of the custom pigment coatings.

2. Materials and Methods

2.1 Functional pigment coatings

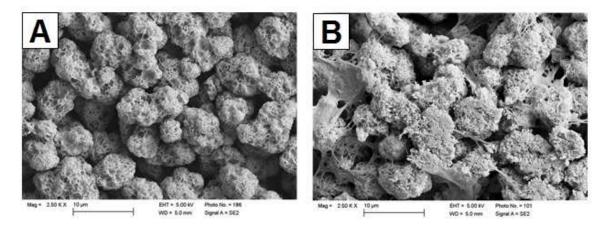


Figure 2. SEM images of FCC particles: (A) plain, (B) combined with micro-fibrillated cellulose (MFC) Arbocel MF-40-7 as binder (right).

Binders tested with FCC included two micro-fibrillated cellulose products Arbocel MF-40-7 (J. Rettenmaier & Söhne GmbH + Co KG) and 22.3462 CMCX-TYPE (Omya International AG), polyvinyl alcohol BF05 (Omya International AG), styrene acrylic latex Acronal S 728 (BASF) and sodium silicate (Merck KGaA, product number 1056212500). Coating colour formulations are shown in Table 1.

Abbreviation	Pigment	Binder	Binder amount	Solids content
			/ pph*	/ w/w %
FCC + MFC A	FCC	micro-fibrillated cellulose	20	11.3
		(Arbocel MF-40-7)		
FCC + MFC B	FCC	micro-fibrillated cellulose	20	4.4
		(22.3462 CMCX-TYPE)		
FCC + PVOH	FCC	polyvinyl alcohol	10	14.3
FCC + latex	FCC	styrene acrylic latex	10	10.0
FCC + sodium	FCC	sodium silicate	50	19.2
silicate				

Tahle	1	Coating	colour	formu	lations
rabie	1.	Couing	colour	jormu	anons.

*pph: parts by weight per 100 parts by weight of pigment

Coating colours were applied with a K202 Control Coater (RK PrintCoat Instruments Ltd.) employing the blue labelled wire-wound rod, applying a 100 μ m thick wet layer with a speed setting of 6 m·min⁻¹. Substituting for barrier coated paper, sheets of impermeable SuperYUPO[®] (Yupo Corporation) pigment filled polypropylene film, of 80 μ m thickness and 62 gm⁻² basis weight, were used as a base substrate for coatings.

2.2 Hydrophobising inks and printing

Hydrophobic inks consisted of 5.0 % by weight hydrophobising agent; either 35 kDa molecular weight polystyrene (Sigma-Aldrich, product code 331651) or solid alkyl ketene dimer, Basoplast 88 (BASF), and 0.1 % by weight Sudan Red G colorant (Sigma-Aldrich, product code 17373) dissolved in p-

xylene solvent (VWR, product code 28984.292). These two inks will be referred to as PS ink and AKD ink.

Inks were applied with a DMP-2831 inkjet printer (Fujifilm Dimatix) with DMC-11610 ink cartridges with 10 pl nominal drop volume. The printer mounting platen and print head were heated to 30 °C. Printing was carried out with all 16 available nozzles. Custom waveforms were designed for the inks.

Samples printed with PS ink were ready immediately after printing. Samples printed with AKD ink were heated for 10 min on a 100 °C hot plate for AKD to react with the hydroxyl groups in the binder.

2.3 Other materials and equipment

SEM micrographs of coatings were obtained with a Sigma VP field emission scanning electron microscope (Carl Zeiss AG). Coating dry thickness was measured with an SE250D micrometer (Lorentzen & Wettre) with a 2 cm² measurement area spindle and measurement pressure of 100 kPa.

For testing the integrity of the hydrophobic patterns, deionised water was dyed with 0.1 % by weight amaranth red (Fluka, product code 06409). Alternatively, aqueous solutions containing 15, 20, 25, 30 or 40 % by weight ethanol and 0.1 % by weight rhodamine B (Kremer Pigmente GmbH & Co. KG) were applied as lower surface tension liquids.

2.4 Test pattern

The amount of hydrophobic ink required to produce effective barriers on the substrates was determined with printed test patterns consisting of rings with nominal 5 mm inner and 9 mm outer diameter. These patterns were printed on the substrates with drop spacing values of 10, 15 and 20 μ m, being equivalent to nominal applied ink layers of 100, 44 and 25 cm³m⁻², respectively. Hydrophobising material quantity was varied by printing patterns with 1-5 layers of PS ink and 1-2 layers of AKD ink.

For each tested combination of ink, substrate, drop spacing and number of ink layers, 10 rings were printed. Barrier properties of the rings were tested by placing a 10 μ l drop of dyed water or ethanol solution inside each ring. Once this drop had dried, the sample was visually evaluated to determine if the liquid had been fully contained within the ring interior. If the liquid had been contained within all 10 rings tested, then that particular setting combination was considered to produce reliable barriers. However, if any of the 10 tested rings displayed any leaking from the ring interior to the printed area or beyond, then the printing combination was disqualified. An example of the test pattern, showing both holding and leaking rings, is shown in Figure 3.

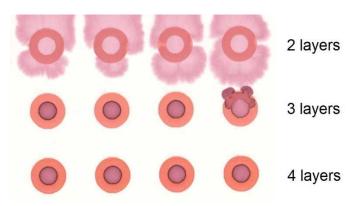


Figure 3. An example of the test pattern, in this case printed with PS ink on FCC + MFC B coated substrate, with drop spacing of 20 µm and 2, 3 and 4 ink layers, respectively.

3. Results and Discussion

3.1 Coating properties

The measured coating properties, in respect to coat weight and thickness, are listed in Table 2.

Coating	Basis weight	Thickness
	/ gm ⁻²	/ μm
FCC + MFC A	14.0 ± 1.3	59 ± 2
FCC + MFC B	4.1 ± 1.4	27 ± 2
FCC + PVOH	17.9 ± 0.8	70 ± 2
FCC + latex	11.8 ± 0.5	60 ± 2
FCC + sodium silicate	22.2 ± 1.7	76 ± 6

Table 2: Measured coating properties.

3.2 Hydrophobic patterning with polystyrene

With PS ink, reliable hydrophobic barriers, such that no leakage outside the inner perimeter was detected, could be produced on all coatings. The number of printed layers required to form those barriers on different substrates with PS ink is listed in Table 3.

Table 3: Number of printed layers needed to produce effective barriers with PS ink. "Does not hold" indicates no reliable barrier could be achieved with the maximum of 5 printed layers.

Coating	Drop spacing		
	10 <i>µ</i> m	15 µm	20 µm
FCC + MFC A	2 layers	5 layers	Not tested
FCC + MFC B	1 layer	2 layers	4 layers
FCC + PVOH	3 layers	Does not hold	Not tested
FCC + latex	1 layer	1 layer	Does not hold
FCC + sodium silicate	3 layers	Does not hold	Not tested

As can be seen in Table 3, with 10 μ m drop spacing, providing the largest tested ink volume per unit printed area, reliable barriers could be achieved with PS ink on all of the coatings, though tending to require multiple printed layers. With 15 μ m drop spacing, reliable barriers could be achieved with three of the coatings. The FCC + latex coating exceptionally could be hydrophobised at this drop spacing with as little as a single ink layer. This is likely due to the reduced pore connectivity and the latex itself making the coating less hydrophilic than other binders.

With 20 μ m drop spacing, PS ink could produce reliable barriers only on the FCC + MFC B coating. This is presumably due to the thinness of the dry coating layer, in which full depth penetration was achievable with a lower ink volume.

For comparison, in an earlier study with the same ink (Koivunen, Jutila and Gane, 2015) on cotton cellulose filter paper Whatman 4 (GE Healthcare), hydrophobisation with 10 or 15 μ m drop spacing could be achieved with a single ink layer. Examples of patterns printed with PS ink on a pigment coated substrate and a filter paper are shown in Figure 4.

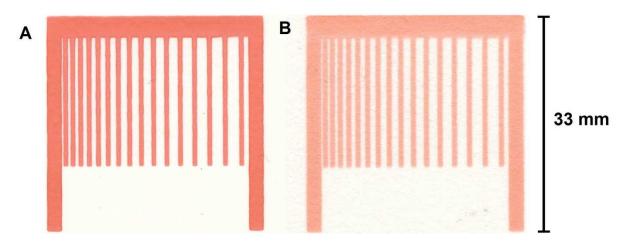


Figure 4. An example channel pattern, printed with PS ink on: (A) FCC + MFC B coating with 20 μm drop spacing and 4 ink layers, (B) Whatman 4 filter paper with 15 μm drop spacing and 1 ink layer.

3.3 Hydrophobic patterning with AKD

In the case of the AKD ink, reliable barriers could be produced on all coatings, except on the one with sodium silicate as binder. The number of printed layers required to form reliable hydrophobic barriers on the various substrates with AKD ink is listed in Table 4.

Table 4: Number of printed layers needed to produce effective barriers with AKD ink. "Does not hold" indicates no reliable barrier could be achieved with the maximum of 2 printed layers.

Coating	Drop spacing		
	10 µm	15 µm	20 µm
FCC + MFC A	1 layer	1 layer	2 layers
FCC + MFC B	1 layer	1 layer	1 layer
FCC + PVOH	1 layer	1 layer	1 layer
FCC + latex	1 layer	1 layer	1 layer
FCC + sodium silicate	Does not hold	Does not hold	Does not hold

Inability to produce effective barriers with sodium silicate bound coating can be explained by the hydrophobisation mechanism of AKD, which is based on a reaction with surface available hydroxyl groups. Neither FCC nor sodium silicate contains such groups.

With the other binders, AKD ink provided effective hydrophobisation with even the largest tested 20 μ m drop spacing, and lower number of ink layers compared to the PS ink. Improved performance at larger drop spacing values suggests that AKD ink, with lower viscosity, penetrates deeper into the substrate than PS ink printed with the same drop spacing. This would match the results achieved previously (Koivunen, Jutila and Gane, 2015) on filter papers. The fewer number of layers needed indicates that AKD is also more effective in hydrophobisation.

3.4 Barrier properties versus low surface tension liquids

In addition to water, two coatings were tested with low surface tension ethanol solutions. Selected coatings were FCC + MFC A, printed with AKD ink (2 layers with 20 μ m drop spacing), and FCC + MFC B, printed with PS ink (4 layers with 20 μ m drop spacing). The chosen print settings produced reliable hydrophobic barriers on these coatings in the previous tests. 10 rings were tested with each of the 5 ethanol solutions.

On FCC + MFC A printed with AKD, ethanol solutions up to 25 % by weight (surface tension = 37 mNm⁻¹) were held by the barriers, with 30 % ethanol solution leaking slightly. On FCC + MFC B printed with PS, ethanol solution of 15 % by weight (surface tension = 44 mNm⁻¹) was held, while 20 % ethanol solution leaked through. The differences between the tested samples are likely to be due to different surface chemistries produced by the two hydrophobising agents, rather than due to differences between MFC binders, suggesting that AKD ink produces in lower surface energy on pores.

4. Conclusions

This study demonstrates feasibility of printing hydrophobic patterning on coatings consisting of FCC combined with a variety of binders. However, such coatings can be more challenging to hydrophobise than conventional cellulose papers, as demonstrated by the cases where multiple layers of ink were needed to achieve reliable hydrophobisation of the coating. This may be explained by the higher specific surface area of the coating as compared to ordinary cellulose fibres and the very hydrophilic nature of the FCC.

While FCC has a highly hydrophilic surface chemistry, it does not solely dominate the nature of the coating. This becomes evident when observing the results achieved by the AKD ink, which, during curing, reacts with the hydroxyl groups in the organic binders but does not interact with the FCC. On the other hand, relatively hydrophobic latex as binder, which also is known to reduce pore connectivity, results in a coating that can be easily hydrophobised, even with the normally otherwise high volume demand needed with PS ink. Thus, the effectiveness of a hydrophobising agent in producing effective barriers on a custom pigment coating is dependent on combined surface chemistries of pigment, binder and hydrophobising agent.

Acknowledgments

Funding for the project was solely supported by Omya International AG. Silvan Fischer (Omya International AG) is acknowledged for the SEM analysis. The AKD sample was donated by Chemigate Oy.

References

Apilux, A., Ukita Y., Chikae, M., Chailapakul, O. and Takamura, Y., 2013. Development of automated paperbased devices for sequential multistep sandwich enzyme-linked immunosorbent assays using inkjet printing. *Lab on a Chip*, 13(1), pp. 126-135.

Bollström, R., Nyqvist, R., Preston, J., Salminen, P. and Toivakka, M., 2013. Barrier properties created by dispersion coating. TAPPI Journal, 12(4), pp. 45-51.

Böhm, A., Carstens, F., Trieb, C., Schabel, S. and Biesalski, M., 2014. Engineering microfluidic papers: effect of fiber source and paper sheet properties on capillary-driven flow. *Microfluidics and Nanofluidics*, 16(5), pp-789-799.

Carrilho, E., Martinez, A.W. and Whitesides, G.M., 2009. Understanding Wax Printing: A Simple Micropatterning Process for Paper-Based Microfluidics. *Analytical Chemistry*, 81(16), pp. 7091-7096.

Carrilho, E., Phillips, S.T., Vella, S.J., Martinez, A.W. and Whitesides, G.M., 2009. Paper Microzone Plates. *Analytical Chemistry*, 81(15), pp. 5990-5998.

Cassano, C.L. and Fan, Z.H., 2013. Laminated paper-based analytical devices (LPAD): fabrication, characterisation, and assays. *Microfluidics and Nanofluidics*, 15(2), pp. 173-181.

Chen, Y., Kuo, Z. and Cheng C., 2015. Paper – a potential platform in pharmaceutical development. *Trends in Biotechnology*, 33(1), pp. 4-9.

Feng, L., Li, H., Niu, L-Y., Guan, Y., Duan, C., Guan, Y-F., Tung, C. and Yang, Q., 2013. A fluorometric paperbased sensor array for the discrimination of heavy-metal ions. *Talanta*, 108(1), pp. 103-108.

Hossain, S.M.Z., Ozimok, C., Sicard, C., Aguirre, S.D., Ali, M.M., Li, Y. and Brennan, J.D., 2012. Multiplexed paper test strip for quantitative bacterial detection. *Analytical and Bioanalytical Chemistry*, 403(6), pp. 1567-1576.

Jayawardane, B.M., McKelvie, I.D. and Kolev, S.D., 2012. A paper-based device for measurement of reactive phosphate in water. *Talanta*, 100(1), pp. 454-460.

Jokerst, J.C., Adkins, J.A., Bisha, B., Mentele, M.M., Goodridge, L.D. and C.S. Henry, C.S., 2012. Development of a Paper-Based Analytical Device for Colorimetric Detection of Select Foodborne Pathogens. *Analytical Chemistry*, 84(6), pp. 2900-2907.

Klasner, S.A., Price, A.K., Hoeman, K.W., Wilson, R.S., Bell, K.J. and Culbertson, C.T., 2010. Paper-based microfluidic devices for analysis of clinically relevant analytes present in urine and saliva. *Analytical and Bioanalytical Chemistry*, 397(5), pp. 1821-1829.

Koivunen, R., Jutila. E. and Gane, P.A.C., 2015. Inkjet printed hydrophobic microfluidic channelling on porous substrates. *Journal of Print and Media Technology Research*, 4(1), pp. 7-17.

Li, M., Junfei, T., Al-Tamimi, M. and Wei, S., 2012. Paper-Based Blood Typing Device That Reports Patient's Blood Type "in Writing". *Angewandte Chemie international edition*, 51(22), pp. 5497-5501.

Li, X., Tian, J., Garnier, G. and Shen, W., 2010. Fabrication of paper-based microfluidic sensors by printing. *Colloids and Surfaces B: Biointerfaces*, 76(2), pp. 564-570.

Määttänen, A., Fors, D., Wang, S., Valtakari, D., Ihalainen, P. and Peltonen, J., 2011. Paper-based planar reaction arrays for printed diagnostics. Sensors and Actuators B: Chemical, 160(1), pp. 1404–1412.

Martinez, A.W., Butte, M.J., Whitesides, G.M. and Phillips, S.T., 2007. Patterned Paper as Platform for Inexpensive, Low-Volume, Portable Bioassays. *Angewandte Chemie international edition*, 46(8), pp. 1318-1320.

Olkkonen, J., Lehtinen, K. and Erho, T., 2011. Flexographically Printed Fluidic Structures in Paper. *Analytical Chemistry*, 82(24), pp. 10246-10250.

Sameenoi, Y., Nongkai, P.N., Nouanthavong, S., Henry, C.S. and Napapricha D., 2014. One-step polymer screen-printing for microfluidic paper-based analytical device (µPAD) fabrication. *Analyst*, 139(24), pp. 6580-6588.

Yang, X., Kanter, J., Piety, N.Z., Benton, M.S., Vignes, S.M. and Shevkoplyas, S.S., 2013. A simple, rapid, low-cost diagnostic test for sickle cell disease. *Lab on a Chip*, 13(8), pp. 1464-1467.

Wang, J., Monton, M.R.N, Zhang, X., Filipe, C.D.M., Pelton, R. and Brennan, J.D., 2014. Hydrophobic sol-gel channel patterning strategies for paper-based microfluidics. *Lab on a Chip*, 14(4), pp. 691-695.

Improving the Electrical Performance and Mechanical Properties of Conductive Ink on thin compound Substrate

Andreas Willfahrt, Thomas Fischer, Gunter Hübner

Stuttgart Media University

E-mail: willfahrt@hdm-stuttgart.de; fischert@hdm-stuttgart.de; huebner@hdm-stuttgart.de

Short Abstract

In printed electronics applications there are several ink/paste and substrate combinations to choose from. Silver ink is usually used due to its high electrical conductivity. Carbon-black and PEDOT:PSS are also very common. Substrates are available in a broad variety. Flexibility, good adhesion of the ink, processability, and a maximum processing temperature compatible with the curing temperature of the functional inks is important. If barrier properties, e.g. against the permeation of oxygen carbon dioxide or water vapour are required, a compound substrate may be necessary, consisting of two or more layers of different materials. The motivation for this investigation was the need for improving the stability and processability of a given substrate chosen for printed batteries. The substrate consists of three layers, namely polyethylene (PE), aluminium (Al) and polyethylene terephthalate (PET). This compound foil is rather thin (100 µm) and very flexible. This is a major requirement for the application. The aluminium sandwiched between two polymer layers provides sufficient barrier properties. PET is commonly used as a substrate for printed electronic applications. PE is not as easy to print on, but with e.g. plasma treatment the adhesion of printing inks is sufficient. The weldability of PE is beneficial for the screen printed battery application, although poor printability without surface treatment and the thermal mismatch of the asymmetric polymer compound (PE – PET) renders processing rather difficult. In this work the authors examined a route for printing on PE without the need of pre-treatment of the substrate with plasma or corona. Instead, it was found that an UV-ink layer used as adhesion promoter provided sufficient adhesion and improved mechanical stability, i.e. cohesion of the successively printed silver ink layer. Additionally, the thermal treatment of the conductive ink was optimized by comparing heat press and hot stamp curing with batch oven curing.

Keywords: printed electronics, printed batteries, substrate, compound material

1. Introduction and background

The choice of substrate in the field of printed electronics is very important. If one considers organic light emitting diodes (OLED) or – as in the underlying case – printed batteries, it is necessary to ensure the encapsulation of the chemical system during manufacturing and lifetime of the device. Without the appropriate barrier materials, the lifetime is drastically lower. (Yoshida, 2006), (Park, 2011), (Bülow, 2014), (Wendler, 2010). Usually, the application determines the inks, the substrates, and also the printing method that can be deployed. Tradeoffs have to be made, if requirements are conflicting. The authors chose a compound laminate consisting of PE-Al-PET as substrate for printed batteries as part of a research project (KoSiF, 2013). The conflict in choosing this substrate lies in the favourable weldability and the poor printability of PE. Due to wettability reasons printing on PET is uncomplicated whereas printing on PE is quite difficult. Pre-treatment of the PE surface is necessary in order to achieve adhesion of the print layer on the polymer, (Lommatzsch, 2007). In roll-to-roll production lines corona or plasma devices can be implemented. However, if a sheet-fed lab machine is used and the available plasma/corona device has a limited spatial work area or if even no corona or plasma device is accessible at all, another approach to the problem may be useful.

One part of this paper is the examination of an ink layer that promotes the adhesion of silver ink on PE without prior application of corona or plasma treatment. A UV-curing printing ink is used as an adhesion agent, since the processability is very good as well as the adhesion of silver ink on top of the UV-ink layer. The second part of the paper concludes the examination of alternative thermal processes

that are capable of curing electrical conductive printing inks. The thermal treatment after printing is essential for optimizing the electrical conductivity. Two alternative routes to improve the adhesion and the mechanical stability of the successively printed inklayers on PE were investigated.

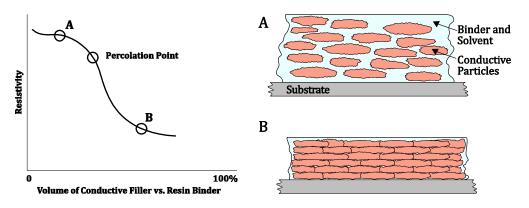


Figure 1: Pigment filled conductive inks need thermal treatment in order to establish electrical pathways between the single particles. Source: (Banfield, 2000)

The mechanism behind metal particle filled inks requires thermal treatment after printing (Banfield, 2000). A curing temperature of 120° C for at least 10 minutes is mostly stated as the minimum requirement for achieving a reasonable conductivity. The conductance, the reciprocal of the ohmic resistance, increases with higher temperature and longer curing time. It is usually the substrate which limits the maximum processing temperature, e.g. PET substrates do not withstand temperatures higher than 150° C. According to Figure 1, the evaporation of the solvent and the shrinkage of the binder reduce the height of the ink layer and thus increasing the contact area of the conductive particles. The continuous service temperature of some polymers, e.g. PE and PP, is below 100° C. Thus, the compound foil examined in this paper is very prone to thermal stress, the asymmetry (PE–PET) leads to a pronounced tendency to curling.

Since the composite film used for the investigation was provided on a roll it showed pronounced memory curl if cut in sheets for processing on a flatbed screen printing press. To prevent curling of the substrate, the compound films were laminated on a rigid carrier substrate which can withstand temperatures of 120° C for several minutes without deforming.

Substrate carrier	Adhesive	Adhesive pattern	
Cardboard	Water based dispersion	Full area	
Phenolic paper	Solvent based	Frame	
		Honeycomb	
Screen mesh	Sefar PET 1500 120-34, 77-48	Sefar PET 1500 120-34, 77-48	
UV ink	SunChemical SunTronic 680, Marabu Ultraswitch UVSW		
Silver ink	Acheson Electrodag® PM-406		

 Table 1: Preliminary tests for identifying the main problems of processing the PE-Al-PET compound foil.

1.1 Preliminary tests

In a preliminary test series the materials listed in Table 1 were examined. The substrate carrier supports the compound foil, which is laminated onto the carrier by printed adhesives. Two adhesives were printed in three different patterns. Without lamination to a carrier the compound foil curls immediately even when heated at low temperature, starting from 40-50° C. The honeycomb pattern showed good adhesion, but the pattern was visible in the image printed on the laminated compound foil. Rigid substrate carriers like phenolic paper improves the processability, whereas cardboard also tends to curl (Figure 3). Both adhesives performed similar and showed promising results. The solvent

based adhesive was chosen for further tests because of ease of use. Based on the results of the preliminary tests the examination of alternative thermal techniques gained more importance.



Figure 2: Effect of oven curing on the dimensional stability of the substrate. When adhesion was too weak, the samples pulled of and curled.

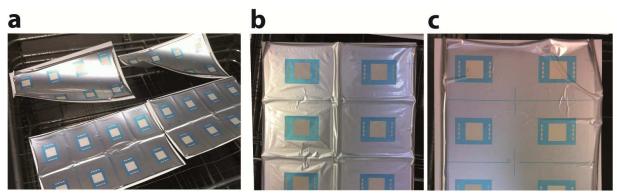


Figure 3: The full area adhesive sheets on cardboard are curling (background a, c), whereas the adhesive free parts of the frame laminated substrates (b, foreground a) act as a kind of expansion joint.

1.2 Alternative Thermal Treatment

Looking for alternative technologies for the thermal treatment of functional layers is a constant topic of research in the field of functional coatings and especially in the field of nanoparticles (German, 1996), (Perelaer, 2006), (Tobjörk, 2012). Two devices, which are commonly used in the graphic arts industry – a heat press usually used for textile prints and a hot stamping machine –, are compared in the following to oven drying. Both machines provide temperature and pressure at the same time. The pressure will be neglected in this examination and will be set to a minimum. The heat press applies heat and pressure on an area of 37.5×27.5 cm. The hot stamping machine utilizes interchangeable stamps such that only the area of interest will is pressed and heated. The stamp used in the tests was of the same size like the test structure. This is considered to be a major advantage of the hot stamping process, because the thermal loading to the substrate and the dimensional instability is assumed to be less with the hot stamping. The thermal and mechanical impact on the printed layers may also have positive impact on the adhesion to the investigated substrate.

2. Materials and Methods

2.1 Materials

Based on preliminary tests the materials and patterns were chosen for further experiments. The experiment setup is listed in Table 2.

The cardboard laminates were susceptible to the temperature impact in the batch oven. Since the investigation is aiming towards less temperature loading to the substrate, cardboard is a good indicator for excessive thermal loading. The frame-patterned adhesive allows for better removal of the attached substrate, whereas the full area printed adhesive may show improved stability during printing and further processing. Both designs were examined.

Substrate Carrier	Adhesive	Adhesive Pattern
Cardboard	Solvent based KIWOPRINT TC 2500/1	Full Area Frame
Screen Mesh	Sefar PET 1500 120-34 & 77-48	
UV Ink	SunChemical SunTronic 680	
Silver Ink	Acheson Electrodag® PM-406	

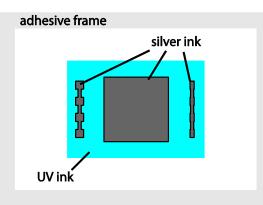


Figure 4: The test layout consisting of an adhesive frame (or full area printed adhesive, not shown), UV curing ink and silver ink test structure for four point probe measurement and area for cross-cut tests

The UV-curing ink is printed on the substrate without plasma treatment. In the preliminary tests the SunChemical ink provided good results. Other UV inks and solvent based inks were not able to adhere to the untreated PE surface. The silver ink test patterns – as shown in Figure 3 and Figure 4 – are used for electrical measurements with a four point probe setup. The printed square was used for cross-cut tests. The ink layer thickness was measured with an optical 3D microscope and verified with a mechanical thickness tester. The electrical measurements were performed with a digital multimeter.

2.2 Methods

The samples were printed on cardboard reinforced PE-Al-PET compound foil with an Ekra X-1 screen printer (EKRA GmbH, Germany). Prior to printing the test samples, the adhesive KIWOPRINT TC 2500/1 was printed onto the cardboard substrate carrier and laminated with the compound foil. The screens were tensioned with PET 1500 120-34 and 77-48 meshes (SEFAR AG, Switzerland). The stencils were made of AZOCOL Z 160 HV emulsion (Kissel + Wolf GmbH, Germany). The UV curing dielectric ink SunTronic 680 (Sun Chemical Corporation, USA) was used as an adhesion agent. The silver ink Acheson Electrodag® PM-406 (Henkel AG & Co. KgaA, Germany) was used for

electrical characterization of the print samples. Plasma treatment was performed with a desktop ambient atmosphere plasma-device (Plasmatreat GmbH, Germany). The oven-cured samples were processed in a batch oven (Binder, Germany). The thermal press picollo plus (Walter Schulze GmbH, Germany) and the hot stamping press (Robertshaw, USA) were used for examining alternative thermal treatment. Four point probe measurements were performed with spring-loaded round-shaped probe heads (Feinmetall, Germany) and a digital multimeter M3510A (Picotest Corp., USA). Adhesion was tested with a Cross-Cut-Tester, 1 mm, DIN/ISO (BYK-Gardner GmbH, Germany). Thickness of the layers and overall roughness were determined with an optical microscope Infinite Focus (Alicona Imaging GmbH, Austria).

3. Results and Discussion

3.1 Effect of Thermal Process on Ohmic Resistance

Thermal treatment of conductive inks printed on sheetfed material is mostly performed in batch ovens. These allow curing of functional inks from room temperature up to roughly 300° C. Curing temperature and time are limiting factors in printed electronics. In this specific scenario the temperature is even more critical, since it causes dimensional distortion due to the low maximum processing temperature of PE and the thermal mismatch of the two polymer layers. This results in alignment shifting of the successively printed inklayers. The batch oven curing process is therefore not useful for this specific process of printing batteries. The samples, which were cured in the batch oven, are reference samples to evaluate the alternative processes. The negative impact of the batch oven curing is shown in Figure 3.

Thermal Treatment in Batch Oven

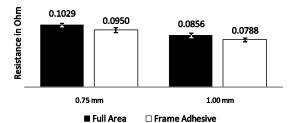
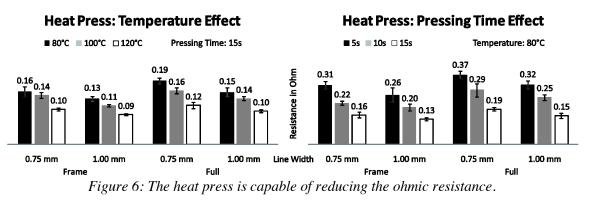
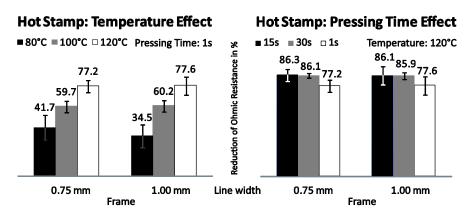


Figure 5: The thermal treatment in the batch oven is the reference for alternative curing processes.



The achievable resistance is in the same order of magnitude as with oven dried samples.

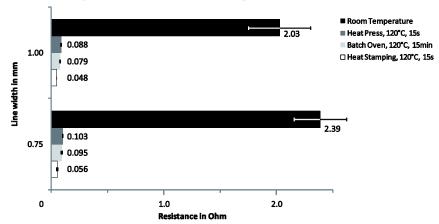
In Figure 6 the impact of the heat press is illustrated. The effect of temperature is shown in the diagram on the left hand side. The higher the temperature is, the lower is the resistance. On the right hand side the effect of pressing time is shown at 5, 10 and 15 seconds. As expected, the ohmic resistance is linearly decreasing with increasing process time. The way the compound foil is attached to the substrate carrier seems to influence the ohmic resistance as well. At least there is a tendency of increased resistance when the adhesive for laminating the compound foil on the carrier covers the full



area. The samples, which are laminated by using only a frame of adhesive surrounding the image area show slightly lower ohmic resistances.

Figure 7: The process of hot stamping provides an efficient way of reducing the ohmic resistance.

The same effect of time on the resistance is evident in the case of the hot stamping processed print samples. In Figure 7 the percentage of resistance reduction is plotted against the process time. Even at low temperature of 80° C and with a very short processing time of one second, the reduction is very high, achieving ~40% resistance reduction. At higher temperatures and longer pressing times of 15 seconds, the resistance is reduced about 86%. Increasing the impact time from 15 to 30 seconds or even to 60 seconds does not reduce the ohmic resistance any further. It seems reasonable that at this point the maximum percolation of the conductive particles is achieved. Moreover the print samples are deformed when pressing for 60 seconds.



Impact of Thermal Processing on Ohmic Resistance

Figure 8: Comparison of the investigate routes for curing conductive silver ink.

As depicted by Figure 8, the samples dried at room temperature show the highest ohmic resistance. The resistance of the heat press samples are slightly higher than the batch oven results. The lowest ohmic resistance is provided by the hot stamp samples. The advantages of hot stamping are fast processing and less impact on the substrate. The heat press is less efficient and more harmful to the substrate than hot stamping.

3.2 Mechanical Stress & Determination of morphological parameters

In order to identify the mechanical stress of the hot stamping and hot pressing on the printed layers, the morphological properties were measured. Important parameters are the surface roughness, layer thickness and adhesion to the underlying substrate. Roughness and thickness values were obtained by optical 3D microscope measurements on the basis of focus variation (alicona infinite focus). Thickness

and roughness data were obtained before and after the treatment with thermal transfer pressing and hot stamping at 120° C for 15 seconds. The adhesion strength of the ink layer was tested in cross-cut and tape-tests.

The heat press processed samples showed thickness differences after pressing. In average, the layer thickness decreased about 30%. Values dropped from roughly 13-15 μ m to about 9-10 μ m. The heat press process did not damage the samples, whereas visual inspection clearly showed the impact of the hot stamp on the samples (imprint of the stamp). The pressure of the hot stamp led to deformation of the substrate, as indicated by Figure 10. Thus, reliable thickness data could only be obtained from the samples, which were processed with the heat press. The results of the thickness measurements for a temperature of 120° C and a processing time of 15 seconds are given in Figure 9. The samples were printed on full area laminated substrates.

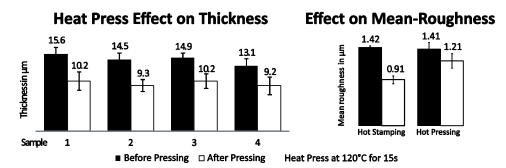


Figure 9: Decreased layer thickness after hot pressing (left) and the pressing effect on the meanroughness

It is likely that the pressing process leads to a densification of the silver layer that decreases the overall thickness. This is supported by the fact that the average surface roughness changed, too.

After thermal treatment by hot stamping or hot pressing, the roughness of the silver layer decreased noticeably. The overall mean roughness seems to be lower for the samples which were processed with the hot stamp. Deviations in terms of surface roughness are in the range of 15% for the hot pressed samples and up to 35% for the hot stamped samples.

It is assumed that the significantly higher pressure of the hot stamping influenced the surface of the samples to a great extent, what can be observed visually. An image, which clearly shows the imprint of the stamp on the sample, is given in Figure 10.

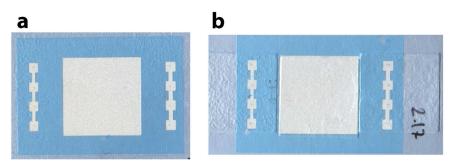
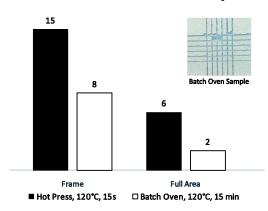


Figure 10: a) Heat press and b) hot stamping. The impact of the stamp is clearly visible.

3.2.1 Cross-cut and tape test

The general adhesion of the ink on the substrate was tested in two experiments: Cross-Cut-Test and tape test. Samples were cross-cut in a 90° angle. This led to 25 equally shaped squares. The test procedure is in accordance with ISO 2409 & ASTM D 3359-97. (Byk-Gardner, 2015) This partially

removed the small squares from the surface. The number of removed squares was counted. This should give an indication of the adhesion of the ink. Figure 11 shows the impact of the crosscut.



Cross-Cut-Test: Removed Squares

Figure 11: Cross-cut test on an oven-dried sample (120° C, 15 min). The pattern of the adhesive seems to have an impact on the adhesion. The quality of adhesion also depends on the thermal process

Cross-cut test on samples which had atmospheric plasma pre-treatment showed very good adhesion on the underlying substrate. They were not harmed by the cross-cutting.

The patterning of the adhesive layer (full area or frame laminated) seems to have an effect on the adhesion. This is exemplarily shown in Figure 11.

The adhesion of the ink layer seems to be temperature dependent as well. Samples which were hot pressed at 80° C for 15 seconds showed less adhesion to the substrate compared to samples which were pressed at 120° C for the same time. However this test was done manually and thus slight variations in downwards pressure and cutting speed cannot be avoided. This could be optimized by an automatic cross-cutting machine, which was not available. Further tests need to be done in order to prove this.

3.2.2 Tape test

A tape test was done on the samples, which partially or fully removed the ink layers from the substrate. This was true for all samples except for samples that were treated with plasma prior to printing. No tendency in terms of temperature related adhesion could be noticed regarding the tape test

Impact of Creasing on Ohmic Resistance

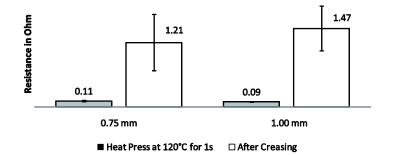


Figure 12: Creasing the printed structures and bending the compound film on the substrate carrier resulted in a massively increased resistance

A manual crease roller was used to simulate mechanical stress on the functional ink layers. After creasing the samples, the compound foil and the substrate carrier were bent 180°. A four point probe measurement was performed after flattening the samples. The increase in ohmic resistance was massive, but most of the samples still showed electric conductivity. It is only possible to judge the outcome of this test qualitatively, because the resistance measurement was not consistent. Thus, the creasing test only provides the mere result of continuity or discontinuity of the electric pathway. The measured values lack of significance and reliability, as indicated by the huge error bars in Figure 12. As shown with this crease roller test even a very violently handling of the print layers does not destroy the functionality.

4. Conclusions

The goal of this examination was to establish a route for screen printing electrically conductive structures on PE-Al-PET substrate. These structures are part of a printed battery that benefits from the properties of the compound substrate, i.e. the PE-weldability and the thin substrate thickness. An UV-curing ink was introduced to act as an adhesion agent. This UV-ink improved the mechanical stability of the print samples. The formerly crumbling silver layers gained elasticity by the underlying UV-ink layer. The adhesion to the untreated substrate improved as well, but it is certainly less effective than a plasma treatment. The thermal press and hot stamping curing showed also an improvement of the adhesion to the untreated substrate. Additionally, the ohmic resistances of the pressed samples were comparable (heat press) to or lower (hot stamping) than the samples cured in the batch oven at 120° C for 15 min. The average thickness and roughness of the screen-printed silver layers seem to be influenced by the hot stamping and the hot pressing process respectively. The cross-cutting and tape test showed certain tendencies that need to be further investigated. Especially the influence of the temperature on the adhesion needs to be considered.

Acknowledgments

Special thanks go to Prof. Dr. Erich Steiner for fruitful discussions, Maryam Zakerighadi and Jürgen Krumbach for supporting the practical work. The authors would also like to thank the industrial partners for their assistance in providing materials and technical: SunChemical, Kiwo, Sefar, Huhtamaki, Feinmetall and Ekra.

References

Banfield, D., 2000. Understanding and measuring electrical resistivity in coductive inks and adhesives, SGIA, 6 pp. 29-36.

Bülow, T., Gargouri, H., Siebert, M., Rudolph, R., Johannes, H. H., Kowalsky, W., 2014. Moisture barrier properties of thin organic-inorganic multilayers prepared by plasma-enhanced ALD and CVD in one reactor. Nanoscale Research Letters. 9(1) pp. 223

Byk-Gardner Byk-Gardner, Intro Adhesion [Online]. - Byk-Gardner, 2015. - 31 05 2015. - http://www.byk.com/fileadmin/byk/support/instruments/theory/physical-properties/en/Intro_Adhesion.pdf.

Chun, K.Y., Oh, Y., Rho, J., Ahn, J. H., Kim, Y. J., Choi, H. R., Baik, S., 2010. Highly conductive, printable and stretchable composite films of carbon nanotubes and silver. Nature Nanotechnology, 5 pp. 853–857.

German R. M. 1996. Sintering Theory and Practice, John Wiley & Sons, p. 568.

KoSiF [Online]. - 2013. - 30 05 2015. - http://kosif.ims-chips.de.

Lommatzsch, U., Pasedag, D., Baalmann, A., Ellinghorst, G., Wagner, H. E., 2007. Atmospheric Pressure Plasma Jet Treatment of Polyethylene Surfaces for Adhesion Improvement. Plasma Processes and Polymers, 4(1) pp. S1041–S1045.

Paints and varnishes - Cross-cut test (ISO 2409:2007); German version EN ISO 2409:2007.

Park, J. S., Chae, H., Chung, H. K., Lee, S. I., 2011. Thin film encapsulation for flexible AM-OLED: a review. Semiconductor Science and Technology, 26 034001 8pp

Perelaer, J., de Gans, B.-J., Schubert, U.S., 2006. Ink-jet Printing and Microwave Sintering of Conductive Silver Tracks. Advanced Materials, 18(16), pp. 2101-2104.

Rossmann, K. 1956. Improvement of bonding properties of polyethylene. Journal of Polymer Science, 19(91) pp. 141-144.

Tobjörk, D., Aarnio, H., Pulkkinen, P., Bollström, R., Määttänen, A., Ihalainen, P., Mäkelä, T., Peltonen, J.,

Toivakka, M., Tenhu, H., Österbacka R., 2012. IR-sintering of ink-jet printed metal-nanoparticles on paper, Thin Solid Films. 520(7), pp. 2949-2955.

Xu L .Y., Yang, G. Y., Jing, H. Y., Wei, J., Han Y. D., 2013. Pressure-assisted low-temperature sintering for paper-based writing electronics. Nanotechnology, 24(35), 355204

Yoshida, A., Sugimoto, A., Miyadera, T., Miyaguchi, S., 2006. Organic Light Emitting Devices on Polymer Substrates, Journal of Photopolymer Science and Technology, 14(2001) 2 pp. 327-332.

A new FEM Simulation Method of Paper Materials by using a Gasket Model

Jian Chen¹, Jann Neumann², Hans Martin Sauer¹, Edgar Dörsam¹

¹Institute of Printing Science and Technology, Darmstadt University of Technology, Magdalenenstr.2, 64289 Darmstadt
 ² Perfecta Cutting Systems GmbH, Schäfferstr.44, 02625 Bautzen

E-mail: chen@idd.tu-darmstadt.de; neumann@perfecta.de; sauer@idd.tu-darmstadt.de; doersam@idd.tu-darmstadt.de

Short Abstract

A number of models have been proposed with the objective of predicting the stress-strain behaviour of paper materials in out-of-plane (ZD) direction, but the simulation research is still quite limited. The mechanical behaviour of paper materials under compression is highly nonlinear, and it is also exhibits quite complicated unloading behaviour when compression is released. The stress-strain curve of paper materials is a typical example of materials with J-shaped compressive curves, which is the same as seen for gasket materials. There is still no model provided in ANSYS or ABAQUS which could be directly used for paper simulation. But the thickness-direction behaviour of gasket materials could be simulated using finite element (FEM) software. The main purpose of this paper is to actualize the compressive simulation of paper materials by using the gasket elements provided in the FEM software. In this paper, the accuracy of this new method was verified by comparing with the experimental results. The compressive simulation of paper under some further selected forces was also calculated and verified based on the material model established. Comparative analysis of the experimental results proves the effectiveness of the simulation.

Keywords: paper simulation, out-of-plane (ZD) direction, FEM software, gasket element

1. Introduction and background

The compressive behaviour of paper in the out-of-plane direction has a very close relationship with many operations in the print production process, such as printing, paper counting, folding, creasing, calendering, cutting etc. The compression properties of paper have been studied in numerous papers.

1.1 Introducing paper models

Depending on whether the surface topography is taken into account, the compressive analysis of paper in the through-thickness z-direction can be divided into two groups.

Some of the literature describes attempts to model the paper as a smooth solid material, and the properties of paper, such as elasticity, plasticity and viscosity, were modelled, respectively, by using spring, dry friction and dashpot elements (Ribeiro, 2007, Gavelin, 1949). According to this approach, the differences between these constitutive models mainly lie in the different combinations of these elements. Ignoring the surface roughness provides convenient analysis of processes such as paper delivery and paper calendering (Eckstein, 2014).

When compressing thin sheets, however, it is very important to be aware of the influence of surface roughness (Rättö, 2005). The knowledge of the surface topography plays an important role in obtaining the stress-strain curve of paper materials. The influence of surface roughness is, therefore, widely discussed. Four different methods were evaluated for characterizing the smoothness of handsheets (Singh, 2008). Paper surface topography under compression was also studied (Teleman, 2004). According to the surface topography, the paper body was considered as being composed of two

rough surfaces and an internal structure (Schaffrath, 1991), the force-deformation relationship of paper was derived by using the Newton formula.

1.2 State of the art in paper simulation

Only very few references attempt to establish the simulation model in finite element (FEM) software, but even in those which did, the results remained inconclusive in some respects. In one study, the simulations applied an orthotropic elastic-plastic constitutive model for describing the material behaviour of paper. The commercial FE-code ABAQUS/Standard was adapted in the numerical analysis and the quadratic isoparametric eight-node planar stress elements used for defining the paper material (Mäkelä, 2003). In another study, a computational micromechanical material model for the unloading behaviour of paper and other nonwoven materials was presented (Ramasubramanian, 2007), and four-node bilinear planar stress elements were used in the ABAQUS/Standard model for simulating the uniaxial tensile test.

In the year 2002, a theoretical framework to account for damage in paperboard was proposed, which consists of two parts: an in-plane continuum model, and an interface model (Xia, 2002). The two models were implemented, respectively, into ABAQUS /Standard as a user-defined material (UMAT) and a user-defined interface (UINTER) for modelling the creasing and folding of paperboard (Nygards, 2005). In addition, under the supervision of Nygards, the simulation of paperboard creasing was completed and improved in a master thesis based on the model proposed by Xia (Andersson, 2006).

1.3 Aim of the research

Unfortunately, because of the complexity of the paper materials, there is still no material model provided in FEM software which could be used directly for paper simulation. All the simulation works presented up to now are based on constitutive models, which need a considerable number of free parameters requiring to be obtained by experiment. Additionally, most of these parameters are very difficult to measure. Furthermore, the compressive behaviour of paper stacks is related to the additional aspect that there is interaction between the individual sheets. These kinds of simulation models are, however, very difficult to be extended to the research of multiple sheets. In this paper, we try to find a new simulation method for evaluating the stress-strain relationship of paper materials.

2. Materials and Methods

2.1 Experimental setup

The laboratory of the Institute of Printing Science and Technology (IDD) is equipped with the universal testing machine Zwick Z050, which can be utilized for strain, shear and bending tests with different substrates and machine components with high accuracy over a cross head speed range of 0.0005 - 2000 mm/min, and a position repetition accuracy of $\pm 2 \mu \text{m}$.

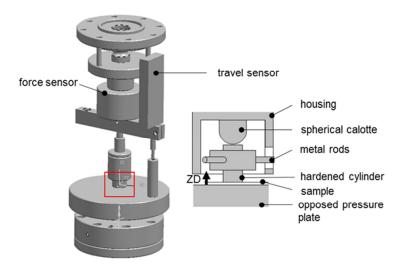


Figure 1: Test equipment for determining force deformation behaviour of paper (Kaulitz, 2009)

The test equipment used at the IDD is shown in Figure 1. Two kinds of force measurement systems are available in the stress testing device, one for high loads up to 1 kN (with diameter of the pressure head d=11.3 mm) and the other for smaller forces under 120 N (with diameter of the pressure head d=6.0 mm). The specific weight of the studies paper specimen was 80 g/m², and actual average thickness was $d=102 \mu m$. The measured force-deformation curve of paper materials is showed in Figure 2.

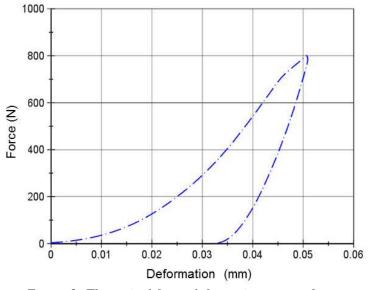


Figure 2: The typical force-deformation curve of paper (A force of 800 N corresponds to about 8 N/mm²)

According to the size of the press head and paper thickness, the force-deformation curve could be easily transferred into a stress-strain curve. The compressive curve of paper material is a typical example of materials with J-shaped stress-strain curves, which is very similar to gasket and soil materials. The curve shows that initially, small increases in stress give large deformations, however at larger deformations the material becomes stiffer and more difficult to be compressed. The data obtained in Figure 2 could be used for defining the material property in FEM software and thereafter compared with the simulation results.

2.2 Simulation process

Gaskets are usually designed as very thin sheet-like structures, acting as sealing components between structural components. They are designed to provide appropriate pressure-closure behaviour by virtue of their ability to adapt their thickness according to contact with a given rigid surface, so the primary deformation behaviour of gasket joints is the one perpendicular to the sheet plane. There is no available model provided in ANSYS or ABAQUS which could be directly used for paper simulation, but both FEM environments offer a variety of gasket elements that generate through-thickness behaviour specifically designed for the study of gaskets. The main purpose of this paper is to actualize the simulation of paper materials by using the gasket elements provided in the FEM software.

The principle of computer simulation of gasket material is basically the same. For gasket materials, most of the simulation processes are very similar to the simulation of other materials. Generally, the process flow of the simulation in FEM software consists of pre-processor, solution control, solve and postprocessor etc. The different simulation processes between ANSYS and ABAQUS are listed and compared in Figure 3. Both of the simulation processes in ANSYS and ABAQUS could be divided into nine steps. The principle of computer simulation of gasket material is basically the same. The simulation based on ANSYS was chosen here as an example of introducing the simulation process. For gasket materials, most of the simulation processes are very similar to the simulation of other materials. The main differences exist in the process of choosing the elements, defining the material properties and meshing.

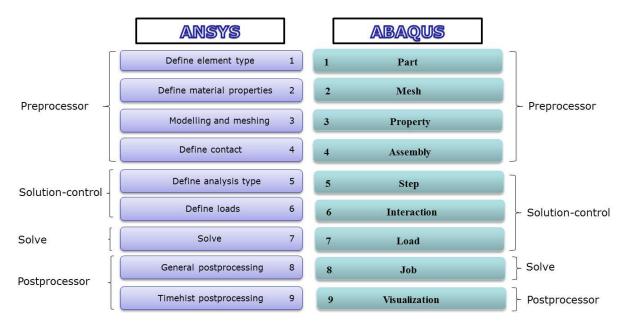


Figure 3: Comparison between the simulations of ANSYS (ANSYS Help, 2012) and ABAQUS (ABAQUS/CAE Help, 2012)

The material data needed as input for the FEM software consist of two parts: general parameters and pressure closure behaviour. The general parameters define initial gap, stable stiffness etc. The pressure closure behaviour includes loading and unloading data. The element SOLID185 (ANSYS Help, 2012) was chosen in this paper for defining the material of stiff platform and press head. Element INTER 195 (ANSYS Help, 2012) was used for defining the paper material.

2.2.1 Fundamental theory of defining the gasket material properties in FEM software

The definition of the material properties was divided into two parts: the loading process and the unloading process, the behaviour of which have each to be defined, respectively. The in-plane stiffness and transverse shear was negligible when compared to the stiffness in the out-of-plane direction. The

loading and unloading curve of gasket material could be obtained from the following compression curve function (Jorwekar, 2006).

Compressibility model for synthetic gasket material were:

$$y = a \cdot \left(1 - e^{-\binom{x}{b}}\right) + c \cdot \left(1 - e^{-\binom{x}{d}}\right) + u$$
 [1]

Slope:

$$\frac{dy}{dx} = \binom{a}{b} \cdot e^{-\binom{x}{b}} + \binom{c}{d} \cdot e^{-\binom{x}{d}}$$
[2]

Tangent modulus:

$$E = \frac{dx}{dy}$$
[3]

Unloading curve function (due to the unrecoverable strain):

$$u(z) = a \cdot \left(1 - e^{-(z_{b}')}\right) + c \cdot \left(1 - e^{-(z_{d}')}\right) - \left[a \cdot \left(1 - e^{-(z_{b}')}\right) + c \cdot \left(1 - e^{-(z_{d}')}\right)\right]$$
[4]

where y is the strain or the percentage of the compression; x is the stress; a and c are the scale factors; b and d are the time variables; z is the maximum stress achieved and u is the loading offset, when u=0, which is used for the loading curve.

According to the equations provided above, the gasket loading and unloading behaviour could be calculated. Equation 1 gives the compression curve function of gasket material, equation 4 gives the unloading curve function of gasket material under the unrecoverable strain.

2.2.2 Definition of the loading material properties

For the loading process, the pressure and closure data are needed for defining the paper property. Some discrete data points $A_i(x_i, y_i)$ (*i*=1, ..., *n*) obtained by experiment (Figure 2) were selected as shown in Figure 4. x_i is the value of closure (reduced thickness in mm), y_i is the corresponding pressure value (MPa).

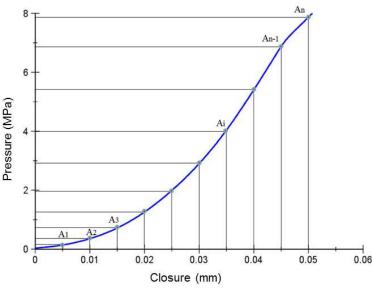


Figure 4: The data selected for defining the loading process

2.2.3 Definition of the unloading material properties

Two different unloading simulation methods are provided in ANSYS: linear unloading simulation and nonlinear unloading simulation. The linear unloading material definition option is a simple way to

define the paper unloading behaviour, in which several unloading slopes (Equation 2) can be defined to accommodate the unloading behaviour. The nonlinear unloading gasket material definition option provides a more comprehensive way of defining gasket material unloading behaviour (Equation 4).

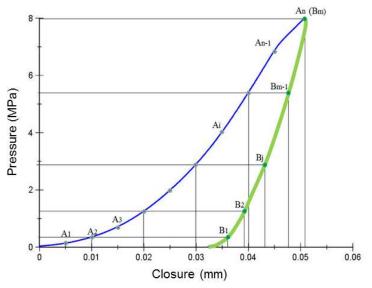


Figure 5: The data selected for defining the unloading process

Some discrete data points $B_j(x_j, y_j)$ (j=1, ..., m) in the unloading process were selected as shown in Figure 5. All the longitudinal coordinate values of these selected points should be the same as the loading points. The main difference between the linear unloading simulation and nonlinear unloading simulation lies in the definition of the employed material parameters. For linear unloading simulation, x_j is the value of closure (mm), y_j is the corresponding unloading slope (-) (Equation 2). For nonlinear unloading simulation, x_j is the value of closure (mm), y_j is the corresponding unloading pressure value (MPa). The point A_n of maximal loading should be the same as B_m , the starting point of the unloading process.

2.2.4 Modelling and simulation

Figure 6 is a simple finite element model created to demonstrate the paper material simulation. Two block elements of type SOLID 185 (ANSYS Help, 2012) were generated for the sample supporter and a gasket element INTER 195 (ANSYS Help, 2012) was created for the paper sample itself. Then meshing, defining the constraint condition and imposing the load are undertaken.

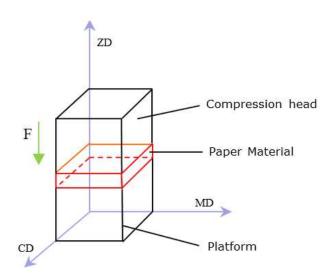
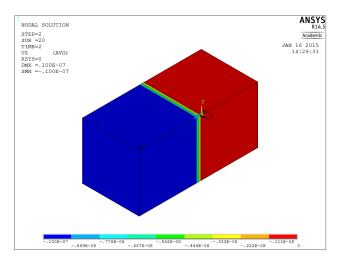


Figure 6: Schematic representation of the simulation model

The simulation results were then derived in the post-processing step. The force-deformation curve could be obtained directly and the data could also be stored as text documents.

3. Results and Discussion



The simulation result of the displacement is shown in Figure 7.

Figure 7: The simulation result of displacement in the through-thickness direction

3.1 Paper simulation under a defined force

For comparison between the simulation result and the experiment, as shown in Figure 2, a force maximum of 800 N was provided in the loading process, and implemented in both the simulation of linear unloading and nonlinear unloading processes. The simulation results are shown in Figure 8 and Figure 9. The picture in Figure 8 is the simulation result under linear unloading, Figure 9 shows the simulation result under nonlinear unloading.

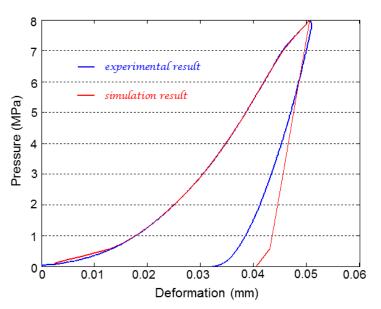


Figure 8: The linear unloading simulation result compared with the experimental result (A pressure of 8 MPa corresponds to about 800 N)

Figure 8 shows the comparison of the experimental and the simulated pressure-deformation curve for ZD compression, the simulation result of loading process is in perfect agreement with the

experimental result. To a certain extent, the linear unloading simulation can qualitatively describe the trend of the actual process, but is still lacking full predictive accuracy.

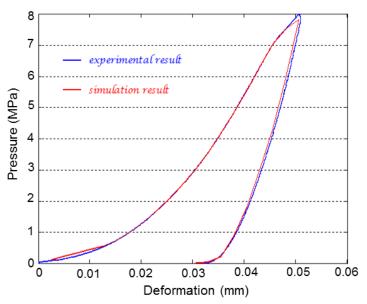


Figure 9: The nonlinear unloading simulation result compared with the experimental result (A pressure of 8 MPa corresponds to about 800 N)

Figure 9 displays the comparison of the experimental and the nonlinear unloading simulated curve. As shown previously, the simulation result is much more precise, and this simulation model can be used with high confidence for doing further simulations of paper compression.

3.2 Paper simulation with variable maximum forces

According to the simulation model of paper material established above, the compression simulation of paper under different forces were conducted, the results are shown in Figure 10. Three groups of simulations were implemented here, in which the respective maximum forces applied are 200 N, 400 N and 600 N.

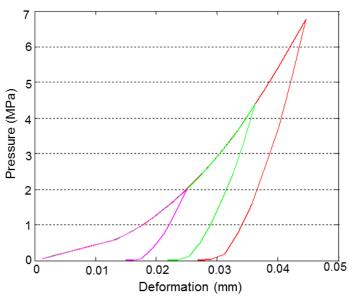


Figure 10: The simulation results under different forces

In order to verify the applicability of the simulation model, some experiments on paper, using different maximum compression forces, were also performed and the results from which are shown in Figure 11. In the experimental process, the maximum forces provided were also 200 N, 400 N and 600 N. The experiments were made on different points of the paper surface. For each load and unload cycle, the moving speed of the pressure head was set as 0.05 mm/min.

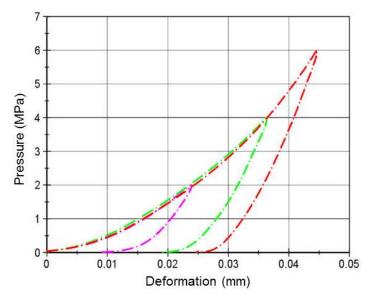


Figure 11: The experimental results under different forces

From Figures 10 and 11, it can be seen that the agreement between FEM simulation and the experimental result is very good. The method can be used with high confidence for the simulation of paper under other different forces.

4. Conclusions

In this paper, a much easier way than previously reported techniques was proposed to simulate the *z*-direction compression of paper, avoiding many difficult measurements (Andersson, 2006) that would normally be needed for defining the parameters. The method is based on finite element modelling using the gasket approximation. This result will also provide a basis for the simulation of multiple sheets or paper stacks. Intrinsically, the simulation method presented here implies some type of nonlinear curve fitting, especially the simulation under a defined force, but it provides a way of doing the simulation of paper under any different forces to a high level of predictive confidence.

Furthermore, the paper material was also modelled here as a smooth solid material, the influence from the surface roughness has not been taken into account. Some simulation work concerning the surface topography should be supplemented in future.

Appendix

During the compression process, for many materials the compressional stress is directly proportional to the compressional strain. Generally, at the beginning of the loading process, the relationship between stress and strain is linear, which is known as Hooke's Law.

For some other materials, such as rubbery materials, an S-shaped stress-strain curve is obtained (Figure 12). The initial part of this curve, where the stiffness decreases with increasing load, can be easily predicted theoretically. However, a clear transition from approximated linear to nonlinear behaviour is observed, and stiffness increases with increasing load, which is the same as paper or gasket materials. The increasing stiffness leads to an increase of the real modulus (the slopes of the

stress-strain curve), which is very difficult to be calculated theoretically because of the nonconvergence, as well as simulation.

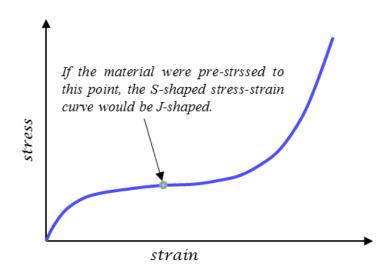


Figure 12: An S-shaped curve compared to a J-shaped curve

If the rubber type material was pre-stressed to the point shown in Figure 12, the S-shaped stress-strain curve would be J-shaped. Many special materials, such as pre-stressed rubber, show J-shaped behaviour only.

Acknowledgments

The authors gratefully acknowledge the financial support from the China Scholarship Council (CSC).

References

ABAQUS/CAE Help, 2012. Version: 6.12-1: ABAQUS/CAE 6.12-1 User's Manual. ABAQUS, Inc

Andersson, T., 2006. A small deformation model for the elasto-plastic behaviour of paper and paperboard. (Master's Thesis, Lund University)

ANSYS Help, 2012. Version 14.5: ANSYS Help system-mechanical APDL-Element reference. ANSYS, Inc

Eckstein, M., 2014. Instabilities and wear propagation in calenders: Interactions with structural dynamics and contact kinematics. (Doctoral dissertation, Darmstadt University of Technology)

Gavelin, G., 1949. The Compressibility of Newsprint, Svensk Papperstidning, 52, pp.413-419

Jorwekar, P.P., Birari, Y.V. and Nadgouda, M.M., 2006. Cylinder head gasket contact pressure simulation for a hermetic compressor. *International Compressor Engineering Conference at Purdue*

Kaulitz, T., 2009. Bilden von Schneidlagen unter Ausnutzung des Nipinduzierten Effekts für die Druckweiterverarbeitung. (PhD dissertation, Darmstadt University of Technology)

Mäkelä, P., Östlund, S., 2003. Orthotropic elastic-plastic material model for paper materials. *International Journal of Solids and Structures*, 40, pp. 5599-5620

Nygards, M., Hallbäck, N., Just, M. and Tryding, J., 2005. A finite element model for simulations of creasing and folding of paperboard. *ABAQUS Users' Conference*

Ramasubramanian, M.K., Wang, Y., 2007. A computational micromechanics constitutive model for the unloading behaviour of paper. *International Journal of Solids and Structures*, 44, pp. 7615-7632

Rättö, P., 2005. The influence of surface roughness on the compressive behavior of paper, *Nordic Pulp and Paper Research Journal*, 20, pp. 304-307

Ribeiro, H. A., Costa, C.A.V., 2007. Modelling and simulation of the nonlinear behaviour of paper: A cellular materials approach. *Chemical Engineering Science*, 62, pp. 6696-6708

Schaffrath, H.-J., and Göttsching, L., 1991, The behavior of paper under compression in z-direction, *Proceedings* of the 1991 Tappi International Paper Physics Conference, Kona, Hawaii, pp. 489–510

Singh, S.P., 2008. A comparison of different methods of paper surface smoothness evaluation. *BioResources*, 3 (2), pp. 503-516

Teleman, A., 2004. Analysis of paper surface topography under compression. STFI, CW217

Xia, Q.S., Boyce, M.C. and Parks, D. M., 2002. A constitutive model for the anisotropic elastic-plastic deformation of paper and paperboard. *International Journal of Solids and Structures*, 39, pp. 4053-4071

42nd International Research Conference, 06 to 09 September 2015



Session 2B

Print for packaging

Monday, 7 September 2015 14:00 – 15:40

Chair: Fons Put

Critical Evaluations of Liquid Absorption Testing Methods for Package Printing

Li Yang and Sofia Thorman

Innventia Box 5604, 114 86 Stockholm, Sweden

E-mail: li.yang@innventia.com; sofia.thorman@innventia.com

Corresponding author: Li Yang, +46 8 676 7134

Short Abstract

Liquid absorption is often of importance in flexographic printing, yet is not easy to pinpoint or visualise, due to its co-existence with surface roughness. To study the relative importance of ink absorption, adequate characterisations on the absorption dynamics are essential. Hence, we have tested and compared two relatively new techniques, Automatic Scanning Absorptometer (ASA) and Automatic Cobb Test (ACT). These instruments offer not only more in-depth information concerning absorption dynamics, but also operational benefits compared to the conventional Bristow Wheel and Cobb testing methods. Absorption tests with specially designed pilot coated boards that cover a wide range of absorbency have been performed. Both ASA and ACT techniques ranked the boards similarly in terms of absorption rates, except for the board with a relatively hydrophobic coating. The ASA measurement indicated there being an absorption delay in this sample, which was not seen with the ACT measurement. Moreover, the ASA offered better time resolution in the short time regime, t < 1 s. Our study also suggests that when surface roughness is not dominating, print density exhibits a correlation with the absorption rate, namely greater absorbency leads to higher print density. Despite this finding, one should not over-generalize this observation, as the samples had been specially designed to provoke large differences in absorbency.

Keywords: absorption dynamics, print density, ink absorbency, flexography.

1 Introduction

Ink absorption and surface roughness are often regarded as two major factors that affect flexographic printing quality. Generally speaking, surface roughness has often been found to cause print quality problems (Lagerstedt and Kolseth, 1995, Aspler *et al.*, 2004, Wågberg and Wennerblom, 1992, Barros *et al.*, 2005, Barros and Johansson, 2006), but there are also occasions when print defects cannot solely be explained by surface roughness. In other words, a smooth surface does not necessarily mean high print quality, because ink absorption may play an important role or even dominates the print quality (Aspler *et al.*, 1998, Sheng *et al.*, 2000). Nevertheless, the true importance of the ink absorption is difficult to pinpoint or visualize, due to the simultaneous presence of surface roughness.

Relevant measurement techniques of ink absorption are highly important for understanding flexographic print quality. Cobb (TAPPI, 1998) and Bristow Wheel (Bristow, 1967) testing methods are two of the most representative and widely accepted measurement techniques for liquid absorbency. However, these techniques have their intrinsic limitations, e.g. typically having far too long a time of absorption compared to the printing process and/or operator dependence. Depending on the printing technique and speed, the absorption time may vary from milliseconds (ms) to seconds (s). Moreover, none of the techniques provides insights into absorption dynamics, which is critical to understand the printing dynamics. We made, therefore, systematic studies with a special focus on short term absorption, using two relatively new techniques: Automatic Scanning Absorptometer (ASA, from KRK KUMAGAI RIKI KOGYO Co., Ltd. Tokyo, Japan) and Automatic Cobb Test (ACT, from FIBRO system AB, Hägersten, Sweden). These two techniques offer not only operational benefits but also details of absorption dynamics.

The objective of this work is to study the impact of ink absorbency on print density with board samples covering a wide range of pore structures, surface chemistries and ink absorbency, to thereby reduce the impact from surface roughness.

2 Experimental details

2.1 Materials

Six pilot-coated packaging boards are included in the study, which span a broad ink absorption range. They all feature the same base board (200 g/m²) and pre-coating (11.8-12.5 g/m²), but have different top-coating formulations (11.3-12.0 g/m²) in order to create different pore-structures and surface chemistry. Different latex types, different amounts of latex and different pigment blends have been used. Details of the top coating compositions and board properties are summarized in *Table 1*. The coating was performed with a bent ceramic blade at 600 m/min.

Table 1: Details of the top coating compositions, surface roughness as standard deviation of height about the mean in the lateral wavelength interval 0.06-1 mm and contact angle of water after 0.1 s.

No.	Notation of the board	Coating compositions of top-coatings and the pre-coating (in addition, 0.5 pph thickener, FinnFix 10, Noviant Oy, Finland, and 0.08 pph caustic soda (NaOH) were used in all formulations)	Roughness, std.dev height [µm]	Contact angle, after 0.1 s, H_2O [°]
1	12.5 Latex A	12.5 pph of vinyl acetate acrylate latex (VAA) CHP 2635EP (CH Polymers Oy, Raisio, Finland), ground calcium carbonates with broad particle size distributions (PSD): 60 pph Hydrocarb® 90 and 40 pph Setacarb® HG. All carbonates in top- and pre-coatings were from Omya International AG, Oftringen, Switzerland.	0.86	71.2
2	15 Latex A	15 pph of the VAA latex. Same pigments as board no. 1.	0.88	75.2
3	20 Latex A	20 pph of the VAA latex. Same pigments as board no. 1.	0.76	80.8
4	15 Latex B	15 pph of styrene butyl acrylate (SBA) latex, Acronal S722 (BASF, Ludwigshafen, Germany), same pigments as board no. 1.	0.89	84.4
5	40 Clay	15 pph of the VAA latex. Pigments; 40 pph Setacarb® and 60 pph Capim NP delaminated clay (Imerys S.A, Paris, France).	0.83	70.3
6	N75 GCC	15 pph of the VAA latex. 100 pph Covercarb® 75, a GCC with narrow PSD.	0.87	73.4
-	Pre-coating	100 pph Hydrocarb® 60 and 13 pph of the VAA latex.		

2.2 Automatic Scanning Absorptometer

The ASA apparatus (*Figure 1, left*) consists of a motor-driven rotating table, a head box (5 mm x 1 mm slit), as well as a liquid supply and monitoring system enabling absorption rate measurement. The absorbed volume is monitored through a communicating capillary tube, where the movement of the meniscus is detected. It operates on the same principle as the Bristow Wheel, but is more effective, as one test-run with the ASA is equivalent to a series of Bristow Wheel tests at different speeds. Whilst moving outwards, the liquid head box scans the sample surface. The amount of the liquid absorbed by the sample surface depends on the absorbency as well as the contact time of the head box with the sample, which is inversely proportional to the rotating speed. The contact time may span a range from 5 ms to 10 s.

Using coloured liquid, the ASA makes a spiral stain (Figure 1, right). The testing liquid contains 12% (volume) of the condensed liquid dye, methylene blue, which is diluted in deionized water. It also contains 8.6 wt-% n-propanol in order to reduce the surface tension similar to flexographic inks.

2.3 Automatic Cobb Tester (ACT)

The ACT makes the conventional Cobb testing process automatic, measuring the amount of water absorbed by a specimen within a pre-set time period, e.g. 60 s. The sample is clamped against a porous glass disc that is filled with water. The water level inside the test chamber is monitored. In addition to the Cobb60 value, the details of the absorption dynamics are registered, such as absorption rate.



Figure 1:Left: ASA instrument. Right: Spiral stain from ASA measurement, the inner & outer circles correspond to longer and shorter contact time, respectively.

2.4 Surface roughness

Topographic maps of the board surfaces were captured with an OptiTopo instrument (Johansson 1999) which is based on a photometric stereo technique. The instrument captures image pairs through sequential illuminations at low angles from two opposite directions. Areas of 16 x 16 mm² with a lateral resolution of 15.6 μ m in both x and y directions were analysed. Surface roughness was calculated as standard deviation of the height about the mean value, with a focus on the lateral wavelength interval 0.06-1 mm.

2.5 Printing and print density

Full-scale printing was performed with an in-line flexographic press at Tetra Pak, Lund, Sweden. Print density of a solid cyan area was measured on five consecutive signatures. A spectro-densitometer, SpectroDens (TECHKON GmbH, Königstein, Germany), was used, calibrated against the board white and with the following settings: D50 illumination, 2° observer, polarization filter and density filter ISO E.

3 Results and discussion

3.1 Interpretation of absorption curves

The absorption curve (absorbed volume versus the square root of time) obtained from a series of measurements with a conventional Bristow Wheel consists of two parts

$$V(t) = V_0 + k\sqrt{t}$$
^[1]

where V_0 is the intercept of the absorption curve with the *y*-axis, *k* the slope of the curve and *t* the time of ink absorption. As explained by Bristow (1967), V_0 corresponds to the volume initially filling the surface roughness, while *k* expresses the absorption rate of the bulk.

The absorption curves given by the ASA, see *Figure 2*, are similar to the traditional Bristow curves. The absorption rate is calculated as the slope, and the impact from filling of the surface roughness is given by the intercept, both calculated from linear regressions in the time interval $t^{\frac{1}{2}} = 0.1$ -0.95 s^{1/2}. For sample 15 Latex B, a different time interval was used because of what appears to be a

wetting/absorption delay. In this case, the intercept ($V_0=4.9 \text{ ml/m}^2$) was estimated from the time interval $t^{\frac{1}{2}}=0.11-0.48 \text{ s}^{\frac{1}{2}}$ and the absorption rate was calculated from time interval $t^{\frac{1}{2}}=0.53-0.95 \text{ s}^{\frac{1}{2}}$.

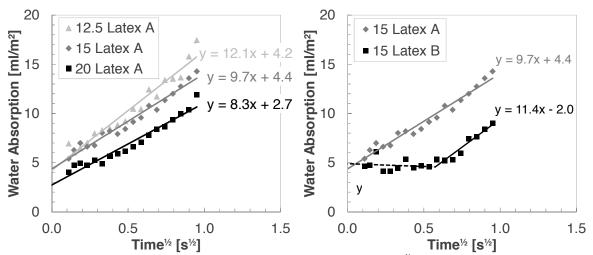


Figure 2: Example of absorption curves (absorption volume versus $t^{\prime/2}$) from the Automatic Scanning Absorptometer. Left: the boards having the same pigment blends but different latex contents. Right: the boards having the same pigment blends but different types of latex.

The absorption curves from the ACT measurements instead start at zero absorption volume, see *Figure 3*, probably as a consequence of the fact that the substrate surface approaches the liquid from above. One can also see that after the initial fraction of a second the ACT slope decreases. Hence, the absorption rate is calculated as the slope of the curves in the time interval $t^{\frac{1}{2}} = 2-8 \text{ s}^{\frac{1}{2}}$, and the intercept with the *y*-axis is extrapolated from the linear regressions in the same time interval.

Due to the different top coatings, the boards in *Figure 2 and 3*, exhibit different pore structures, porosities and contact angles, and, hence, different absorption behaviours. In the graphs to the left, all the boards have the same coating pigments, but different amounts of latex (type A). Increasing latex content leads to a less permeable pore structure, and so the absorption rates decrease. In the graphs to the right, boards with the same coating pigment and same latex content, but different latex types, are compared. Board 15 Latex B comprises of the latex type B, which results in a more hydrophobic coating surface as observed by an increased water contact angle. When measuring with the ASA instrument, *Figure 2*, the absorption dynamics of this board can clearly be divided into two regimes, revealing an absorption delay, which indicates that a pre-wetting process was needed before capillary driven absorption took place.

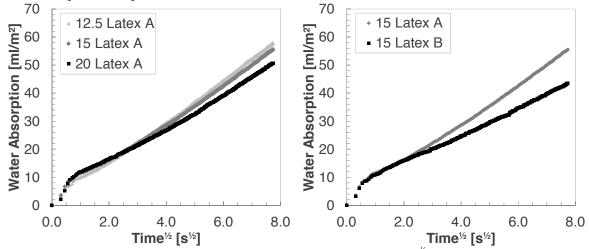


Figure 3: Examples of absorption curves (absorption volume versus t^{1/2}) from the Automatic Cobb Tester. Left: the boards having the same pigment blends but different latex contents. Right: the boards having the same pigment blends but different types of latex.

3.2 Comparison between ASA and ACT measurements

The absorption rates given by the ACT and ASA correlated fairly well with each other for all samples with one exception, 15 Latex B, see *Figure 4*. As previously explained, the board 15 Latex B has a relatively hydrophobic top coating, which requires pre-wetting as revealed by ASA absorption curve (black spots), *Figure 5*. Only after the wetting delay, the capillary driven absorption takes place. The ACT curve (grey spots) shows no wetting delay, rather a slow absorption during this period of time. With the aid of alcohol, the surface tension of the liquid in the ASA was lowered compared to the pure water used in the ACT testing. Hence, one does not expect the absorbency of the two liquids to comply fully and especially not when measuring a low surface energy paperboard as 15 Latex B. Besides, the ASA also showed better time resolution in the short time scale, $t^{1/2} < 1$ s^{1/2}.

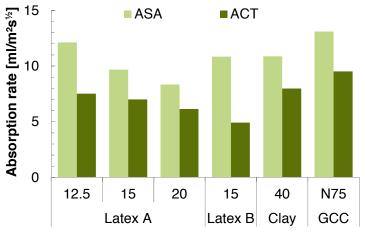


Figure 4: The absorption rates measured with the ASA and ACT respectively.

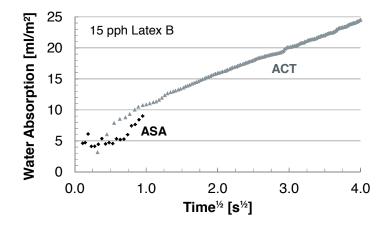


Figure 5: Example of dynamic absorption curves from the ASA and ACT for sample 15 Latex B.

Figure 6 depicts the surface roughness values measured with the OptiTopo and those estimated from the absorption curves of the ASA and the ACT measurement, using Eq. 1. An $R^2 = 0.73$ correlation between the OptiTopo and ASA was obtained. This implies that the contribution from filling of the surface roughness can indeed be estimated in the same way as with traditional Bristow curves. As to the ACT measurement, there is no such correlation observed with the OptiTopo measurements. In other words, with ACT one is not capable of measuring the roughness filling effect.

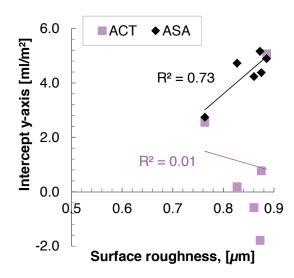


Figure 6:Intercepts with the y-axis from ASA and ACT absorption curves vs. surface roughness (OptiTopo height standard deviation, wavelength interval 0.06-1 mm).

3.3 Impact on print density

Figure 7 depicts the correlations of the print density with the absorption rate (left) as well as with the surface roughness (right) of the paper boards. As shown, the print density has a stronger correlation with the ASA measurement and weaker correlation with the ACT measurement due to involvement of an outlier. On the contrary, the correlation between print density and surface roughness is low.

It is important to point out that there is no pressure pulse applied to the liquid by either absorption measurement apparatus. This may differ significantly from flexographic practice where the printing plate in contact with the substrate will deliver an impression force. The pressure pulse may lead to forced wetting of even quite hydrophobic surfaces, such as the coating with Latex B. Hence, the prewetting phase that was seen with the ASA, or the slow absorbency that was seen with the ACT, may not cause any wettability issue when printing on this paper board. Similarly, flexographic ink formulations typically contain surfactant, which will aid surface wetting and addition of alcohol to the liquid in the ASA measurements helps mimic this.

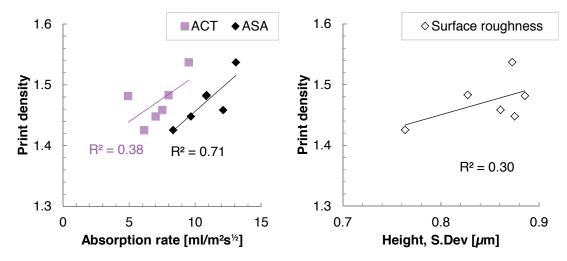


Figure 7: Correlations between print density and absorption rate (left) as well as with surface roughness (right).

4 Conclusion

Two relatively new absorption testing methods, ASA and ACT, have been tested and evaluated. Six pilot coated boards with specially designed top coatings were used in the study, which span a broad

145

range in pore-structure and surface chemistry, hence also a broad range of wetting and absorption behaviour. The results show that the absorption rates, given by these techniques, ranked the samples similarly, except for the one with a relatively hydrophobic coating. Moreover, compared to ACT, the ASA offers better time resolution in the short time regime $t^{\frac{1}{2}} < 1 \text{ s}^{\frac{1}{2}}$, which is relevant to a printing process and print quality. It is important to point out that there is no pressure pulse applied to the liquid by either measurement apparatus. This may differ significantly from flexographic practice where the printing plate in contact with the substrate will deliver an impression force. Our study also shows that, on these paper boards, the absorbency had a greater impact than the surface roughness regarding flexographic print density. However, one should not over-generalize this observation as the boards chosen in the study were specially designed to create large differences in absorbency. The results suggest that when surface roughness is not having a dominating impact on the print density, boards of higher absorbencies will also have higher print density. This is most likely linked to higher ink transfer.

Acknowledgements

The authors wish to thank KRK KUMAGAI RIKI KOGYO Co., Ltd. and FIBRO system AB for supporting with measurements and allowing us to use their equipment. BillerudKorsnäs, Stora Enso, and Tetra Pak are also acknowledged for financial support. The Research Institute of Sweden (RISE) and the Knowledge Foundation via VIPP Industrial Research College – Values Created in Fiber Based Processes and Products – at Karlstad University, Sweden, are also acknowledged.

References

Aspler, J., Cormier, L. & Manfred, T., 2004. Linerboard surface chemistry and structure affect flexographic print quality. In: *International Printing and Graphic Arts Conference*. Montreal, Canada, 4-6 Oct. 2004. PAPTAC: Montreal, Canada. pp. 167-177.

Aspler, J., Jordan, B., Zang, H. & Nguyen, N., 1998. Print Quality of Linerboard in Commercial Water-based Flexography. In: *1998 TAGA Annual Technical Conference*. Chicago, USA, 26-29 April 1998. TAGA office. pp. 749-781.

Barros, G. G., Fahlcrantz, C. M. & Johansson, P. A., 2005. Topographic Distribution of UnCovered Areas (UCA) in Full Tone Flexographic Prints. *TAGA Journal*, vol. 2, no. 1, pp. 43-57.

Barros, G. G. & Johansson, P. A., 2006. Prediction of uncovered area occurrence in flexography based on topography: a feasibility study. *Nord. Pulp Paper Res. J.*, vol. 21, no. 2, pp. 172-179.

Bristow, A., 1967. Liquid absorption into paper during short time intervals, *Svensk Papperstid.*, vol. 70, no. 19, pp. 623-629.

Johansson, P.-Å., 1999. *Optical homogeneity of prints*. Doctoral Thesis, KTH - Royal Institute of Technology, Stockholm, Sweden.

Lagerstedt, P. & Kolseth, P., 1995. Influence of surface energetics on ink transfer in flexo printing. In: 23rd International Research Conference of iarigai. Paris, France, 17-20 Sept. 1995. pp. 269-299.

Sheng, Y. J., Shen, W. & Parker, I. H., 2000. Importance of the substrate surface energetics in water-based flexographic printing. *Appita J.*, vol. 53, no. 5, pp. 367-370.

TAPPI, 1998. T 441 om-13. Water absorptiveness of sized (non-bibulous) paper, paperboard, and corrugated fiberboard (Cobb test). Test method TAPPI/ANSI.

Wågberg, P. & Wennerblom, A., 1992. A Correlation between Results Achieved with an Optical Profile Tester, Conventional Paper Evaluation and Printability. In: *1992 International Printing and Graphic Arts Conference*. *Pittsburgh, USA, 18-21 Oct. 1992. TAPPI Press: Atlanta, USA.* pp. 187-196.

The Choice of a LCIA Method from a Practitioner's Perspective – an Example of Laminating Films in the Printing and Packaging Industry

Kirsten Radermacher^{1*}, Ulrich Jung¹, Joachim M. Marzinkowski²

¹Department of Print and Media Technologies, University of Wuppertal, Rainer-Gruenter-Strasse 21, 42119 Wuppertal, Germany

²Department of Safety Engineering, University of Wuppertal, Rainer-Gruenter-Strasse 21, 42119 Wuppertal, Germany

E-mail: radermacher@uni-wuppertal.de, ujung@uni-wuppertal.de, marzinko@uni-wuppertal.de

**Corresponding author*: Kirsten Radermacher, Tel.: +49 (0)202-439 1313, Fax +49 (0)202-439 1310

Short Abstract

The LCIA method 'ILCD2011' was developed and recommended for practitioners to be the preferable one from a scientific point-of-view. This recommendation is independent from the product system in question and solely based on scientific knowledge. In this paper, the LCIA methods 'EDIP2003', 'CML2001', 'ReCiPe2008' and 'ILCD2011' were investigated from a practitioner's view-point. We analyzed whether the choice of a LCIA method should be product-related. Is the choice of the ILCD method eligible to assess the impact of laminating films? We found out that this method is also suitable for products of the printing and packaging industry which are mainly organic in nature.

Keywords: life cycle assessment, impact assessment, plastics, laminating film, toxicity

1. Introduction and background

The life cycle assessment had emerged to be a comprehensive approach, which is accepted by experts, researchers and industry for assessing the environmental impact of products. However, the research work is ongoing in concern to special issues. An important task is the choice of an impact assessment (LCIA) method. It was found that in some impact categories, the LCIA methods differ fundamentally in their theoretical background and influence the LCA results (Dreyer et al, 2003; Pant et al., 2004; Radermacher et al., 2013b). These differences are particularly recognized in the toxic impact categories. Due to include these impact categories, a full LCA requires that the LCIA method is chosen carefully (Radermacher et al., 2014). The researchers, who worked together in the European study initiated by EC-JRC (EC-JRC, 2010, 2011a,b; Hauschild et al., 2013), pointed out that the LCIA methods available are preferable for some impact categories but not for all. Thus, the EC-JRC generated a new LCIA method out of the existing ones representing most closely the reality of intake and transportation of substances in the environment and humans from a scientific point-of-view. They called it 'ILCD2011'. This is the newest LCIA method and supposed to be the most suitable one. Due to evaluate whether 'ILCD2011' is also appropriate from the practitioner's perspective, we analyzed the general focus of 'ILCD2011' and the processes emphasized in comparison to various other LCIA methods. This research questioned how the choice of impact assessment methods is able to change the statement in a comparative analysis of products in LCAs and whether contrary processes and process types are prioritized in a specific product system of the printing and packaging industry. For this purpose, the comparative LCA of the polypropylene- and cellulose-based laminating film, were used according to Radermacher et al. (2014). How could a LCA practitioner be sure that an appropriate LCIA method for the product system in question was chosen?

2. Materials and Methods

In the study, a set of LCIA methods was chosen including 'EDIP2003' (Wenzel et al., 1997; Hauschild and Wenzel, 1998; Hauschild and Potting, 2005; Potting and Hauschild, 2005) and 'CML2001' (Guinee et al., 2001) which had been the commonly used LCIA methods for a long time and 'ReCiPe2008' (Goedkoop et al., 2009) which is one of the newest developments considering recent knowledge.

The overall results of a comparative analysis applying these LCA methods give a general view on the effect of the choice of a LCIA method. Therefore, a LCA of two laminating film types was performed in accordance to Radermacher et al. (2013a, 2014). We investigated if the statements in the LCA study could be reversed by using another LCIA methods and which of the impact categories are mostly affected. These impact categories were analyzed in more detail in the following.

The detailed analysis was concentrated on the toxic assessment. The toxic impact categories are those categories which are able to present the differentiation of LCIA methods most clearly. In ILCD, the USEtox method is recommended for freshwater ecotoxicity and human toxicity. The marine ecotoxicity and terrestrial ecotoxicity is not included in 'ILCD2011' because none of the existing LCIA methods fulfil the requirements from a scientific point-of-view reported in EC-JRC (2010, 2011a,b). Thus, they are not considered in this paper. The detailed analysis of the freshwater toxicity and human toxicity was separated into two parts: (a) A priority analysis of substances emitted by the product system and (b) the analysis of life cycle stages and process types creating these emissions.

In the first part of the analysis, the substances emitted by the product system were grouped by their chemical properties. The substances mentioned in the published lists of characterization factors for 'CML2001' (v4.2), 'EDIP2003' (version unknown), 'ReCiPe2008' (v1.08) and 'ILCD2011' (USEtox v1.10) were considered. The first step of the detailed analysis helps to visualize which of these substance groups are broadly prioritized in the various LCIA methods.

In the second part, the life cycle stages und process types were analyzed to investigate the environmental potential of laminating films applying the LCIA methods. Therefore, the Multi-level approach of Radermacher et al. (2014) was utilized. A first impression about the influence of the LCIA method chosen could be taken from the analysis of the life cycle stages (level 1); the analysis on level 2 and level 3 reveal which of the process types and process groups contribute mainly to the toxic potential: the product-related or the supporting processes. The methods are implemented in the life cycle assessment tool 'OpenLCA' (v1.4.0/1) and utilized in this part of the analysis.

3. Results and Discussion

The overall results of the comparative life cycle assessment of laminating films in accordance to the report in Radermacher et al. (2013a, 2014) is shown in Figure 1. Here, the differences of LCIA methods 'EDIP2003', 'CML2001', 'ReCiPe2008' and 'ILCD2011' were identified for various impact categories. 'max. Delta' implies the gap between the methods with lowest and highest ratio between the impact potential of the product alternatives. Generally, the choice of a LCIA method does not affect the ranking of laminating films in the LCA study meaningful. However, the results were reversed with a gap of 4% in the freshwater ecotoxicity if ReCiPe would be chosen in the impact assessment.

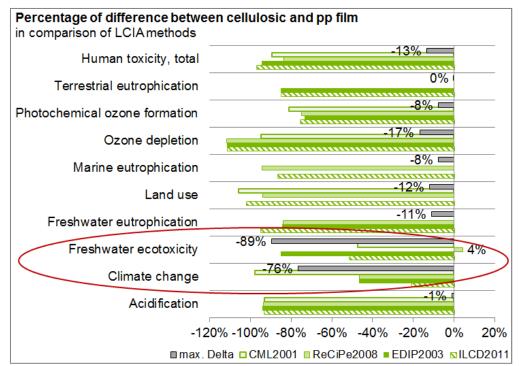


Figure 1. List of the comparative results of the LCA on cellulosic and pp-based laminating films in accordance to Radermacher et al. (2013a, 2014): Percentage of differences for each impact category

Great differences of the comparative LCA results are found in the toxic assessment and the climate change potential (GWP). The comparative LCA study including one product manufactured from renewable and one which consists of non-renewable materials is mainly influenced by the calculation of biogenic carbon dioxides. Thus, the effect on the global warming potential could be significant. In accordance to the LCI method pack of OpenLCA (v1.5.3), the carbon dioxides consumed in the wood growing are credit to the product with a positive effect factor in 'ILCD2011'; not in the other methods. This fact changes differentiating results in the method comparison. Another great difference is located in the freshwater ecotoxic potential. This impact category is part of the detailed analysis below.

In the first step of the detailed analysis, the substances were grouped in 15 clusters of different chemical nature and toxic potential: aliphatic compounds (A1: alcohols, aldehydes, ketones, oxides, A2: chlorated, bromated and fluorated aliphatic compounds; A3: organic acids, esters, ethers; A4: Amines, Amides, quaterny ammonium compounds; A5: (cyclo-)alkanes, alkenes, alkines, paraffins), benzene derivates (B1: chlorated, bromated, fluorated and iodized aromatic compounds; B2: aromatic nitrogen compounds; B3: other aromatic and poly-aromatic compounds not found in B1, B2 and B4; B4: aromatic oxygen compounds), anorganic compounds (I1: acids and non-metallic oxo-compounds; I2: alkaline compounds, betaines, salts; I3: oxidants and reducing agents), F (fungizides/herbizides), G (gases), M (metals) and S (organic sulphuric compounds and thiophosphoric acid compounds). A scheme of the clustering is shown in Figure 2.

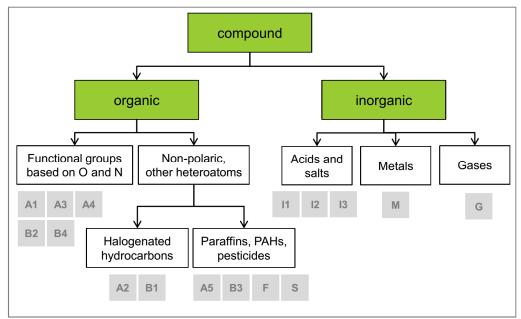


Figure 2. Substance groups built from the chemical properties of emissions

On the basis of the substance groups, the inventory data of the laminating film 'pp-based film' were exemplarily assigned to the characterization factors of 'CML2001', 'EDIP2003', 'ReCiPe2008' and 'ILCD2011'. Priorities of substance groups will present results about the specialty of the LCIA methods (cf. Figure 3 and 4). A specific handling in the analysis is needed for the aquatic ecotoxicity: The aquatic ecotoxicity is distinguished in marine and freshwater in CML, ReCiPe and ILCD; EDIP reports the aquatic toxicity in general. This situation was considered in the interpretation of the results.

The priority analysis of the LCIA methods for marine and freshwater toxicity is presented in Figure 3 in percentage of the total toxic potential for each LCIA method if the substance group of metals is ignored which is high in EDIP, ILCD and CML and medium in ReCiPe in the freshwater ecotoxcity.

In CML, besides metals, inorganic substances are predominant in the marine ecotoxicity whereas the freshwater toxicity includes broadly organic substances. The substance groups prioritized in CML (freshwater) are similar to these of EDIP. However, in combination, the inorganic substances are mainly contributing to the toxic potential in CML. ReCiPe is equal in the priority of marine and freshwater toxicity. In both cases, the same inorganic substance group is included besides metals. The freshwater toxicity in ILCD is concentrated on metals, fungicides/herbicides and (poly)-aromatic benzene derivates.

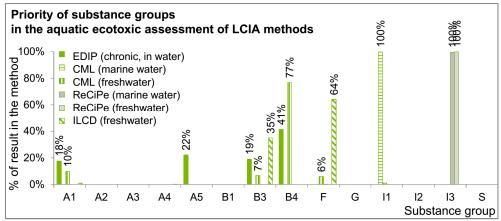


Figure 3. Priority analysis of substances in the aquatic ecotoxicity of EDIP, CML, ReCiPe and ILCD for marine and freshwater; Metals are not included

In the human toxicity (cf. Figure 4), besides metals, ReCiPe prioritizes inorganic substances, whereas EDIP is focussed on organic substances and gases, as in CML. Additionally, CML prioritize inorganics. ILCD shows a balanced consideration of different organic substance groups. Metals have a high percentage of consideration in CML and ILCD and low percentage in EDIP and ReCiPe.

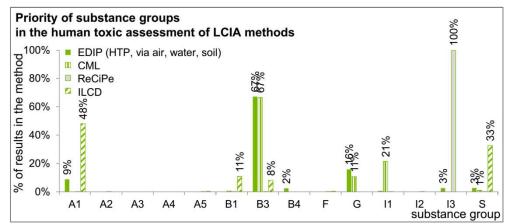


Figure 4. Priority analysis of substances in the human ecotoxicity of EDIP, CML, ReCiPe and ILCD; Metals are not included

In the second part of the analysis, the main contributing life cycle stages and process types for the product system 'cellulose-based laminating film' were investigated applying the different LCIA methods (cf. Figure 5 and 6). The figures shown include six graphics each with three levels of detail for the impact categories 'Human toxicity' and 'Aquatic ecotoxicity'. The three-level-approach was implemented following the Multi-level approach presented in Radermacher et al. (2014): In level 1, the environmental impact is partitioned into life cycle stages. Level 2 presents the process types of the film manufacturing step incl. their upstream processes. The impact potential is grouped in level 3 according to the material origin where the substances are emitted.

The level-1-analysis illuminates that the film manufacturing is the main contributor for the human toxicity and the aquatic ecotoxicity in EDIP, ILCD and ReCiPe. In CML, the film manufacturing contributes mainly to the human toxicity; whereas, the pulp production is high in the aquatic ecotoxicity. More than 80% in CML is caused by the upstream processes. On level 2, there are some similarities in the partitioning of process types between CML and EDIP. In all LCIA methods auxiliaries and energy are important process types. Taking the results of level 3 into account, in the human toxicity, EDIP emphasizes processes of fuels, organics and metals. The results of ILCD are dominated by metals, fuels and inorganics. CML shows priorities on fuels, metals and organics. Metals, organics and fuels are predominant in ReCiPe. Even in the aquatic ecotoxicity potential, the dominating process types in ILCD are metals in addition to fuels and organics. Others, fuels and organics are relevant in EDIP. 'Others' is mainly caused by unspecified burdens from waste treatment. A similar contribution is found in the aquatic toxic potential in CML. ReCiPe focusses on organics, others, fuels and metals. The end-of-life phase is a great contributor of the aquatic ecotoxicity in ReCiPe. This priority is continued in Level 2 and 3: the process type 'direct emissions' and the process groups 'others' and 'organics' include this waste treatment of the product.

A specific view on the auxiliaries and their upstream processes in the film manufacturing stage enables to confirm a general conclusion for the toxic assessment: EDIP focusses on fuels of energy production processes and organic materials, like auxiliaries and waste. ILCD highlights processes of metallic compounds. These are e.g. auxiliaries in the film manufacturing counting 34% out of the 47%-portion metals in the aquatic ecotoxicity and 53% out of 66% in the human toxicity. CML prioritize fuels and metallic compounds from energy processes. Similar to ILCD, ReCiPe emphasizes metals of the auxiliaries and other inorganics in the human toxicity. Metals and fuels of energy processes are relevant in the aquatic toxicity in both of the LCIA methods.

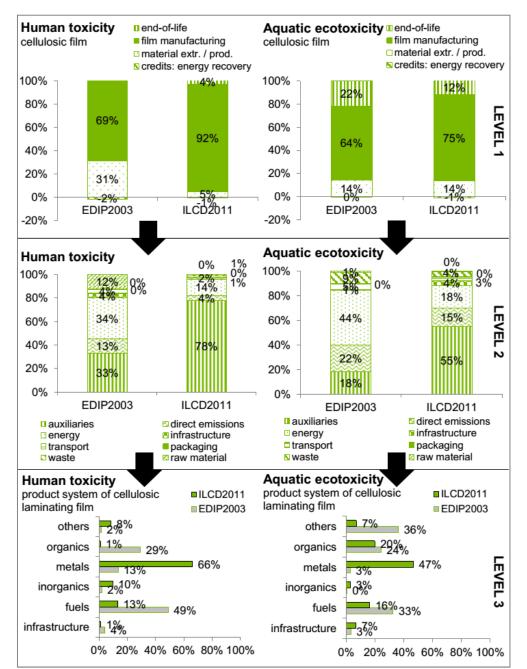


Figure 5. Multi-level toxic assessment of the cellulosic film applying EDIP2003 and ILCD2011 in OpenLCA 1.4.1 in accordance to the 'Multi-level' approach of Radermacher et al. (2014)

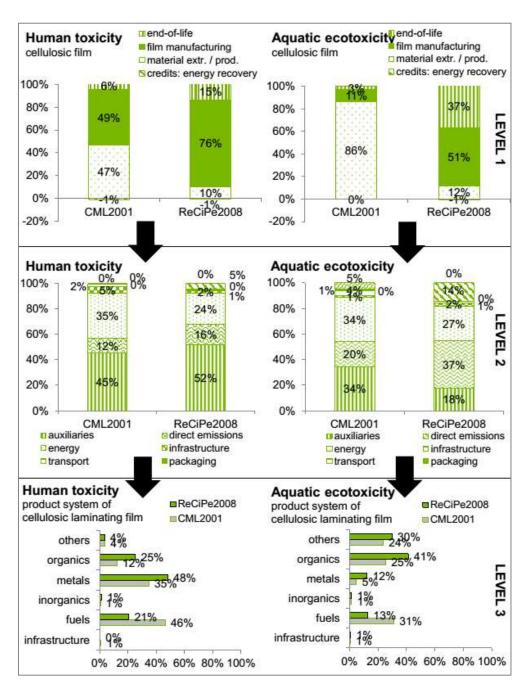


Figure 6. Multi-level toxic assessment of the cellulosic film applying CML2001 and ReCiPe2008 in OpenLCA 1.4.1 in accordance to the 'Multi-level' approach of Radermacher et al. (2014)

4. Conclusions

The investigation of the impact of products is a great challenge in the life cycle assessment. Different LCIA methods are available and could be chosen conducting LCA studies. The extension of the lists of characterisation factors are still in progress and have been implemented in the newest versions of LCA tools. Practitioners aim to choose an appropriate LCIA method. The newest recommendation is 'ILCD2011'. It emerged from the results of a study which questioned if the existing LCIA methods are appropriate from a scientific point-of-view (EC-JRC, 2010, 2011a,b; Hauschild et al, 2013). In our study, we analyzed the LCA results of two laminating film types to answer the main research question: Is the choice of 'ILCD2011' appropriate from a practitioner's perspective? Should the LCIA method be chosen depending on the product system in question? For this purpose, 'EDIP2003', 'CML2001' and 'ReCiPe2008' were chosen to be compared with 'ILCD2011'. The gaps between the potential

impacts of the product alternatives are meaningful in the climate change and the toxic assessment. In reference to former publications, the toxic impact categories reveal the main differences between LCIA methods most clearly. Further investigations were, thus, concentrated on these toxic categories.

The research question in this paper was observed from a highly aggregated perspective to clearly present the general focus of the LCIA methods. Previous publications discuss the differences in the single substances considered for different product examples. It is known that LCIA methods vary in the substances considered and could affect the LCA results decisively. Another reason for differences is the currency of the effect factors implemented. So, analyses on the LCIA methods constantly represent the current situation. In this paper, we chose the aggregated perspective to overcome the time-dependency of the research results and to achieve a robust study.

The analysis was separated into two parts: a priority analysis of the emissions emitted by the product system and a detailed analysis of the life cycle stages, process types and process groups where the emissions occur. In a first step, the method comparison based on the prioritized groups of substances emitted from the product system 'polypropylene-based laminating film'. To achieve this, emissions of the laminating film were assigned to the emissions reported in the lists of characterisation factors published. Additionally, we generated clusters of substances to illuminate which kinds of emissions are prioritized in the LCIA methods. The clustering was based on information about the chemical properties of the substances and their toxic effects. The metals have an extraordinary position in all of the LCIA methods. However, the priority is somewhat different. Besides metals, EDIP emphasizes organic compounds. Inorganic substances are predominant in ReCiPe. Organic substances are important in the freshwater ecotoxicity in CML and ILCD. A combined consideration of marine and freshwater toxic potential applying CML reveals priorities of groups with inorganic compounds, whereas EDIP are exclusively based on organic substance groups besides metals. This result leads to a preferred comparison of ILCD and EDIP in a second step matching the organic nature of the product system in question. Additionally, the LCA results of ReCiPe and CML were shown. In the second step, the analysis was concentrated on the life cycle stages and process types in which the emissions occur. The 'Multi-level' approach by Radermacher et al. (2014) was utilized to reveal main contributors on different levels of detail. With special focus on EDIP and ILCD, process types and process groups prioritized are product-related in both cases: The film manufacturing including energy and auxiliaries is the main contributor. In level 3, the metals emphasized in ILCD are mainly generated by auxiliaries in the film manufacturing. This is the main contributing process group in the main contributing life cycle stage. EDIP presents a great portion of fuels and organics. This impact assessment method is focussed on energy-related processes, like energy production and transportation. Metals are of smaller interest in EDIP than in ILCD. CML and EDIP have some similiarities in the contributing proces groups in level 3; great differences between these LCIA methods were found in the consideration of metals. The impact potential in ReCiPe is influenced by the end-of-life phase of the laminating film in level 1. This priority is continued on Level 2 and 3 of the 'Multi-level' approach. It leads to a high ratio in the process group 'organics'. In the aquatic ecotoxicity, 37% out of 41% is caused by the end-of-life treatment. In general, this dependency of the LCA results leads to a reversed product ranking in the comparative study in Figure 1.

In general, a clear prioritization of supporting processes was not found in any of the LCIA methods in our research. Infrastructure (buildings, machines and transport) and packaging do not contribute decisively to the total toxic impact.

As a conclusion, the recommended ILCD method was developed from a scientific point-view to assess most suitable the toxic impact in the environment and on humans. It was questioned if this proceeding conflicts with a product-related perspective of LCA practitioners. This assumption could not be confirmed. The balanced consideration of organic and inorganic substances in the ILCD method is product-related. The highlight of organic materials is also possible if the product system is mainly based on organic materials. This is the case in the product system 'polypropylene-based laminating film'. The ILCD method seems to be an eligible one, also for products of the printing and packaging industry whose material origin is mainly of organic nature.

References

Dreyer, L.C., Niemann, A.L. and Hauschild, M., 2003. Comparison of three different LCIA methods: EDIP97, CML2001 and eco-indicator 99. *International Journal of Life Cycle Assessment*, 8(4), pp. 191-200.

European Commission-Joint Research Centre EC-JRC (ed.), 2011. Framework and requirements for Life Cycle Impact Assessment models and indicators, International Reference Life Cycle Data System, European Union, ILCD Handbook (Part II). <u>http://lct.jrc.ec.europa.eu/assessment/publications</u>.

European Commission-Joint Research Centre EC-JRC (ed.), 2011. *Recommendations for Life Cycle Impact Assessment in the European context, International Reference Life Cycle Data System*, European Union, ILCD Handbook (Part III). <u>http://lct.jrc.ec.europa.eu/assessment/publications</u>.

European Commission-Joint Research Centre EC-JRC (ed.), 2010. Analysis of existing Environmental Impact Assessment methodologies for use in Life Cycle Assessment, International Reference Life Cycle Data System, European Union, ILCD Handbook (Part I). <u>http://lct.jrc.ec.europa.eu/assessment/publications</u>.

Goedkoop, M., Heijungs, R., Huijbregts, M., De Schryver A., Struijs, J. and van Zelm, R., 2009. *ReCiPe 2008:* A life cycle impact assessment method which comprises harmonised category indicators at the midpoint and the endpoint level, Technical report.

Guinée, J.B., Gorree, M., Heijungs, R., Huppes, G., Kleijn, R., de Koning, A., van Oers, L., Sleeswijk, A. Wegener, Suh, S., Udo de Haes, H.A., de Bruijn, H., van Duin, R. and Huijbregts, M. A.J., 2001. *Life cycle assessment – an operational guide to the ISO standards*.

Hauschild, M. and Potting, J., 2005. Spatial differentiation in Life Cycle impact assessment – The EDIP2003 methodology, Technical Report 80-2005. Danish Environmental Protection Agency. Hauschild, M. and Wenzel, H. (1998), Environmental Assessment of Products: Volume 2: Scientific background, University Press, Cambridge (GB).

Hauschild, M.Z., Goedkoop, M., Guinee, J., Heijungs, R., Huijbregts, M., Jolliet, O., Margni, M., De Schryver, A., Humbert, S., Laurent, A., Sala, S. and Pant, R., 2013. Identifying best existing practice for characterization modeling in life cycle impact assessment. *International Journal of Life Cycle Assessment*, 18(3), pp. 683-697.

Hauschild, M. and Wenzel, H., 1998. Environmental Assessment of Products. Volume 2: Scientific background. Capman & Hall.

Pant, R., Hoof, G. van, Schowanek, D., Feijtel, T.C.J., Koning, A. de, Hauschild, M., Pennington, D.W., Olsen, S.I. and Rosenbaum, R., 2004. Comparison between LCIA methods for aquatic ecotoxicity and a product environmental risk assessment – Insight from a detergent case study within OMNIITOX. *International Journal of Life Cycle Assessment*, 9(5), pp. 295-306.

Potting, J. and Hauschild, M. (2005), Background for spatial differentiation in LCA impact assessment – The EDIP2003 methodology, Technical Report 996-2005. Danish Environmental Protection Agency.

Radermacher, K., Jung, U. and Marzinkowski, M.J., 2014. Life cycle oriented analysis of laminating films for the printing and packaging industry using a Multi-level approach. *Journal of Print and Media Technology Research* 3(3) pp. 163-174.

Radermacher, K., Jung, U. and Marzinkowski, J. M., 2013a. A life cycle approach to environmental aspects in the printing and packaging industry. In: Enlund, N. and Lovreček, M., eds. *Advances in Print and Media Technology, Vol. 40*, IARIGAI, pp. 169-177.

Radermacher, K., Jung, U. and Marzinkowski, J. M., 2013b. Analysis of life cycle impact assessment methods for products in the printing and packaging industry. *5th International Scientific Conference 'Printing Future Days 2013*'. Institute of Print and Media Technology, pp. 191-196.

Wenzel, H., Hauschild, M. and Alting, L., 1997. Environmental Assessment of Products – Volume 1: Methodology, tools and case studies in product development. Kluwer Academic Publishers, Boston/Dordrecht/London.

Does the use of Black Ink still comprise the "Darkest" Issue of CMYK Printing?

Y. Kuznetsov, M. Ermoshina

North-West Institute of Printing of the St. Petersburg State University of Technology and Design

E-mail: yurivk@mail.ru, tel.: +7 (911) 226-88-03

Short Abstract

The relationship, degree and effect of the variety of black ink functions use in CMYK printing is discussed on the background of prepress evolution from analogue scanners of 60ies up to precise digital color control of today. There are also presented the results of comparative colorimetric analysis of CMY and CMYK printing revealing the black ink particular effect of expanding the CMY gamut by providing the chromatic colors which aren't available for any combination of the other three process ones.

Key words: color, printing, gamut

1. Introduction

Looking back in the latest history of graphic technology developments one can find that thirty-forty years ago there were a number of scientifically approved recommendations on direction and degree of tone and color values correction for print quality improvement. However, even at the times of color electronic prepress, there was a lack of means for proper control in providing the desired variations. In 70ies of last century it was used, for example, to indicate in advertising specification of a scanner its ability to replace the CMY achromatic component by the black ink as "up to 75%" (against the 65% of the competing model). The clear knowledge of what had to be done with an image data faced, as well, the lack of means to realize the task in some other relations.

The digital image processing of today allows for practically unlimited print parameter variation in any direction with the discretion of just 25 square microns of ink coverage. However, quite a contrary situation of adequate resources but lacking in knowledge of what should be done is often met and the need arises of additional research or training which could substantiate the recommendations and performing methods for effective use of such precise, recently appeared control facilities [Kuznetsov, 2012].

There is variety of purposes and reasons of black ink use in CMYK printing. It can be in different degree applied for reproduction of achromatic, chromatic colors as well as the achromatic component of chromatic ones. Its use can also differentiate from the vast, stationary image area to sharp edges and fine details. So, there is theoretically infinite continuum of CMY to CMYK transformations which can result in the same or better colorimetric print values.

The beginning of K-ink use within the CMY triad stems from the times of photoengraving, camera prepress. The facilities of black control according to certain rendering intent and, especially, in isolation from its other effects on resulting color were rather restricted. However, the mostly heuristically found, scanty collection of black ink settings is until now used in wide practice. One of the reasons is in some isolation of numerous participants (publishers/advertisers, prepress operators, quality managers, printers...) from each other. Lack of facilities or time for finding the optimal adjustments which would match the job/process specifics makes them to follow the narrow path of guaranteed standard parameters (Euroscale, SWOP...) or of settings stipulated by the available ICC profile.

The other reason of non-optimal black ink use is in vague interpretation of its settings and their relationship essence in "black boxes" of prepress software applications or commenting manuals. When

appealing to their "help" option the user is sometimes sent to get an advice from a printer. In this relation E. Enoksson [Enoksson 2004] notes, for example, that only about a quarter of the Swedish print houses have people ever heard of the UCR and GCR functions of Photoshop.

Not so much "help" the user can get from academic sources. Problems start here from providing the proper definitions for these functions because of similar sense of their abbreviations meanings. For example both Under Color Removal and Grey Component Replacement indicate in fact the "removal of chromatic inks (CMY) achromatic component by replacing it with the black (K) one". Meanwhile the "Complete Color Glossary" defines the UCR procedure as related just to the dark neutral colors [Southworth, Southworth 2004]. In "Handbook of Print Media" one can find the attempt to distinguish UCR, GCR, UCA functions by the examples of varying just the volume of CMY achromatic part replaced by the black ink [Kipphan, 2001], though this volume can be varied within each of these procedures as well. At last, the "Digital Color Imaging Handbook" stands out GCR as a "generalization" of UCR and K addition [Bala, 2003]. There were also attempts to modify UCR under the names of such procedures as PCR (Programmed Color Removal), ICR (Integrated Color Removal) etc. and the new names are still proposed [Enoksson 2004].

The number of other explanations of fourth ink application suffers from mixing the purposes and methods of these purposes destination on the background of vast variety of CMYK combinations continuum. So, it is not out of place to separately discuss the following black ink functions:

- replacement the achromatic component of three chromatic inks combination;
- reproduction the image achromatic colors;
- expanding the print color gamut.

2. Relationships of black and balanced CMY in achromatic component of chromatic inks combination

2.1 Volume of replacement

This parameter is often used to be illustrated by the diagram of the kind presented on figure 1, where figures 1b and 1c show the examples of partial (50)% and complete (100%) volumes of achromatic share replacement.

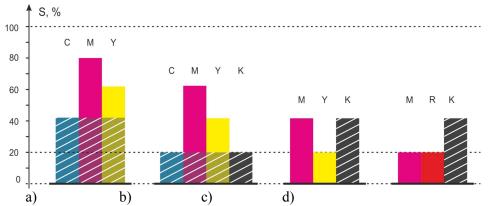


Figure 1. Providing formally the same chromatic color: without black ink (a); at 50% and 100% CMY achromatic share (hatched) replacement by black (b, c); with further removal of equal M and C on behalf of an orange ink in HiFi printing

This variation is historically related to UCR function which is first of all used for the darker image areas because of total ink limitation. In fact, it can be varied from 0% up to 100% independently of the given pixel brightness, i.e. in the highlights, middle tones or shadows.

Volume and range of $(CMY)_{min}$ replacement is, except of ink limit, stipulated by the number of other technological, economic, operating and image quality considerations including:

- ink consumption costs;
- fidelity and stability of the grey balance within a run;
- color disbalance due to rosettes geometry variation, as well as moiré and rosettes visibility [Daels, Delabastita, 1994];
- gamut mapping intents;
- use of inks which colors are complementary to that of CMY inks in Hi-Fi printing, etc.

Nature of these reasons is well known or described in hereinabove referred literature while the last one is cleared up by the figure 1d. It shows that the 100% removal of one of CMY inks is compulsory over all tonal range to make worthwhile the use of an additional ink of opposite, complementary color (red or orange one in this example) to expand the print color gamut. This makes it clear too that the screen of complementary color can safely use the angle of its corresponding process ink for the latter should be completely removed in particular image area to get higher chroma.

2.2 Varying the replacement volume within the tone range

The volume variation of this replacement within the tonal range can be illustrated by diagram of figure 2, where the straight line 1 corresponds to printing of the whole grey scale of color image exclusively by CMY inks. For simplicity the balanced CMY inks amount changes along lines of this graph in equal proportion (C = M = Y) thereby related to the use of some "ideal inks".

With taking the line 1 for a reference the other curves of figure 2 demonstrate the possible variants of achromatic CMY and K-ink volumes relationship along the tone range. Thus, curve 2 indicate at its upper point the $(CMY)_{min}$ withdrawal of 40% where initial C = M = Y = 100% is replaced by combination of C = M = Y = 60% and K = 40% at 220% of ink total. Following to lighter areas of this curve the use of black ink is gradually reduced to 0% at middle tone. Starting from C = M = Y = 50% and until the white point the achromatic component is again reproduced only by CMY.

Curve 3 illustrates the constant replacement value of 40% along the whole grey scale. Here, for example, middle tone is presented by C = M = Y = 30% and K = 20% with the latter comprising 40% of achromatic value defined at this point by C = M = Y = 50% of the reference curve 1.

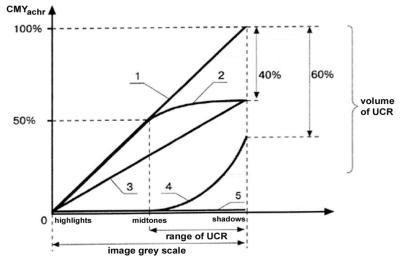


Figure 2. Variation of balanced CMY and K values along the image grey scale: 1 – without K;

2 – gradual reduction of CMY replacement by K from 40% in shadows to 0% in middle tones;

3 - with 40% constant volume of replacement along the whole scale;

4 – with gradual growth of CMY replacement by K from 60% in shadows up to 100% in middle tones; 5 – without CMY along the whole scale Gradual replacement increase from 60% in the darkest areas up to 100% in middle tones is shown by curve 4. It also indicates the reproduction of lighter part of the scale exclusively by black ink. At last, the curve 5 coincides with horizontal axis of the diagram and relates to complete replacement

volume of 100% along the whole scale. It corresponds to the so called "binary chromatic + black" strategy where CMY inks can't be altogether found in any chromatic area of a print.

As result, this diagram describes two different dimensions of $(CMY)_{min}$ withdrawal the first of them relating to volume of K-ink use while the other one indicating the location of its certain volume within the tone range. It's however used to separate the whole this continuum just on two strategies: UCR and GCR relating the first of them to curves with shoulder, of the kind of curve 2, and the latter to concaved ones like curve 4. The freedom of choice and manipulating these curves faces in wide practice the difficulties which are concerned of their affecting the printing system color profile. Coming from one curve to the other inevitably changes color of the same print area due to variations in halftone dots overlap, their summary perimeter, etc. resulting in the shift of an ink trap, physical and optical dot gain.

2.3 One else function of black ink

The black ink spectral reflectance is characterized by the sharp growth in the near infra red band. That makes it much more visible under corresponding intensive illumination than C, M or Y and thereby provides the facility of non-costly K-ink use for print security purposes.

It's possible to modulate the $K = (CMY)_{min}$ component by the auxiliary image of the kind of well known Eiffel tower. After such modulation the part of K which exceeds this image signal is returned to the balanced CMY to keep the color of basic print non-touched. Such auxiliary image stays to be concealed from the viewer under any day light but somehow reveals in the strong red lightening.

3. Achromatic colors reproduction

Providing the maximum of reproducible grey levels was outlined by R. Hunt [Hunt, 1997] and stipulates the priority of black ink use in color print. The whole achromatic component can be considered as the basic one for image formation while the CMY inks as auxiliary ones, i.e. responsible just for the image chromaticity. Moreover, due to well known degradation of color vision sensitivity with decreasing of detail angular, spatial dimension, the small details and high contrast sharp transitions can also be reproduced with greater share of black or even, as it's done in color TV, completely achromatic.

The effect of K and accurately balanced CMY combined use for b/w image is especially apparent when the latter is simultaneous printed on the same sheet with multicolor ones. Our research based on UCR offset atlas [Avatkova, 1987] has shown, for example, that at ink limit 290% the addition of C = M = Y = 62% to K solid expands the grey scale optical density range on about 0.3.

Various ways of such combining are formally possible. Balanced CMY inks and K ink amounts can be uniformly distributed along the grey scale or each of them alternatively concentrated in its highlights or shadows [Kuznetsov, 2002].

It has to be assumed that above considerations to certain extent artificially separate the formation of print achromatic color and achromatic component of chromatic one. In prepress software these functions are merged in each other and altogether governed by the same curves. Moreover, achromatic color is also influenced by the so called Under Color Addition (UCA) settings. However, the discerning of these functions may look purposeful in creating or operating the updated prepress software of foreseen future.

4. Expanding the print color gamut

Discussed above combined use of K and CMY inks expand the gamut in close vicinity to L axis of LCh space where colors can still be considered achromatic with taking into account their permissible delta E, for example of 5 units, from neutral as illustrated by figure 3.

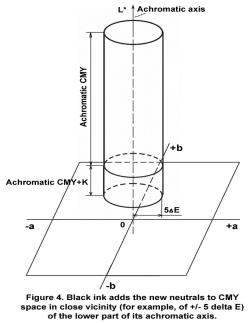


Figure 3. Black ink adds new neutrals to CMY color space in close vicinity of the lower part of its achromatic axis

At the same time, it's practically used to add some K amount to C, M or Y solids to get new, darker chromatic colors. However the gamut expanding due to chromatic colors which exclusively appear with applying the fourth, black ink wasn't separately considered in literature. Moreover, one can find the contrary statement of color gamut reduction with black ink introduce [Bala, 2003]. Nevertheless, the issues of print gamut expanding by the use of intensive inks, addition to CMY process colors of their complimentary RGB ones within the so called Hi-Fi Color concept, etc. were widely discussed in last decades.

Our research was provided by comparative analysis of CIE Lab colorimetric data from offset atlas [Avatkova, 1987], as well as from test step wedges especially produced by variety of three and four inks combinations on digital printer. Examples of these data are visualized at LCh color space vertical slices for magenta and blue hues on figure 4. The upper line of the left diagram shows the L reduction with continuous adding the magenta tone value to substrate from 0% to 100%, while the upper line of the right one — with the similar adding of M + C.

It may look that the further reducing of these colors lightness can be provided by adding to their solids the complementary process color. In the first case such color is green provided by the balanced sum of cyan and yellow, while for the blue tint it's yellow. However, the lower lines of both diagrams demonstrate much purer colors generation by simple adding of the black ink. The increase of saturation achieves 30 chroma units for magenta at L = 30 level and about 40 units for blue at L = 23.

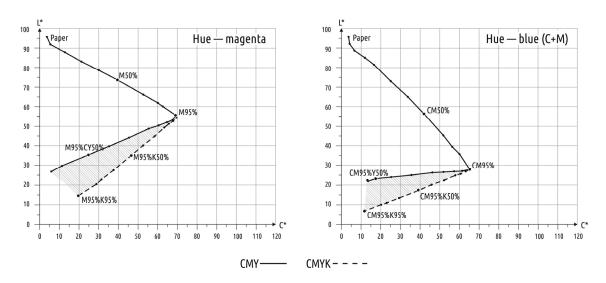


Figure 4. Coordinates of CMY and CMYK dark chromatic colors on meridian sections of CIE Lch space for magenta (left) and blue (right) hues.

Such difference can be physically explained by that the black ink more or less uniformly reduces reflectance without distortion of source (magenta or blue) spectrum profile, while the inks of opposite colors darken the source one by suppressing reflectance at their inherent in bands of spectrum. The hatched areas of the both diagrams demonstrate the expanding of lower part of color gamut with the use of the fourth, K color. According our three dimensional calculations it comprises about 10%.

5. Conclusions

It's purposeful to consider the black ink use in CMYK process in relation of its three basic functions:

- replacement of achromatic share of CMY originated chromatic colors;
- reproduction of achromatic colors in combination with balanced CMY;
 - creating the new chromatic colors unavailable for CMY.

Within the whole its variants continuum the first of these functions is in full characterized by two dimensions:

- volume of K as the share of a reference (CMY)_{min};
- location of certain K volume application within the tone value range of a print.

Contrary to the first one, the other two of these functions are completely additional and have no alternatives.

New chromatic colors provided by adding black ink expands the CMY gamut on about 10%. So, the move from CMY to CMYK which took place in the 30ies of last century can be concerned as the first step to Hi-Fi color technologies aimed nowadays on a similar effect by once again increasing the number of process inks.

The results of research and discussion can help to explore the effect of described black ink functions in multicolor halftones.

References

Avatkova N. et al. *Atlas of chromatic colors minimization in offset printing*. Experimental print shop of VNII poligrafii, Moscow, 1987 (Rus.).

Bala R. *Device characterization*. Ch. 5 in Digital Color Imaging Handbook, ed. by G. Sharma, CRC Press, 2003, p. 366, p. 358.

Daels K., Delabastita P. Tone Dependent Phase Modulation in Conventional Halftoning / Recent Progress in Digital Halftoning.- IS&T, 1994.- P. 46-49.

Enoksson E. Image reproduction practices / TAGA Proc. 2004, pp. 318-331.

Hunt R. Why is Black and White so Important in Color? Recent Progress in Color Science.- IS&T, 1997.- pp. 123-126.

Kipphan H. Handbook of Print Media, - Springer, 2001, pp. 519, 578-579.

Kuznetsov Y.V. Image data processing technology. "SPb Institute of Printing", Moscow-SPb, 2002 (Rus.).

Kuznetsov Y.V. Providing the balance of an "old" and "new" knowledge in education content: examples from graphic technology teaching. Advances in Education Research. USA, Vol. 30, pp. 59 - 65, 2013.

Southworth M., Southworth D. Complete Color Glossary // The Color Resource.-1992.

42nd International Research Conference, 06 to 09 September 2015



Session 3A

Printed functionality 2

Monday, 7 September 2015 16:05 – 17:45

Chair: Tim Claypole

Co-Innovating novel Printed-Electronic Products

Kaisa Vehmas, Thea Sipiläinen-Malm, Anu Seisto, Maria Smolander, Himadri Majumdar

VTT Technical Research Centre of Finland, P.O. Box 1000, FI-02044 VTT, Finland

E-mail: kaisa.vehmas@vtt.fi; thea.sipilainen-malm@vtt.fi; anu.seisto@vtt.fi; maria.smolander@vtt.fi; himadri.majumdar@vtt.fi

Short Abstract

In this study co-innovation and co-development with the consumers were utilized for the first time in a project presenting future printed electronics applications. Even if large scale manufacturing of disposable printed electronics products is not yet feasible, it is possible to evaluate consumer attitudes towards such products. Owela platform, developed at VTT in 2007, was utilized to involve consumers in the co-innovation and co-development process.

Examples of novel biodegradable printed electronics applications were presented to the participants in Owela. The participants were able to develop these ideas further and innovate new ones. They were not confused with detailed information or any possible limitations; they were still able to ideate freely. The participants were interested in the novel solutions presented. Especially with applications in themes of gardening and wearables positive feedback was received. In some cases people were concerned that the sensors would decrease the interplay between people or increase the price of the product or service. Novel innovations besides the given examples related mostly to health and wellness. People would be interested in utilizing novel sensor applications in treatment of different diseases and determining the condition of their own body.

Keywords: Printed electronics, biodegradable, co-innovation, Owela

1. Introduction and background

Extensive use of electronics in communications, data processing and entertainment, is leading to huge amount of electrical and electronic equipment waste and to a rapid exhaustion of natural elements. Printed electronics, as being produced by additive manufacturing processes, could dramatically reduce the environmental impact, especially when biobased materials are used. Printed Electronics is forseen as the most viable solution for large-area electronics applications. This includes use of large-area displays, solar-cells, smart buildings with integrated sensors and electronics and, also, wearables. To enable such applications, electronics components, including resistors, capacitors, diodes, transistors and electronic circuits are essential and they need to be printed on large-area substrates.

Printed electronics is produced by conventional printing techniques for patterning materials (semiconductors, conductors, insulators) and manufacturing electronic components onto a wide range of flexible and organic substrates, such as paper, plastic foils, and labels (Sun et al., 2010). For printing electronics on large-area substrates traditional printing processes, namely screen, flexography, offset, inkjet, gravure, and also novel, application-specific printing methods, such as super-inkjet, reverse-offset etc., are used. Each printing process has its advantages and disadvantages regarding printed electronics. General problem facing printed electronics currently are registration and alignment accuracy. In some cases, e.g. for printed transistors, accuracy in order of 5-10 microns are required for efficient device performance. Research and development are ongoing to eliminate these obstacles.

Sustainable printed electronics technology means free of toxins and rare earth elements and low energy consumption, both in the manufacturing process and in operation. Sustainability in electronics refers to biobased, biodegradable, biocompatible, bioresorbable, or even metabolisable electronics.

Printed electronics will offer many new possibilities towards more environmentally friendly applications and improving safety. The bulkiness of electronics is minimised and extremely light and flexible products may be produced. Conventional complex production of electronic equipment also results in much more waste compared to printing process of electronics. However, several scientific and engineering challenges still need to be addressed, e.g. new materials with better electrical performance that offer additional functionality (mechanical flexibility, and optical and magnetic properties) need to be developed. Energy-efficient processes with fewer steps to achieve accurate and less defective fabrication also need to be designed.

Even though at the moment there are no significant investments into printed electronics by large printing houses, different stakeholders like electronic, paper, energy and chemical companies have invested into research in this topic. There is still a gap between technology development and market-demand for the printed electronics technology. However, with the onset of Internet of Things the prospect of large-area (opto)electronics is great for distributed sensing, lighting, signage and power-generation applications.

However, even if large scale manufacturing of disposable printed electronics products is not yet feasible, it is possible to evaluate consumer attitudes towards such products. In this study, we involved ordinary consumers to express their interest in several future use cases, in which both disposability and affordable printed electronics applications play a role.

2. Potential in biodegradable printed electronics

Many materials with bio-origin have been identified as suitable substrates for the fabrication of organic electronics (Irimia-Vladu, M. 2014), (Irimia-Vladu, M., et al. 2012). Many of these materials demonstrate properties, processability and functionalities which present low-cost, non-toxicity, biodegradability, and often biocompatibility and even bioresorbability. One of the oldest and most familiar 'substrate' materials of natural origin is paper, whereas nanocellulose materials seem to be promising new materials for flexible electronics. Another natural material with a long history is silk. Silk is fully bioresorbable and can be safely implanted into the body. Silk has been used as a substrate for passive RFID circuits that can be integrated directly onto food as sensors of food quality (Tao, H., et al., 2012).

Polysaccharides can be used as biocompatible substrate materials. Polymers made of starches and polylactic acid have been commercially mass-produced as biodegradable plastics. Caramelized glucose was explored as an exotic substrate for electronics. Deoxyribonucleic acid, DNA, is extracted in large amounts from waste products of the fishing industry, and can be produced on an industrial scale. The individual nucleobases (guanine, adenine, thymine, and cytosine) are extracted commercially for medical and cosmetic applications and have been implemented as gate dielectrics for OFETs (Irimia-Vladu, M., et al., 2012).

Research in biodegradation and biocompatibility of organic semiconductors remains limited. Chlorophyll, beta-carotene, indigo and Tyrian purple are examples of cheap and nontoxic materials for bio-originating semiconductors. Conductors are the most underdeveloped branch in the field of biobased electronics. Melanin and chitosan are potential conductor materials. However, synthetic conducting polymers such as polyaniline, polypyrrole, and polythiophenes have demonstrated excellent biocompatibility in biological applications (Angione, M. D, et al, 2011), (Serrano, M. C. et al., 2010).

Recent demonstrations of high performance organic electronics based on biomaterials have shown that biodegradability and sustainability in electronics have potential and, hopefully, are poised to make a positive impact in the future.

3. New technology and technology acceptance

To be successful, innovative solutions must take into account opportunities provided by new technology and materials but they cannot lose sight of the users. Companies have understood how important it is to understand the needs and expectations of the end users of the product or service. Users are experts on user experience and thus as a significant source of innovation (Tomke and von Hippel, 2002). It is easier for the users to develop products that are already familiar to them or comment on incremental improvements for existing products or services than completely new ones but when the ideation is supported in a proper way, users are able to create radical innovations (Heiskanen et al., 2007).

Technology acceptance models aim at studying how individual perceptions affect the intentions to use (information) technology and further the actual usage as presented in Figure 1. According to the Technology Acceptance Model for Mobile Services (Kaasinen, 2005), user acceptance of mobile services is built on three factors: *perceived value of the service, perceived ease of use and trust*. These three factors affect the intention to use a mobile service.

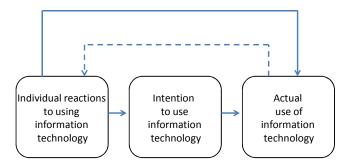


Figure 1: The basic concept underlying user acceptance models (Venkatesh et al., 2003).

Seisto et al. (2012) have studied the user experience in utilization of Augmented Reality (AR) technology in an advertisement in a printed magazine. According to their results, the user experience plays a significant role in *value perception of the service*.

4. Methods

Open Web Lab, Owela, was used in this project to involve consumers in the co-innovation and codevelopment process. Owela is designed for user centric studies and can be used as a collaboration and communication platform between users, developers and researches. By using Owela it is possible to get fast and direct feedback from users and create an interactive relationship with them. By using online tools it is possible to reach a large amount of users quickly and cost-efficiently. Online tools enable more easy and effective participation due to the fact that the users can participate from the place they want and at the time suitable for them (Näkki and Antikainen, 2008).

There are different types of tools in Owela that can be widely used in the development process. Tools are selected separately for each study depending on its purpose. Owela methods can be combined with other user centric methods, like face-to-face interviews and real world Living Lab studies. (Friedrich, 2013)

A separate workspace was created for this project in Owela. Participants for the discussion were recruited from Owela panel, existing Owela users, and a link to the projects' Owela workspace was

also shared in Facebook and Twitter. Owela suits well for co-development in all different phases of an innovation process. In this project Owela was used in very early stage of the ideation process; people were asked to co-innovate and co-develop products that do not exist yet.

The goal of this Owela discussion was to clarify, how people feel about sensoring in their everyday life. Different possibilities for sustainable printed electronics solutions were presented to the participants in the themes of gardening, wearables, toiletries, domestic animals and packages. People were asked to comment and develop these ideas further and they were also able to ideate totally new printed functionality solutions.

In this project co-innovation and co-development with the consumers was utilized for the first time in printed electronics project. The challenge was that the consumers are necessarily not familiar with the terms of printed electronics or biodegradability. The project group ideated five novel solutions where printed electronics was utilized in every-day contexts and where sustainability issues were taken into consideration in a way that people could benefit from biodegradable possibilities. These solutions were presented to the participants with simple examples including pictures and text to help their innovation but not to direct them too much for certain solutions. Three of these solutions, gardening, wearables and toiletries, are presented in this paper, see Figures 2-4. The discussion platform in Owela was open during two weeks. Together 48 Finnish consumers participated the discussion and totally 275 comments for the discussion themes were achieved. 20 of the participants were men (42 %) and 28 women (58 %). The age distribution was 18-89 and the average age of the participants was 51 years.



Figure 2: Case gardening: watering sensor for your private garden



Figure 3: Case wearables: Sensoring and screening of the speed of the runner



Figure 4: Case toiletries: Sensoring the moisture in the nappies of the babies

5. Results

According to the results people were well disposed towards novel printed electronic solutions and to use them in their everyday life. They were willing to openly innovate novel products and services. Still, the participants were a bit concerned if the additional functionality in the products increase the price, decrease the usability or make people more incapable to handle their everyday life.

5.1 Case gardening

People liked the idea of watering sensor and were willing to use this kind of solution in their own garden, especially if it could be possible to have a plant or species specific watering systems. This moisture sensoring could be combined with automatic watering system in the garden and it could be connected to a monitor with light signals to easily see the condition of the plant. participants ideated also that an alarm system could be included in the application and it should be connected with the weather forecast.

Participants pointed out that it would be interesting to sensor some other properties from the garden like nutrient content and air humidity. Indoors it would be useful to have sensors also for lightness and temperature. On public places sensoring of swaying of bigger trees would be useful to avoid bigger

accidents. Sensors could also be utilized in nursing the grass; what is the optimal time for cutting and the stage of the grass and weeds.

In addition to participants' own garden they listed other possible targets for this application. These kinds of sensors could be used also in indoors with potted plants, in professional glasshouse cultivation, gardens at the stores, silviculture and agriculture and also in transportation of the plants and landscaping in public places.

5.2 Case wearables

Sensoring and screening of the speed of the runner in his/her number tag got mainly positive comments. Participants thought that this application could be used on the side of a pulse counter and it could also work as a personal trainer, for example, to ensure that the training movements have been done correctly. Participants ideated that a similar application could be used in determining the location of children, elderly people, people with severe illnesses, domestic animals, hikers, people in accidents etc. The target groups for this kind of application could be found from day care centers, sport clubs, hospitals / nursing homes, event organizers or from police and fire service.

In the case of wearables participants innovated clothes that can be used to follow the vital function of the patients (epilepsy, diabetes, heart defect...). They would also be willing to have luminous tags for outdoor clothes with twilight switch and leds.

5.3 Case Toiletries

People did not like the idea of sensoring the moisture in the nappies of small children. According to the participants this would decrease the use of common sense and communication between parents and their children. Still, the participants saw potential in this moisture sensoring application in hospitals and in nursing aged people. In the case of elderly people it would be useful to sensor other values from urine also, such as sugar or protein.

5.4 Other applications

Participants were asked to innovate some totally new ideas different from presented examples. Most of the ideas related to health and wellness services; people were interested in applications that would help taking care of e.g. sleep apnea, epilepsy and diabetes, but also to control heart defects. In addition to following their health, it would be useful to have sensors to determine different human body related issues like weight, fluid balance and percentage of fat. Handicapped people could benefit e.g. from distance sensors in their everyday life.

On the basis of these results we believe that the same factors, described in Technology Acceptance Model for Mobile Services (Kaasinen, 2005) can be applied also in this case. Consumers were able to innovate and they would be willing to use these kinds of novel applications if they see that they will perceive value of the service, the service is easy to use and they are able to trust the service.

6. Discussion

In today's service innovation studies, user centricity and user experience are central perspectives. Researchers have pointed out that user involvement often speeds up the development process: instead of separate market research exercises, ideas are received directly from the participating users (Alam and Perry, 2002).

In this study we presented consumers different future use cases with a common focus on biodegradable printed electronics applications. Participants were able to actively participate in the discussion of different use cases, even though none of the cases presented are commercially available at the moment. The use cases were presented with pictures and text, which proved to be enough to motivate the participants to comment on the given examples and to start their own ideation process. In all cases, comments were received on how to improve the product or service further and who would be the most potential users if not the ones presented in the original use case. In the case of gardening, the service concept was widened considerably in the co-development process. Instead of helping the gardener to know when to water the plants, it was suggested that the service would be connected to automatic watering, include an alarm system and utilize weather forecast information, making it a fairly complicated IoT service.

From the viewpoint of technology acceptance, ease of use and trust were not raised up in the discussions. This is most probably due to the early innovation stage, and it is possible that these topics will be of concern in the following phases with prototypes of the services. However, based on earlier studies with AR technology (Seisto et al., 2012), also the brand of the service provider plays an important role in how trust in the service is perceived.

The participants had no problem in trusting that the use cases presented would be possible to implement in practice and they had no questions on what kind of materials would be used on the products. This can also be expected to be due to the early stage of the innovation process. It is possible that the ways in which sustainability and biodegradability of the products is communicated to the consumers have on effect on the trust and final acceptability of the proposed service.

Perceived value of the product and service was clearly the most important viewpoint in the discussions. The participants were quite strict and honest about their feelings towards how suitable the proposed use cases were. The gardening use case was perceived as very valuable, and many additional ideas were written to support the service. The use case was clearly one where the participants could see a lot of potential and practical benefits. Also examples dealing with health and wellness topics were found valuable, possibly due to the mega trend of "guantified self" in which different applications are used to monitor e.g. steps taken during the day. It was also interesting to note that in the use case was not correct. They didn't see that a sensor application would bring the most value to the parents of small children, but a better focus group could be elderly people.

7. Conclusions

The most important evaluation criterion at this stage of the innovation process of future biodegradable printed electronics applications by the participants was found to be the perceived value of the product and service. The materials and the sustainability aspects did not emerge into the discussion in this stage.

User acceptance is crucial to the success of new technologies. Therefore, it is important to involve consumers from the very beginning of the innovation process. Then, it is possible to clarify their attitude towards the new technology including their needs and expectations. These results can then be combined with the service providers perspective; how do they see their future, what are the challenges and possibilities and what is the business potential of their application. In close co-operation with consumers, companies and research partners it is possible to clarify the future steps and ensure that the novel solutions are relevant and accepted. During the development process it is reasonable to continue the discussion with the consumers when something new, like a new model or prototype of the application is available.

Combining new technologies and materials with user needs, and adopting them in new unexpected ways give rise to products that present new commodity categories. The challenge is to drive forward innovations from laboratory stage to production on an industrial scale.

References

Alam, I. and Perry, C. (2002). A customer-orientd new service development process. Journal of Services Marketing 16(6), 515-34.

Angione, M. D, Pilolli, R., Cotrone, S., Magliulo, M., Mallardi, A., Palazzo, G., Sabbatini, L., Fine, D., Dodabalapur, A., Cioffi, N., Torsi, L. (2011). *Carbon based materials for electronic bio-sensing* Mater Today (2011) 14(9), 424-433.

Friedrich, P. (2013). *Web-based co-design: Social media tools to enhance user-centred design and innovation processes*. Espoo 2013. VTT Science 34. 185 + 108 p.

Heiskanen, E., Hyvönen, K., Repo, P. and Saastamoinen, M. (2007). *Käyttäjät tuotekehittäjinä*. Teknologiakatsaus 216/2007. Tekes, Helsinki. [Users as product developers]

Irimia-Vladu, M. (2014). "Green" electronics: biodegradable and biocompatible materials and devices for sustainable future. Chem. Soc. Rev., 2014, 43, 588-610.

Irimia-Vladu, M., Glowacki, E.D., Voss, G., Bauer, S. and Sariciftci, N.D. (2012). *Green and biodegradable electronics*. Materials Today, 2012, 15 (7-8), 340-346.

Näkki, P. and Antikainen, M. (2008). Online tools for Co-design, User Involvement through the Innovation Process. Tapir Akademisk Forlag, pp. 92-97.

Seisto, A., Aikala, M., Vatrapu, R., Kuula, T. (2012). Augmented Reality as a technology bringing interactivity to print products. IARIGAI Conference, 2012.

Sun, J., Zhang, B., Katz, H.E. (2010). Materials for printable, transparent, and low-voltage transistors. Adv Funct Mater 21, pp. 29–45.

Olsson, T., Lagerstam, E., Kärkkäinen T. & Väänänen-Vainio-Mattila, K. (2011) *Expected user experience of mobile augmented reality services: a user study in the context of shopping centres*. Pers Ubiquit Comput. 20 Dec. 2011. DOI: 10.1007/s00779-011-0494-x.

Serrano, M. C. Chung, E.J. and Ameer, G.A. (2010). Advances and Applications of Biodegradable Elastomers in Regenerative Medicine Adv Funct Mater, 2010, 20, 192–208.

Tao, H., Brenckle, M.A., Yang, M., Zhang, J., Liu, M., Siebert, S.M., Averitt, R.D., Mannoor, M.S., McAlpine, M.C., Rogers, J.A., Kaplan, D.L. and Omenetto, F.G. (2012). *Silk-Based Conformal, Adhesive, Edible Food Sensors*. Adv Mater, 2012, 24, 1067-1072.

Thomke, S. and von Hippel, E. (2002). *Customers as innovators: a new way to create value*. Harvard Business Review, 80(4), pp. 74-81.

Venkatesh, V., Morris, M. G., Davis, G. B. and Davis, F. D. (2003) User acceptance of information technology: Toward a unified view. MIS Quarterly, Vol. 27. No. 3, September, pp. 425.478.

Fully printed Biodegradable Nanocellulose-Based Humidity Sensor for SMART LABEL Applications

Tomáš Syrový¹, Vinay Kumar², Martti Toivakka², Chunlin Xu³, Lucie Syrová¹, Jan Řeboun⁴, Douglas W. Bousfield⁵

¹University of Pardubice, Faculty of Chemical Technology, Department of Graphic Arts and Photophysics, 533 53 Pardubice, Czech Republic

²Åbo Akademi University, Laboratory of Paper Coating and Converting and Center for Functional Materials, Turku/Åbo, FI-20500 Finland

³Åbo Akademi University, Johan Gadolin Process Chemistry Centre, c/o Laboratory of Wood and Paper Chemistry, Turku/Åbo, FI-20500 Finland

⁴University of West Bohemia, Faculty of Electrical Engineering/RICE, Department of Technologies and Measurement, 306 14 Plzeň, Czech Republic

⁵University of Maine, Department of Chemical and Biological Engineering, Orono, ME, USA

E-mail: tomas.syrovy@upce.cz; vinay.kumar@abo.fi; martti.toivakka@abo.fi; cxu@abo.fi, lucie.syrova@upce.cz; jreboun@ket.zcu.cz;bousfld@maine.edu

Short abstract

Resistive-type humidity sensors were fabricated by using nano- (NFC) or microfibrillar cellulose (MFC) as a sensing layer and printing substrate at the same time. The variation of impedance within a range 20-90 % of relative humidity was measured across a printed carbon based interdigitated structure. The NFC-based sensors exhibit logarithmic sensitivity to moisture up to 75 % RH, beyond which saturation is observed. The MFC-based sensors have proper sensitive behaviour in the range 40-90 % RH. Based on the results, it is obvious that sensor behaviour is controlled by the differences surface chemistry and nanoscale fibrillar structure of the cellulosic materials, which lead to differences in interaction with moisture. The response of sensors in the whole range of RH can potentially be optimized by proper combination of both types of cellulose materials.

Keywords: printed sensor, humidity sensor, nanocellulose, microcellulose, NFC, MFC

1. Introduction and background

Numerous different types of humidity sensors have been proposed in literature (Chen and Lu, 2005; Reddy *et al.*, 2011; Mahadeva *et al.*, 2011; Yang *et al.*, 2006). Typically, the sensing principle is based on a functional layer of either inorganic – Al_2O_3 , SnO_2 (Yawalw *et al.*, 2007); TiO₂ (Su and Huang, 2007), ZnO₂ (Chang *et al.*, 2010) or organic – small molecules (Rakusan *et al.*, 2011; Syrový *et al.*, 2013) or polymers – PVA (Yang and Chen, 1998), PSSNa, PDEB (Li and Yang, 2002), PPBT, PMMA (Ralston *et al.*, 1994), PET (Pérez and Freyre, 1997). However, many of these materials are problematic or not compatible with printed electronics-based manufacturing. Typical challenges are difficulties in formulating the functional materials as inks (Syrový *et al.*, 2013) and coating fluids in sufficient amounts, the need to use high annealing temperatures, and lack of sufficient long time stability of the fabricated sensors.

To enable their wide use in different applications, printed humidity sensors should strive for simplicity in the detection of humidity. While measurement of relative humidity (RH) from a DC resistance signal is straight-forward, the sensors suffer from poor stability, caused by migration or degradation of the involved materials. The objective of present work was to demonstrate printed humidity sensors which utilize nano- (NFC) or microfibrillar cellulose (MFC) as sensing layers. The two types of cellulosic materials exhibit differences in their hygroscopic behaviour, which can be a benefit in sensors of this type. NFC and MFC are also suitable as printing surfaces, since they can be processed into strong, transparent films. Depending on the application, the natural biodegradability of cellulosic materials can also be an advantage. Using the NFC/MFC as both a sensing layer and a printing substrate simplifies the sensor fabrication by eliminating one printing/coating step. An assumption of the current study is that the cellulosic base sensing layer is sufficiently stable during AC measurement of impedance, which depends on RH and temperature.

2. Materials and methods

In presented work, MFC and NFC films were used as substrates for fabricating humidity sensors. These films were prepared by casting and evaporation under controlled air conditions of 23°C and 50% RH. The fiber suspension was stirred at 700 rpm for 30 min using a magnetic stirrer (RCT Basic from IKA) before casting into polystyrene petri dishes (dia. 8.5 cm). Glycerol (SigmaUltra, 99% (GC) with $M_w = 92.09$) was used as plasticizer (15% by weight) in NFC films as they were otherwise very brittle and difficult to handle.

The two types of nanocellulose, i.e. MFC and NFC were prepared using different methods. MFC was produced using mechanical treatment (refining) at the Process Development Center of the University of Maine. A bleached softwood Kraft pulp was dispersed with a beater at 2.5% solids and sent to a refiner equipped with specialized plates and operated with low clearance and careful gap control. The pulp was circulated through the refiner until the fines content reached over 90% as measured with a standard fiber size analyzer (Morfi, Techpap) (Kumar et al., 2014). NFC was prepared using a chemical pre-treatment (TEMPO-mediated oxidation) followed by mechanical disintegration of cellulose fibers as described by Liu (Liu et al., 2014). Two grams of cellulose fibers were dispersed into 100 mL distilled water and stirred for 4 h at room temperature. The dispersed fiber suspension was subsequently mixed into a 100 mL solution containing 32 mg TEMPO (0.1 mmol/g fiber) and 200 mg sodium bromide (1.0 mmol/g fiber). The pH of the slurry was adjusted to 10.0 by adding 0.5 M NaOH. To this slurry, a desired amount (10 mmol/g fiber) of 10% NaClO was added dropwise to start the oxidation. The total volume of the NaClO was added within one third of the designated reaction time while maintaining the pH at 10.5 by adding 0.5 M NaOH. No further NaOH consumption was assumed to indicate the equilibrium stage of the reaction. The oxidized cellulose was precipitated in ethanol with a ratio of 1:3 (v/v) and thoroughly washed with distilled water by filtration, and the residual ethanol was removed by rotary evaporation. A domestic blender (OBH Nordica 6658, Denmark) was applied for 2 min at an output of 300 W to fibrillate the oxidized cellulose at a consistency of 0.5%. The obtained NFC was stored in a cold room before further analysis. The obtained MFC and NFC suspensions are shown in Figure 1. It can be observed from TEM micrographs of these suspensions in Figure 2 that NFC fibers are much smaller in diameter and length compared to MFC fibers.

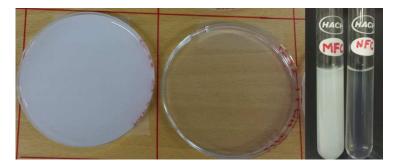


Figure 1. MFC (left) and NFC (right) suspensions.

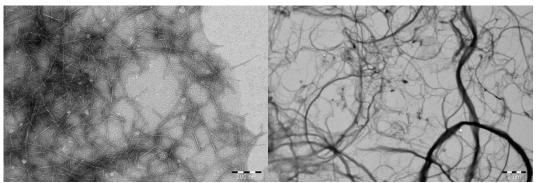


Figure 2. TEM microphotograph of NFC (left) and MFC (right) suspensions

The NFC/MFC-based sensors were expected to behave similarly to sorption sensors, for which the sensor resistance or impedance is influenced by humidity and temperature. To measure this, carbon-based interdigitated (ID) structures were printed on the NFC/MFC substrates using carbon/graphite GWENT D1 ink formulation. The width of electrode "fingers" and the gaps between them were 200 μ m (Figure 3). The sensors were printed on semiautomatic screen-printing machine S 200 HF equipped with the aluminium flat bar and polyurethane sandwich squeegee with hardness of 75/90/75 Shore A. The mesh count was 100 threads per centimetre. Alongside the cellulosic films, a typical non-absorbing substrate PET (Melinex ST-504) was used to provide information about RH sensing behaviour of the printed ID structures.

Measurements of NFC and MFC sensorial properties were done in a climatic chamber Votch VC7018 with controlled temperature and RH. Humidity dependence was controlled fully automatically in the range from 20 % to 90 % RH for temperatures from 20 °C to 50 °C. The RH was increased continuously from 20 % to 90 % at 0.5 % RH/min speed. The whole measurement cycle is shown in Figure 4. Prior to the measurement cycle, all sensors were exposed to high humidity and temperature environment for the stabilization of the sensors. The total duration of a cycle was 24 hours. The measurement of all sensors was repeated three times. Electrical parameters of sensors were measured by AC RLC bridge Agilent E4980A equipped with 10 channel multiplexer.

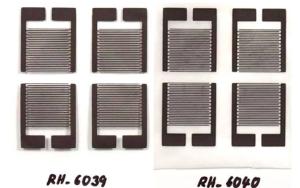


Figure 3. NFC- (RH_6039) and MFC- (RH_6040) based RH sensors.

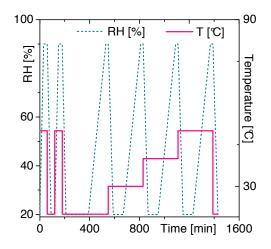


Figure 4. Humidity and temperature cycle in climatic chamber

3. Results and discussion

3.1 Physical characteristics of NFC/MFC films

The physical properties of MFC and NFC films are presented in Table 1. NFC films had a higher tensile strength and elastic modulus, but they were more brittle compared to MFC films as is indicated by values of elongation at break.

Sample	Tensile strength	Elastic	Elongation at	Thickness
	(MPa)	modulus (GPa)	break	(µm)
	(IVII <i>a)</i>	(01 a)	(%)	(µ111)
MFC	81 ± 3	5.5 ± 0.3	3.86 ± 0.27	25 ± 1.6
NFC	109.5 ± 10.2	7.2 ± 0.3	2.7 ± 0.7	23 ± 1.4

Table 1. Mechanical properties of NFC/MFC substrate/sensing layers.

3.2 Sensing characteristics of NFC/MFC based RH sensors

Initially, during the first RH cycles, the impedance characteristics at 1 kHz frequency were measured.

For evaluation of sensitivity of carbon based ID structure itself to the moisture, non-porous PET substrate was used. The ID structures printed on PET using carbon based ink formulation exhibit sensing properties dependant on humidity from 70% RH and higher (Figure 5). The sensing behaviour of ID structure due to humidity is well known phenomenon, and it's given by the fact that the high relative humidity increases the capacity between the electrodes, and decreases the resistance between ID electrodes at the same time. Both these factors cause a decrease in impedance approvingly. The impedance is also dependent on the distance between fingers of ID structure.

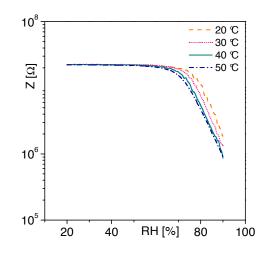


Figure 5. Dependence of impedance on RH and temperature for carbon based ID on PET.

Cellulose based sensor structures using same interdigitated carbon electrodes printed on NFC and MFC substrates show high sensitivity to RH.

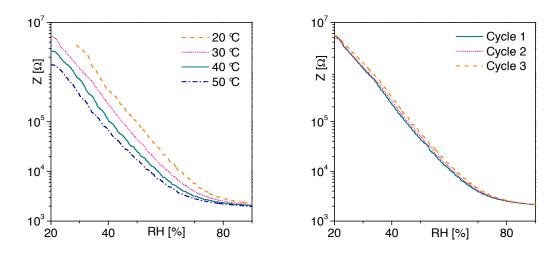


Figure 6. Left: Dependence of impedance on RH and temperature for NFC-based sensors. Right: stability of sensor response in repeated cycles.

The impedance-humidity characteristics of the sensor structure on the NFC substrate (Figure 6) exhibits high impedance changes at low humidity levels. This behaviour is uncommon in chemiresistive sensors and is very promising for the construction of fully printed RH sensors. The sensor structure is sensitive to humidity levels up to about 75 % RH. The saturation of impedance was observed at higher RH values. The maximal impedance is below 10 M Ω and three orders of magnitude change in layer impedance can be observed when RH changes from 20 % to 75 %.

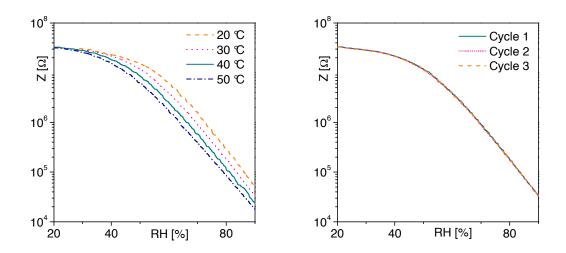


Figure 7. Left: Dependence of impedance on RH and temperature for MFC-based sensors. Right: stability of sensor response in repeated cycles.

The impedance-humidity characteristics of the sensor structure on the MFC substrate (Figure 7) shows, in contrast to the NFC sensors, sensitivity at high humidity levels. The sensor shows a change in impedance starting from the humidity level of about 40% RH without any saturation at high humidity values. When RH increases from 40 % to 90 %, three orders of magnitude change in layer impedance can be observed. The impedance-humidity characteristic is almost exponential with the maximal impedance below 20 MΩ. It is obvious from Figure 5 and Figure 7 that sensors based on PET and MFC substrates are more sensitive at higher levels of RH, but MFC base sensor exhibits much higher sensitivity in range 40-75% RH. This could be caused by larger surface area and more hydrophilic nature of NFC films.

Some mechanical deformation of the NFC and MFC substrates was observed after the humidity cycles, which can influence the obtained results. It is obvious from the stability plots (Figure 6, right & Figure 7, right), that the MFC sensors exhibit very high reproducibility of sensor characteristics, whereas the stability of the NFC-based sensors was inferior to that of the MFC. This can be contributed to the higher deformation (swelling) of the NFC substrate at high RH. To improve the mechanical and sensing stability of the NFC sensors at high humidity, the NFC can be fixed/laminated on to a carrier substrate, e.g. paper, or various modifications can be done to the NFC material itself.

4. Conclusions

It is obvious from the obtained results that the MFC and NFC substrates represent very promising sensing layers for humidity sensing applications. The nature of their sensing behaviour is dissimilar, and probably related to differences in their nano- and microfibrillar structure and surface chemistry, which leads to differences in hygroscopic properties. It is clear that each type of sensing material is limited to sensing at a specific range of RH. Based on the current findings, future work will investigate use of combinations of NFC and MFC to widen the measurable RH range, and coating of optimized NFC/MFC mixture to a carrier substrate such as paper.

Acknowledgments

The research has been performed thanks to created relationships and support within COST Action FP1104. This research was supported by the research project FLEXPRINT No. TE01010022 under the auspices of the Czech Competence Centres Programme.

References

Chang S., Chang S-J., Lu Ch., Li M., Hsu Ch., Chiou Y., Hsueh T., Chen I., A ZnO nanowire-based humidity sensor, Superlattices and Microstructures, Volume 47, Issue 6, June 2010, Pages 772-778, ISSN 0749-6036, http://dx.doi.org/10.1016/j.spmi.2010.03.006.

Chen Z., Lu Ch., Humidity Sensors: A Review of Materials and Mechanisms, SENSOR LETTERS, Vol. 3, 274–295, 2005.

Kumar V., Bollstrom R., Yang A., Chen Q., Chen G., Salminen P., Bousfield D. and Toivakka M. (2014) Comparison of Nano- and Microfibrillated Cellulose Films. Cellulose, 21:3443–3456

Li Y., Yang M., Bilayer thin film humidity sensors based on sodium polystyrenesulfonate and substituted polyacetylenes, Sensors and Actuators B: Chemical, Volume 87, Issue 1, 15 November 2002, Pages 184-189, ISSN 0925-4005, http://dx.doi.org/10.1016/S0925-4005(02)00233-2.

Liu J., Korpinen R., Mikkonen K. S., Willför S. and Xu C. (2014) Nanofibrillated cellulose originated from birch sawdust after sequential extractions: a promising polymeric material from waste to films. Cellulose, 21:2587–2598

Mahadeva S.K., Yun S. and Kim J. (2011) Flexible humidity and temperature sensor based on cellulose-polypyrrole nanocomposite. Sensors and Actuators A: Physical, 166:194–199.

Pérez J. M. M., Freyre C., A poly(ethyleneterephthalate)-based humidity sensor, Sensors and Actuators B: Chemical, Volume 42, Issue 1, 15 July 1997, Pages 27-30, ISSN 0925-4005, http://dx.doi.org/10.1016/S0925-4005(97)80308-5.

Rakusan J., Karaskova M., Hamacek A., Reboun J., Kubac L., Cerny J., Rais D., And Nespurek S.: Double layer humidity sensor based on phthalocyanine derivatives. Proceedings of Internetional Conference Nanocon 2011, held in Brno, Czech Republic September $20^{th} - 23^{rd}$ 2011, pages 1241 – 1247.

Ralston A. R.K., Tobin J. A., Bajikar S. S., Denton D. D., Comparative performance of linear, cross-linked, and plasma-deposited PMMA capacitive humidity sensors, Sensors and Actuators B: Chemical, Volume 22, Issue 2, November 1994, Pages 139-147, ISSN 0925-4005, http://dx.doi.org/10.1016/0925-4005(94)87013-6.

Reddy A. S. G., Narakathu B. B., Atashbar M. Z., Rebros M., Hrehorova E., Bazuin B. J., Joyce M. K., Fleming P. D. and Pekarovicova A. (2011) Printed Capacitive Based Humidity Sensors on Flexible Substrates. SENSOR LETTERS, 9:1–3.

Su P., Huang L., Humidity sensors based on TiO2 nanoparticles/polypyrrole composite thin films, Sensors and Actuators B: Chemical, Volume 123, Issue 1, 10 April 2007, Pages 501-507, ISSN 0925-4005, http://dx.doi.org/10.1016/j.snb.2006.09.052.

Syrový T., Syrová L., Kaplanová. M., Černý J., Kubáč I., Řeboun J., Hamáček A., Printed humidity sensor for measuring in wide range of temperatures and relative humidities, Utility model PUV 2013-27805, 2013.

Yang B., Aksak B., Lin Q. and Sitti M. (2006) Compliant and Low-cost Humidity Sensors using Nano-porous Polymer Membranes. Sensors and Actuators B: Chemical, 114(1):254-262.

Yang M., Chen K., Humidity sensors using polyvinyl alcohol mixed with electrolytes, Sensors and Actuators B: Chemical, Volume 49, Issue 3, 1 July 1998, Pages 240-247, ISSN 0925-4005, http://dx.doi.org/10.1016/S0925-4005(98)00134-8.

Yawale S. P., Yawale S. S., Lamdhade G. T., Tin oxide and zinc oxide based doped humidity sensors, Sensors and Actuators A: Physical, Volume 135, Issue 2, 15 April 2007, Pages 388-393, ISSN 0924-4247, http://dx.doi.org/10.1016/j.sna.2006.08.001.

Printed Batteries, Overview, Status, Recent Developments, Future Perspectives

*Gunter Hübner*¹, *Martin Krebs*², *Patrick Rassek*¹, *Andreas Willfahrt*¹

¹Hochschule der Medien, Institut f
ür Innovative Anwendungen der Drucktechnologien (IAD), Nobelstr. 10, 70569 Stuttgart, Germany www.hdm-stuttgart.de ²VARTA Microbattery GmbH, Daimlerstraße 1, 73479 Ellwangen, Germany. www.varta-microbattery.com

Email: huebner@hdm-stuttgart.de, Martin.Krebs@varta-microbattery.com, rassek@hdm-stuttgart.de, willfahrt@hdm-stuttgart.de

Short Abstract

Autarkic smart objects need power or energy sources. For flexible, thin and geometrically special devices the printed batteries are the best choice due to their design flexibility. Printed batteries are on the market for about 20 years but today mainly used for medical or cosmetic patches. The paper gives an overview about printed batteries, explains the main principles and properties, discusses the current status, recent developments and some future aspects. The batteries typically are screen printed on plastic foils with the need of good barrier properties in order to keep the electrolyte humid during lifetime of the battery. It shows some characterising methods and reports from reel-to-reel manufacturing of primary cells

Keywords: Printed batteries, thin film batteries, Printed Electronics, functional printing

1. Introduction and background

Presently the Organic and Printed Electronics Association OE-A divides the targeted market into 5 groups, see [OE-A 2013]

- Organic LED (OLED) Lighting
- Printable, Organic Photovoltaics (OPV)
- Flexible Displays
- Electronics and Components (printed memory and batteries, active components and passive components)
- Integrated Smart Systems (including smart objects including NFC/RFID, sensors and smart textiles)

Lighting, Displays, active components and especially the integrated smart systems have in common that they need a power source. If they should work autarkic, then they need a battery for power supply. If these devices are thin (around or less than 1mm), flexible or designed in a special geometric way, then the batteries should adapt to these properties. In this case classical rigid batteries (e.g. AA-cells or button cells) are not suitable and therefore the flexible design of printed batteries are extremely advantageous.

In the mid-1990s a new type of cells, the so-called pouch cells, have been introduced. These cells are lithium polymer batteries (i.e. Lithium-Ion with polymeric electrolyte) and are wrapped in a barrier foil. If a failure occurs the cells inflates due to internal gassing. The pouch cells paved the way to printed, flexible batteries.

Thus, since about 20 years printed batteries appeared on the market and nowadays several research groups work in that field.

The easiest printable electrochemical system is the $Zn-MnO_2$ one. It is related to the so called Alkaline-batteries that are very well known to the public. Since 1997 an Israeli company, called "Power Paper", offers such type of printed batteries for medical and beauty applications (cosmetic patches), see Figure 1. Today they seem to be China based since the website moved to China.

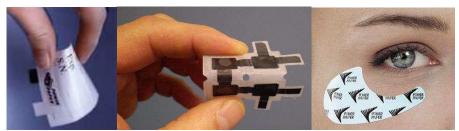


Figure 1 Thin flexible batteries by Power Paper (left: source <u>www.powerpaper.com</u> around 2007, middle and right: source: <u>www.powerpaper.cn 2014</u>)

Other enterprises a little later started to offer printed flexible batteries, too. For instance, the German based company KSW-Microtec as shown in Figure 2. This company no longer exists.



Figure 2 Thin flexible batteries by KSW-Microtec (source: <u>www.ksw-microtec.de</u> around 2007.The website no longer exists)

Another vendor offering flexible batteries (see Figure 3) was Thin Battery Technologies, Inc. (TBT) launched in 2003 based on knowledge of the company Eveready (now Energizer). Thin Battery Technologies now merged into the company Blue Spark Technologies located in Westlake Ohio, USA.



Figure 3 Thin flexible batteries Thin Battery Technologies, Inc. (TBT) (source: www.thinbattery.com around 2007. The website now belongs to Blue Spark Technologies)

Also around 2007 the company Enfucell based in Vantaa, Finland entered the market. Nowadays they offer three types of so-called "SoftBattery" a larger single cell (Figure 4), a smaller one and a series connection of two small cells.



Figure 4 Thin flexible batteries SoftBattery by Enfucell (source: <u>www.enfucell.com</u> 2014)

In July 2009, the Fraunhofer Research Institution for Electronic Nanosystems (ENAS) in Chemnitz, Germany announced in a press release a screen-printed battery. This printed battery is also based on the zinc-manganese system and combines two cells in series connection (Figure 5).



Figure 5 Screen-printed battery. Source: Fraunhofer-Gesellschaft (http://www.fraunhofer.de/presse/presseinformationen/2009/juli/batterien-drucker.jsp)

On the web some more offerings on Zinc/Manganese Dioxide printable batteries can be found. There are the Austrian company Prelonic <u>www.prelonic.com</u> (Plastic Electronic) or Thin Profile Technologies in Champlin, Minnesota USA <u>http://www.thinprofiletech.com/</u> or even more.

Other types of electrochemical systems for printed batteries have only been shown in lab stage, so far, no commercial application is known to the authors.

A project funded by the EU called GREENBAT led by VARTA Microbattery (Ellwangen, Germany) started in 2008 and aimed at environmentally friendly "green" batteries based on Lithium Ion. It was finished 2011.

The University of Applied Science, Hochschule der Medien Stuttgart, Germany started around 2007 together with VARTA Microbattery to investigate Ni/MH rechargeable batteries. Since then, several prototypes (e.g. shown in Figure 6) have been developed, presentations given at conferences and results published [Wendler 2014], [Wendler 2011], [Wendler 2010]. The work is ongoing in a project called BatMat.



Figure 6 Printed NiMH-Battery, (source www.hdm-stuttgart.de/iad).

In the USA, some Berkeley researches founded an enterprise called Imprint Energy, Inc. <u>http://www.imprintenergy.com/</u> that work on Zinc-Polymeric electrolyte rechargeable printable batteries.

Newer publications [Sun 2013] report of tiny batteries (Li-ion) directly coming from 3-D printers. The US based company Solicore (<u>www.solicore.com</u>) just recently announced an inkjet-printed Lithium-polymer primary cell (3V).

Today the market for printed batteries concentrates on beauty patches using $Zn-MnO_2$ non rechargeable systems. There are offerings called Iontopatch that contain printed batteries from Blue Spark Technologies (<u>http://www.bluesparktechnologies.com/</u>) see Figure 7 or from the Finnish company VTT (see Figure 8).



Figure 7 Iontopatch medical patches source: http://www.iontopatch.com/



Figure 8 Beauty patch from VTT (source: http://www.oe-a.org/article/-/articleview/136419)

2. Materials and Methods

Batteries can either be rechargeable (multiple use; in some countries called accumulator) or non-rechargeable (single use and dispose). Basically, a battery consists of two electrodes (comprising of electrochemical active materials) that are held apart by a separator to avoid shorts. The separator is soaked with electrolyte allowing the ion transport between the electrodes.

The positive terminal of the battery is called anode, and the negative end is called the cathode. The complete assembly is embedded in a gasketed housing.

The basic combination of anode, cathode and electrolyte is called single battery cell. Each cell offers a characteristic nominal voltage, e.g. a lead-acid cell has 2.1 V. If higher voltages are required, cells can be connected in series, thus, for example in a car six lead-acid cells are combined to a 12 V starter battery.

Detailed information about classical battery technology is given in textbooks [Linden 1995]. There are very many electrochemical systems that can form batteries but only a few are suitable for being printed, see Table 1 [Huebner 2015].

electrochemical systems	voltage	electrochemical reaction	Electrolyte	applicability for printing	
non rechargeable					
Zinc/Manganese Dioxide	1,5V	$Zn + 2 MnO_2 + H_2O \rightarrow ZnO + 2 MnO(OH)$	Zinc Chloride	++	
Zinc/Air	1,4V	$2 \operatorname{Zn} + \operatorname{O}_2 + 2 \operatorname{H}_2\operatorname{O} \rightarrow 2 \operatorname{Zn}(\operatorname{OH})_2$	Alkaline	(Cathode complicated)	
Zinc/Silver oxide	1,5V	$Zn + Ag_2O \rightarrow 2 Ag + ZnO$	Alkaline	0	
Lithium/ Manganese Dioxide	3,0V	$\rm Li + MnO_2 \rightarrow MnOOLi$	Organic (aprotic)	- Humidity/Water sensitive	
Rechargeable					
Nickel/Metal hydride	1,2V	Metal-H + 2 NiOOH \rightarrow Metal + 2 Ni(OH) ₂	alkaline 25%KOH	0	
Lithium-Ion	3,7V	$Li_{1\text{-}x}Mn_2O_4 + Li_xC_n \rightarrow LiMn_2O_4 + nC$	organic	- Humidity/Water sensitive	
Zinc/Air	1,45V	$Zn + 1/2 O_2 \rightarrow ZnO$	alkaline	(Cathode complicated)	
Post Li	different	Different systems using Air-, Sulphur- Cathode or solid state components	organic	today impossible	

It is hard to tell which system is the best for which application. For low-cost or single use applications primary cells are best suitable. Long term use or more expensive applications match better with secondary batteries. The more complex the systems are a charge management system can be helpful. Self-discharge can be a problem especially of NiMH systems. The systems differ in energy density and power density.

The basic design of a printed thin film battery is typically a sandwich-type arrangement as shown in Figure 9.



Figure 9 Basic design of single cell in stack or sandwich design

The printing technologies provide easy and cheap manufacturing processes for mass production of such batteries. The layer thicknesses are as follows:

- Current Collector $10-15\mu$ m
- Electrodes $100-150\mu$ m

The Separator/Electrolyte layer can either be a fleece soaked in electrolyte or it can be printed, too. Then the electrolyte must ideally be brought into a gum-like state. The electrolyte should penetrate a bit into the electrodes for better performance.

Since the coarseness of the particles in the anode and cathode are in the range of 10 up to 50μ m the only printing technique that is suitable for applying these materials is screen printing.

By using the printing technologies it is easy to expand the single cell to a series connection as shown in Figure 10.

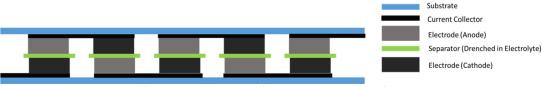


Figure 10 Principal layout of series connection of 5 single cells.

The layout for single cells as used at the research group IAD at the University of Applied Science, Hochschule der Medien in Stuttgart, Germany is shown in Figure 11, where several cells has been put on a drying trolley after printing the electrodes. The next step for these cells is the insertion of the separator/electrolyte and the folding/flapping both sides together.

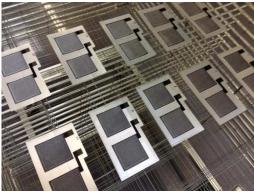


Figure 11 Printed cells on a drying trolley.

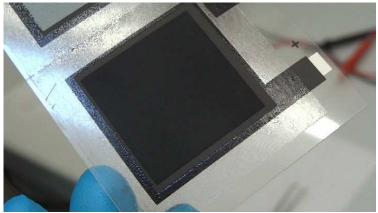


Figure 12 Printed glue for sealing the cells.

One of the biggest problems that needs to be solved for stable printed batteries is that the electrolyte separator has to be kept humid for the lifetime of the battery cell. Therefore, first the substrate must have sufficient barrier properties. For instance PET alone is not quite impermeable enough. Also, for alkaline KOH electrolyte it is necessary to keep the CO2 out in order to avoid carbonisation. Laminate foils, e.g. PET/Al/PET perform much better, but maybe not flexible enough. Second, where the substrates are in contact after the folding together the contact area must be sealed. Therefore, as shown in Figure 12, a layer of glue is printed around the electrodes. The glue is activated by pressure and/or heat.

3. Results and Discussion

The most important property of a battery is the ability of providing energy for the electrical load of the electronic device. The performance can be summarized in simple discharge curves, see Figure 13. These are curves of the voltage vs. time. Typically there is an initial loss of voltage in a very short period of time then there is more or less a plateau and the end of lifetime is characterized by a final,

steep drop. The shape of the curves is depending on the type of load (either constant current or constant resistor). For secondary batteries the curve is expanded by a charging period. Charging and discharging are repeated what is called cyclization.

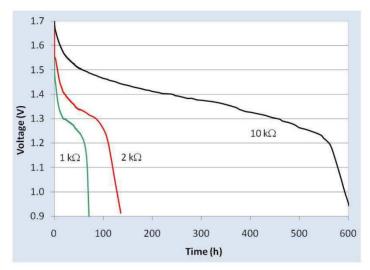


Figure 13 Discharge curves of SoftBattery® 1,5V with various loads at room temperature. Source: http://files.kotisivukone.com/enfucell.kotisivukone.com/tiedostot/discharge.pdf

A more sophisticated way to characterize batteries is the EIS-method (electro impedance spectroscopy). Figure 14 shows how a typical result for a battery with the assumed equivalent circuit diagram could look like. It is a plot, the so called Nyquist plot, of the complex impedance Z, where the impedance is measured during a frequency sweep from very low frequencies 0.01Hz to frequencies around 10kHz. Figure 14 also shows an equivalent circuit diagram comprising of three ohmic resistors (internal resistance) two capacitors and the Warburg impedance.

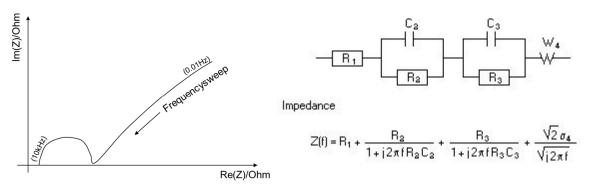


Figure 14 left: Nyquist plot for batteries; right: equivalent circuit diagram

For high frequencies (> 1 kHz) the current flows through C_2 and C_3 completely. W_4 is also not that important. If R_1 is low high currents are possible.

For middle range frequencies (1 - 1000 Hz) the small capacitance starts blocking the current and the impedance increases. For low frequencies (0.01 - 1 Hz) both capacitances block and the impedance is determined by the resistors R_1 , R_2 and R_3 .

For longer periods of time the Warburg-Impedance starts playing a role due to diffusion processes. At the conference more details of the composition of the printing pastes and the electrochemical results will be presented.

4. Conclusions

The printing technologies provide easy and cheap manufacturing processes for mass production of printed batteries.

One of the final achievements of the project BatMat was the demonstration of reel to reel manufacturing of printed batteries at Schreiner Group, Munich. The following images (Figures. 15 to 18) are taken from this manufacturing experiment.



Figure 15 Reel to reel screen printing press with large dryer.



Figure 16 Flapping/folding for finishing the battery.

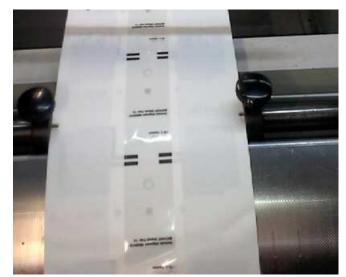


Figure 17 Finalized cells two double cells side by side.



Figure 18 The final Roll-to-roll printed Primary Zinc/Carbon battery

These demonstrators won the first price at LOPE-C 2014 in the category "Best Publicly Funded Project Demonstrator".

The future perspectives of the printed batteries seem to be quite promising. After the publication of the winning of the award a lot of request could be noticed.

A very interesting application of the printed batteries is being developed in the project KoSiF, (complex systems in foil). Figure 19 shows the concept of integrating a lot of functionality on one foil and Figure 20 shows the application with the foil attached to the skin of a bionic gripper. The complex system detects the bending curvature of the gripper and sends information to a base station.

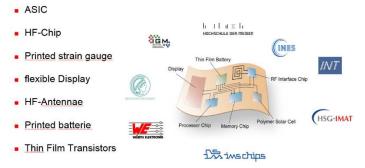


Figure 19 Concept of project "KoSiF" complex systems in foil driven by a flexible, printed battery



Figure 20 Demonstrator "Smart Skin" functionalities on foil attached to a bionic gripper

Other activities towards the commercialization of printed batteries:

Enfucell has showcased a cold chain monitoring system for biopharmaceutical products. Together with the Dutch electronics enterprise NXP, Enfucell has developed a disposable temperature logging foil demonstrator containing NXP's NHS3100 IC, Enfucell's printed battery and an NFC antenna.

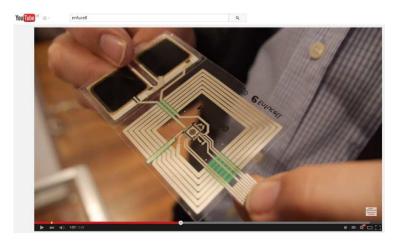


Figure 21 Temperature logger (screenshot taken from www.youtube.com/watch?v=HTXaa317P0s)

Further, a sensor patch has been shown by Enfucell that is attached to golf clubs and measures data like the hitting forces. It communicates with a laptop via Low Energy Bluetooth (BLE).



Golf sensor patch powered by Enfucell SoftBattery

Figure 22 Sensor patch for golf club (screenshot taken from www.youtube.com/watch?v=3eQBKi7Vl-

The future of printed batteries will show more sophisticated series connections up to 10 cells and more and the introduction of other chemical systems like zinc-air, especially when unwanted dendritic growths can be mastered. Lithium based systems still will have the problem that special dry and oxygen free atmospheres are needed for printing, making the process difficult. In a range of several years from now so called post-lithium systems may have achieved a state that is mature enough in a way that printing can be considered.

End 2014 a project run by VARTA and partners called FastStorageBW started that will develop so called PowerCaps. The technology of the PowerCaps is located in-between batteries and supercaps. They will not quite have the capacity of batteries but can be charged and discharged almost as fast as (super-)capacitors. The electrodes of the PowerCaps comprise of thin carbon based layers. The printing technologies may play a major role here, too.

References

Linden, D., Handbook of batteries McGraw-Hill handbooks. McGraw-Hill, New York, 2nd edition, 1995.

Wendler M., Claypole T.C., Steiner E., Krebs M., "Performance optimization of fully printed primary (ZnMnO²) and secondary (NiMH) batteries", *Journal of Print Media Technology Research*, Vol. 4 (2014) pp 241-251.

Wendler M., Krebs M., Huebner G. "Development of Printed Thin and Flexible Batteries", IC International Circular of Graphic Education and Research, The International Circle of Educational Institutes for Graphic Arts, *Technology and Management*, No 4 (2011) pp 32-41.

Wendler M., Krebs M., Huebner G., "Screen Printing of Thin, Flexible Primary and Secondary Batteries", *Advances in Printing and Media Technology*, Vol. 37 (2010) pp 303-312.

Sun K., Wei, T. S., Ahn, B.Y., Seo, J.Y., Dillon, S.J., Lewis, J.A., "3D Printing of Interdigitated Li-Ion Microbattery Architectures", *Advanced Materials*, Vol 25(33) (2013) pp 4539–4543.

Huebner G., Krebs M., "Printed, Flexible Thin Film Batteries and Other Storage Devices", Chapter 18 in "Handbook of Flexible Organic Electronics" edited by Stergios Logothetidis, Woodhead Publishing, 01/2015 pp 429-447

Zinc Oxide-Based Inks for Semi-Conductive Applications

Shreyas Pathak, Alexandra Pekarovicova, Paul D. Fleming

Western Michigan University, Center for Ink and Printability A-231 Parkview, Kalamazoo MI 49008

E-mail: shreyas.pathak@wmich.edu, a.pekarovicova@wmich.edu, dan.fleming@wmich.edu

Short Abstract

ZnO inks were doped with metals, such as aluminum or silver, to observe if small additions of these metals have any effect on semiconductive electrical properties of ZnO inks. The inks were printed on PET substrates on a Gravure K-Proofer and sintered using a NovaCentrix® PulseForge 1200. The formulated inks exhibited semiconductive behavior and the study pointed out that selection of binder and doping levels affect the semiconductive performance of the ink. ZnO was doped with nanosilver ink, which reduced its band gap and allowed it to perform as a semiconductor. However, the nanosilver ink used for doping affected the printability of ZnO ink causing a mottled appearance.

Keywords: semiconductive ink, ZnO, silver dopant, band gap

1. Introduction and Background

Printed Electronics (PE) brings together printing and electronics using conventional printing processes, such as gravure, flexo, screen printing and inkjet processes by modifying their ink components so that they can be used to produce transistors or diodes, which are building blocks for almost all electronic products today (Hrehorova, 2011; Bhore, 2013). Applications of PE have been demonstrated previously in the manufacturing of batteries, LED's, displays, speakers, sensors and fully printed RFID labels (Kattumenu, 2011).

Conventional printing technologies, being additive in nature, offer many advantages over the processes that are currently being used for manufacturing electronic devices. Single or multiple layers of functional materials that are conductive, semiconductive, resistive or dielectric can be deposited on flexible substrates to produce flexible electronic devices. Currently, such electronics incur a high manufacturing cost and low throughput compared to printing when manufactured using processes, such as physical and chemical vapor deposition, sputtering and spin coating. Printing the same features offers high throughput and less expensive investments. With more focus on advancing process refinement and control in printing processes, they can be elevated as highly efficient manufacturing platforms (Emerson, 2009). Moreover, printing of electronic devices can be done using different flexible and rigid substrates using a variety of conductive materials in the inks.

With all the focus on developing printing as a platform for high volume manufacturing of electronics, development of inks for the different processes has also taken a center stage.

Functional inks suitable for gravure and flexographic printing processes have been developed recently and have demonstrated compatibility with novel thermal processing techniques, such as photonic sintering (Lim, 2013). Advances in the field of ink technology such as particle and binder manufacturing have enhanced performance of inks in terms of conductivity, flexibility, line resolution and their compatibility to various substrates. Amongst all the conductive metals, silver is the primary choice for PE due to its high conductivity ($\sigma = 6.3 \times 10^7 \ \Omega^{-1} \text{m}^{-1}$), stability and performance. Most silver inks were made of micron sized silver flakes until the recent introduction of nano-silver inks. These nano-silver inks have the capability of good print quality and can be cured at lower temperatures compared to their larger particle counterparts (Gordon, 2014). Other highly conductive metals that have also found use in PE are copper ($\sigma = 5.96 \times 10^7 \ \Omega^{-1} \text{m}^{-1}$), gold $(\sigma = 4.4 \times 10^7 \ \Omega^{-1} \text{m}^{-1})$ and aluminum $(\sigma = 3.78 \times 10^7 \ \Omega^{-1} \text{m}^{-1})$. For applications that do not require very high conductivity, conductive carbon based inks are used (Gordon, 2014). At present, silver metal-based ink is the most widely commercially available conductive ink for PE (at least 10 companies across the world are manufacturing silver inks). The main challenge today is to replace silver by less expensive metals, such as copper and aluminum, however, they oxidize at ambient room conditions (Kamyshny, 2011). For the manufacture of semiconductors, zinc oxide (ZnO) seems to be a promising material (Girgis, 2013). The past 10-15 years has witnessed a lot of research activity in regards to the use of ZnO as a semiconductor (Janotti, 2009). ZnO is a well-known n-type semiconductor, which is useful for some applications. Doping impurities in ZnO can make a p-type semiconductor, which increases its potential in many more applications. Previously, dye-sensitized solar cells have been successfully fabricated by the inkjet printing of ZnO as one of the components. In modern electronics, semiconductors are the basic foundation in many devices, which include transistors, solar cells, light emitting diodes (LED's), etc. Semiconductors exhibit a number of properties like passing current in a single direction, variable resistance and sensitivity to heat. Conductivity in semiconductors occurs through the unrestricted movement of free electrons and holes collectively known as charge carriers. Based on which charge carrier is in majority, semiconductors are classified as p-type (higher hole mobility) and n-type (Higher electron mobility). By adding impurities to a certain type of semiconductor, it can be converted to the other type and vice versa (Ray, 2014). Some of the properties of ZnO that make it suitable for using in semiconductor applications are as follows (Janotti, 2009):

Direct wide band gap: At low temperatures, the band gap of ZnO is 3.44eV and 3.37eV at room temperature. This enables its application in optoelectronics in the blue/UV region including light-emitting diodes, laser diodes and photo detectors.

Large excitation binding energy: The free-excitation binding energy of ZnO is 60 meV as compared to 25 meV in GaN. This indicates that with such large and efficient binding energy, excitonic emission in ZnO can persist at room temperature and higher. Since the oscillator strength of excitons is much larger than a typical direct band semiconductor, the large exciton binding energy makes ZnO a promising material for optical devices (Janotti, 2009).

Strong Luminescence: ZnO has strong luminescence in green-white region of the spectrum thus making it a suitable material for phosphor applications. The characteristic n-type conductivity of ZnO makes it suitable in vacuum fluorescent display and field emission display applications.

Strong sensitivity of surface conductivity: ZnO thin films are very sensitive to the exposure to various gasses. This property enables it to be used as a less expensive smell sensor to detect the freshness of foods and drinks (Janotti, 2009).

High Thermal Conductivity: ZnO is used as an additive due to its property of high thermal conductivity. It is added to rubber to increase the thermal conductivity of tires. High thermal conductivity transforms to high efficiency of heat removal during device operation. ZnO can be useful for making temperature sensors, too. Composite films made of $(ZnO)_{1-x}(TiO_2)_x$ can be used for solar cell applications (Girgis, 2013).

In this work, ZnO inks with various dopants were formulated with the aim to create gravure semiconductive ink for printed electronics.

2. Materials and Methods

Zinc Oxide ZnO from Alfa Aesar® was used. Particle size was 74 micron. To make these particles suitable for gravure printing, they were first dispersed in isopropyl alcohol (IPA) and milled by applying 2 passes on the laboratory three-roll mill. Solvent-based acrylic resins Elvacite® 1010 and Elvacite® 2045 were used to formulate the inks. Elvacite® 1010 and Elvacite® 2045 were supplied by Lucite International. Elvacite 1010 is a low molecular weight methyl methacrylate polymer. The key feature of this resin is that it maintains a fairly low viscosity. Elvacite 2045 is a high molecular weight iso-butyl methacrylate resin, has superior binding strength and good burn out characteristics as outlined by the manufacturer. The source of silver dopant was silver ink from InkTec® PR-020. The inks contained silver nanoparticles up to 20% weight. The first phase of the experiments was focused on determining the parameters critical for formulating the ink, printing and photonic curing. Parameters critical to the formulation, printing and curing were evaluated to determine the levels that could be used to design further experiments.

2.1 Ink

In the very beginning of the work, inks were formulated only with ZnO and acrylic resins to observe if plain ZnO ink would exhibit any semi-conductive or resistive behavior. ZnO was available in very large chunky particles that were 44 microns or less as per the manufacturer specifications. In order to formulate gravure printable inks, these particles were first dispersed in isopropyl alcohol (IPA) using a disperser at high speed for a period of 60- 90 minutes. The dispersed pigments were further passed through a three roll mill at least twice to reduce the particle size to below 2 microns. Inks were further formulated with two different acrylic resins Elvacite® 1010 and Elvacite® 2045. Elvacite® 1010 had a molecular weight between 4000 and 10,000; while Elvacite® 2045

had a MW of 193,000. The resin contents were varied between 4% and 8% and also combined to observe its effect on adhesion and resistivity of ink on PET substrate before and after photonic curing.

Small quantities of aluminum and silver were further used to dope the ZnO inks to see if they had any effect on electrical properties of ZnO. Further into the trials, the inks were doped with nano silver (Ag) ink InkTecTM PR 20 and aluminum flakes in small percentages ranging from 1% to 5%. Drawdowns were taken in each case using Mayer rod no. 26 and ink films were cured by photonic sintering and tested using a two probe Keithley multi meter for resistance properties. Adhesion was tested by performing tape tests using 3M #610 Scotch tape, both before and after curing. The initial formulations for ZnO with the acrylic resins and silver inks were as shown in the Table 1.

Experiment – Low Mol. Wt. Resin Elvacite® 1010					
Components	NameAmount (%)Dry Wei		Dry Weight (g)		
Filler	ZnO	20 - 30	32.5 - 48.75		
Binder	Elvacite 1010	4 - 8	8.12 - 16.24		
Dopant	Silver and aluminum	1 – 5	1.62 - 8.1		
Solvent	IPA: Ethyl Acetate	75 – 57			
Target Solids (%)		25 - 43			

Table 1: Ink Formulations using Elvacite® 1010

Experiment -High Mol. Wt. Resin Elvacite 2045					
Components	Name	Amount (%)	Dry Weight (g)		
Filler	ZnO	21 - 30	32.5 - 46.4		
Binder	Elvacite 2045	4 - 8	6.5 - 13		
Dopant	Silver and aluminum	1 – 6	1.62 - 9.72		
Solvent	IPA: Iso Propyl Acetate	73.9 - 56			
Target Solids (%)		26-44			

Table 2: Ink Formulations using Elvacite® 2045

Experiment -Combination of High and Low Mol. Wt.					
Components	NameAmount (%)		Dry Weight (g)		
Filler	ZnO	25	32.5 - 46.4		
Binder	Elvacite 1010	4	8.12		
Binder	Elvacite 2045	2-4	3.25-6.5		
Dopant	Silver	2 - 6	1.62 - 9.72		
Solvent	IPA: Ethyl Acetate : Isopropyl Acetate	67.94 - 61			
Target Solids (%)		33 - 39			

Table 3: Ink Formulations combining Elvacite® 1010 and Elvacite® 2045

Inks were formulated step by step to determine the low and high levels of every factor. The inks were printed on the gravure K- Proofer to determine the printability of inks. Inks without doping seemed to perform well on the proofer, while the same inks with doped silver or aluminum showed a highly mottled print. Trials on the proofer also helped determine the solvent ratio to be used in the main experiments. For the low molecular weight a combination of isopropyl alcohol and ethyl acetate was used in a 90:10 ratio respectively, while for the higher molecular weight formulations isopropyl alcohol and isopropyl acetate was used in an 80:20 ratio. This phase helped determine the low and high levels of every ingredient of ink as well as helped determine factors such as the target solid contents and viscosity required for the performance of inks.

One of the most important observations in the grinding stage of making the ink was that the particles being fairly large initially, it was difficult to grind since particles did not hold together and most of the solvent was lost by evaporation in the process. 2% binder was further introduced to the initial dispersion in order to hold the particles together during the grinding process on the three roll mill. For all the further formulations, this practice were kept constant.

2.2 Gravure Printing

With all the initial formulation work, printing was carried out on the K-Proofer mainly to determine which parameters of the engraved plate deliver the best results especially given the larger particle size of ZnO. Printing was carried out on 200 μ m thick PET substrates that were UV treated using the UVO oven to enhance PET surface energy. Screen ruling of 100 LPI and 160LPI was employed. Both these plates were engraved with a 120° diamond stylus tip at 45° compression angle.

2.3 Photonic Curing

The NOVACENTRIXTM Pulse Forge 1200 has a number of parameters, which determine the effect of curing. The SimPulseTM software package in this stage was quite helpful in determining the approximate temperature that ZnO could reach on the given substrate and condition. Based on the simulation, speeds of 10 feet/min, 15 feet/min and 20 feet/min were set in combination of various parameters, such as diode voltage between 300 – 430 V and pulse length between 600 – 1000 μ s of the basic pulse. Various combinations were tried in order to sinter both inks before and after doping. Since doping was done with silver inks that had a lower melting temperature than zinc oxide, the parameters were optimized such that they could accommodate the highest level of doping without affecting the substrate and the material to burn off from the substrate. Experiments not only helped determine the optimum sintering parameters, but also helped determine the highest level of silver ink that could be used for doping and cured without burning the material. Following are the set of parameters as well as different levels that were investigated in combinations and conditions:

Parameters	Levels
Diode Voltage (V)	300 - 430
Pulse Length (μ s)	600 - 1000
Web Speed (feet/min)	10 – 20
Web Height (mm)	25
Energy (W/cm ²)	3.5 k – 5.2 k
Sim Pulse® Temperature (°C)	1070° C

Table 4: Sintering Parameters for PulseForge® 1200

Design of experiments (DOE) was done using the MINITAB[™] statistical software. A full factorial DOE with three factors at 2 levels resulted in 8 different runs:

Run no.	ZnO (%)	Resin (%)	Doping (%)
1	30	7	2
2	30	7	4
3	22	7	4
4	22	4	4
5	30	4	2
6	30	4	4
7	22	7	2
8	22	4	2

Table 5: Design of Experiments (DOE) runs with three factors at 2 levels

The sintering process excites the electrons in the zinc oxide films from the valence band to the conduction band. As a result, the films exhibit resistive behavior for a very short period. The response for the above experiment was an average of five readings recorded immediately after sintering for every run. Readings were also recorded after intervals of 2 minutes and 4 minutes to observe how stable the inks were in terms of resistance readings after the particular time periods. The best combination of parameters obtained from the main effects of the DOE was replicated using ZnO and nano silver inks to observe if there was any similarity in the behavior of nano particle formulations and formulations having larger particle size. While performing the runs of DOE and making drawdowns it was made a common practice to treat the PET substrates using the UVO cleaner before using it for either printing or drawdowns.

3. Results and Discussion

The best trial from the DOE (Table 5) was repeated using nano ZnO particles and doped with nano silver inks. While sintering, the diode voltage had to be reduced to 400V from 430V, since the edges of the nanoparticle inks appeared burnt. The nano particle ink exhibited slightly better resistance readings upon sintering. This also showed that the nano particle ink required a lower curing temperature as compared to the micron sized particles, which points to the fact that particle size of ZnO might play an instrumental role in the properties of semiconductivity.



Figure 1: Nano ZnO ink sintered at original settings and after reducing diode voltage

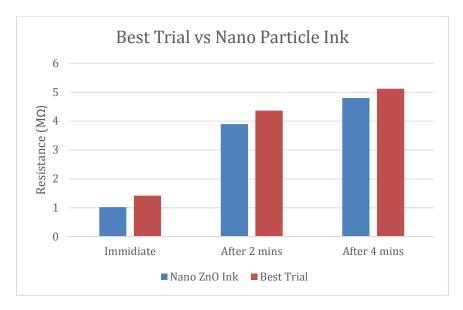


Figure 2: Resistance comparison between nano ZnO inks and best ink from DOE trial

Figure 2 shows the comparison of the best trial from the DOE with the same composition ink using nano particle ZnO.

3.1 UV-Vis Spectroscopy

The UV-Vis spectroscopy showed a similar trend in absorbancy of plain ZnO drawdown on PET as the analysis of powdered ZnO. Plain ZnO did not exhibit substantial absorbancy in the visible range of 400- 700 nm. However, moving towards the shorter wavelengths and wavelengths below the visible range, the absorbancy increased substantially. The band gap was calculated based on the cut – off wavelength, which was found to be 3.11 eV as compared to 3.37 eV reported by Janotti (2009). The "noise" at shorter wavelengths results from electronic transitions of higher energy than 3.11 ev, but the edge is clear.

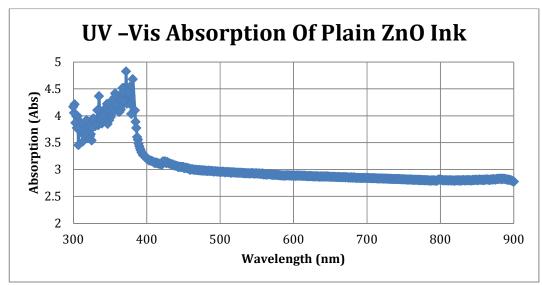


Figure 3:UV-VIS absorption spectrum of plain ZnO ink

The UV- Vis spectral readings were recorded for all the inks and it was observed that all the doped inks exhibited absorption in the visible, which pointed out that doping with silver ink did lower the band gap. However, it was more important to observe and compare the absorption behavior of the best DOE trial (Ink 4) as well as the ink formulated with the nano ZnO. On comparing both the inks showed very similar absorption trends in the visible range. At the same time, the absorption curves were plotted showing plain ZnO nano ZnO and Ink 4 on the chart as well to show the reduction in band gap.

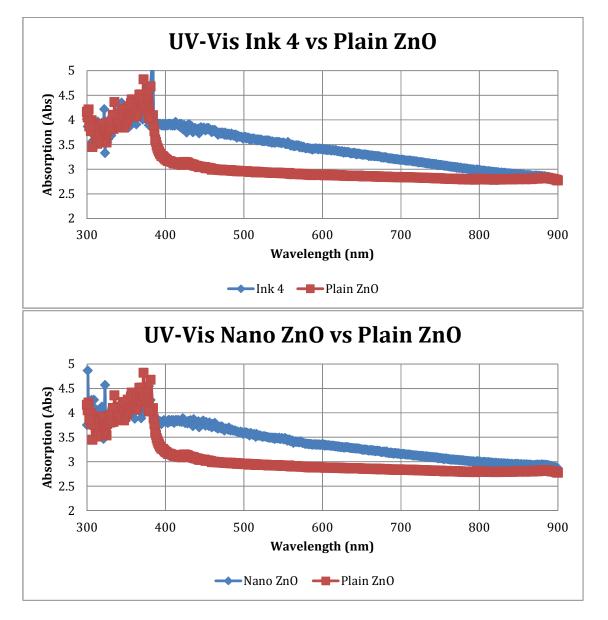


Figure 4: UV-VIS Spectrum of best ZnO ink and doped nano ZnO ink

The results indicate that ZnO inks exhibit semiconductive behavior with reduced bandgap when doped with nano silver. The fact that the inks exhibit electrical conduction when sintered at high temperatures and eventually diminish, confirms semiconductive behavior. Silver metal can be thus used to effectively reduce the band gap of the zinc oxide semiconductive ink. Previous research of doping ZnO with silver has shown that higher doping content has been effective in substantially reducing the band gap of ZnO. The color of the doped ZnO also gets darker when the doping level is increased, which is a similar observation to that of the previous research. ZnO has a wide bandgap that can only be activated by UV light of wavelength below 385 nm. However, on doping ZnO can be activated at wavelength above 385 nm too. The UV-VIS absorption spectra in this study also showed the same trend pointing out that silver doping in ZnO ink formulation effectively reduced the band gap.

4. Conclusion

Various solvent based ZnO inks were formulated, gravure printed, sintered by photonic curing and tested for semiconductive behavior. Only silver doped ZnO inks demonstrated significant absorption in the visible spectrum at 400 - 700 nm pointing out to the fact that doping with silver reduced the band gap of ZnO, and enhanced semiconductivity.

References

Bhore S.S.: Formulation and Evaluation of Resistive Inks for applications in Printed Electronics, Western Michigan University, Master's Thesis, 2013.

Emerson, N. J. Investigation of the Effects of Process on Performance of Gravure Printed ITO on Flexible Substrate. Woodruff School of Mechanical Engineering: Georgia Institute of Technology. Master's Thesis, August 2009.

Girgis E., Fang Mei, Kathab N., Hassan E., Rao K.V., *Ink-jet-printed* $(ZnO)_{1-x}(TiO_2)_x$ composite films for solar cell applications. J. Mater. Res., Vol. 28, No. 3, Feb 2013. Cambridge Journals, pp. 502-505.

Gordon S.E., Dorfman J.R., Daniel K., Kerry A.: *Advances in Conductive Inks across Multiple Applications and Deposition Platforms*. DuPont Microcircuit Materials. As published in IPC Printed Circuit Expo, APEX & Designer Summit Proceedings, Accessed January 15, 2014.

Hrehorova E., Rebros M., Pekarovicova A., Bazuin B., Ranganathan A., Garner S., Merz G., Tosch J., and Boudreau R., *Gravure Printing of Conductive Inks on Glass Substrates for Application in Printed Electronics*. Journal of Display Technology, Vol. 7, No. 6. June 2011, p. 318.

Janotti A., Van de Walle Ch. G., *Fundamentals of Zinc oxide as a semiconductor*. Reports on Progress in Physics, Vol. 72 (2009) IOP Publishing, p. 29.

Kamyshny A., Steinke J., Magdassi S.: *Metal-based Inkjet Inks for Printed Electronics*. The Open Applied Physics Journal, 2011. 1874-1835/11. P- 19-36.

Lim, S. Joyce, M., Fleming, P.D., Aijazi, A., and Atashbar, M., Inkjet Printing and Sintering of Nano Copper Ink, Journal of Imaging Science and Technology, 57 (5) Sept.-Oct. 2013.

Kattumenu, R., Rebros, M., Joyce, M., Hrehorova, E., and Fleming, P.D.: Evaluation of Flexographically Printed Conductive Traces on Paper Substrates for RFID Applications. *TAGA J.*, 4, 219, 2011.

Ray W.J., "Printed Semiconductors". Nth Degree Technologies. Semiconwest.com. February 2014.

42nd International Research Conference, 06 to 09 September 2015



Session 3B

User and audience experience 1

Monday, 7 September 2015 16:05 – 17:45

Chair: Leopoldina Fortunati

Christin Götz¹, *Ulrich Nikolaus*

Leipzig University of Applied Sciences (HTWK Leipzig) Gustav-Freytag-Str. 42, 04277 Leipzig, Germany

E-mail: christin.goetz@arcor.de, ulrich.nikolaus@htwk-leipzig.de

Short Abstract

Cascading Style Sheets (CSS) have already been the *de facto* standard for the visual representation of digital content for some time now – but new features intended for the formatting of printed layouts have been added only recently. With CSS level 3, which is still under development, several new features have been added to the standard, such as, for example, the definition of marginalia, footnotes, running heads or the support for advanced micro-typographic settings like OTF-features. In theory, these new features could be the key to a significant simplification of cross media publishing, based only on the use of XML or (X)HTML and CSS 3. In this paper, the current status of implementation of CSS 3 features for the formatting of XML-based print layouts is discussed and its current support by rendering engines analysed. The results suggest that CSS 3 can be used at present for the formatting of simply structured content, but not for visually or semantically complex print layouts.

Keywords: CSS 3, cross media publishing, electronic publishing

1. Introduction and background

The more the publication of e-books or other digital content formats becomes firmly established, the more important effective procedures for multichannel publishing become for publisher competitiveness (Kleinfeld, 2013; Ott, 2014; Quin, 2014). Today, print layouts can already be generated automatically by applying the eXtensible Stylesheet Language-Formatting Objects (XSL-FO) to media independent data, often stored using the eXtensible Markup Language (XML) (Ott, 2014; Quin, 2014). However, XSL-FO has never become very popular within the publishing industry (McKesson, 2012; Ott, 2014). Cascading Style Sheets (CSS), on the other hand, are extremely popular for the formatting of web content as well as e-books, but has up till now hardly been used for the formatting of print layouts. Nevertheless, an (albeit rather limited) *paged media* model has been added to CSS level 2 as early as 1998 – which in principle makes it possible to generate and format print layouts out of HTML or XML content using CSS as well (Ertel & Laborenz, 2014; Harold & Means, 2004; Lie & Day, 2005).

Since 1998, the CSS print layout functionality has been improved gradually but continuously by the World Wide Web Consortium (W3C). Thus, the current version CSS 3 (with its modular design intended to simplify parallel development, cf. Meyer, 2012) – comprises several CSS modules defining several new rules, functions, properties, values and selectors which can be used for the automatic generation of print layouts (Graham, 2014; Quin, 2013). Amongst others, CSS 3 includes modules to

- define and design multi-column layouts for elements in the CSS Multi-column Layout Module (W3C, 2011a),
- define color and opacity using different color models like RGB, HSL or CMYK in the CSS Color Module Level 3 and 4 (W3C, 2011b, 2014a) or
- make various micro-typographic settings for instance in the CSS Fonts Module Level 3 (W3C, 2013b) and the CSS Text Module Level 3 (W3C, 2013c).

¹ corresponding author

Apart from these more general CSS 3 modules that can be used for other purposes (such as the formatting of digital content) as well, CSS 3 introduced several new modules that are of particular importance for formatting print layouts, such as:

- the CSS Paged Media Module Level 3 (CSS3PAGE): CSS3PAGE describes a page model (or box model) for print layouts and defines rules, properties and special page selectors for the formatting of paged media (e. g. the definition of page size, orientation and margins) or the customization of headers and footers (size, styling and positioning). Furthermore, it allows for the positioning of content such as page counters in headers and footers (W3C, 2013a).
- the CSS Generated Content for Paged Media Module (CSS3GCPM): This module contains features to generate and to place content in special page areas automatically and to add, for instance, running heads, footnotes and cross-references to paged media (W3C, 2014b).
- the CSS Fragmentation Module Level 3 (CSS3BREAK): CSS3BREAK includes properties to control pagination and defines fragmentation rules for page and text breaks which should be observed and applied when generating a static layout (W3C, 2014c).

2. Methods

The objective of this paper was to explore how the new features of CSS 3 can be used to render paged media layouts out of media independent data. In order to achieve this aim, a constructional approach was used: First, structure, content and design of thirty textbooks and other non-fiction books (only Latin characters) with complex print layouts were analysed to identify specific design elements that characterise paged media. Based on this analysis a test document was designed to combine all these features in one single book. Next, a media independent XML file describing content and structure of the test document and a CSS 3 document containing the corresponding paged media formatting instructions were prepared. The content and layout files were validated (using both validation tools of the *<oXygen/>*XML editor and the W3C's online CSS Validation Service) to ensure the conformity of the XML content file with the semantic language DocBook V5 and the CSS layout document with the specification of CSS 3. From these two files, a paged media PDF document was generated automatically using two different rendering engines: *YesLogic Prince* and *Antenna House Formatter* which already support the formatting of XML-based PDF documents by using CSS 3 (for more detailed descriptions, cf. Götz, 2014). Finally, the generated PDF documents were analysed and the conformity of the resulting paged media layout to the initial layout specification was tested.

3. Results and Discussion

In contrast to previous CSS versions, CSS 3 includes a wide range of new elements, properties and selectors to format complex structured print layouts. The CSS 3 modules support various new macroand micro-typographic settings for print layouts and provide a formatting functionality for paged media that is comparable to professional typesetting software applications, such as creating and designing running heads and footnotes, adding marginalia, defining baseline grids, floating images or specifying advanced typographic settings for Open Type Fonts (OTF).

However, the test results show that currently less than half of these new functionalities are fully supported by the two rendering engines named above (cf. Table 1) – although these are among the most sophisticated tools available to date to generate paged media PDF documents out of XML content. Thus, the test results were all in all dissatisfying.

On the other hand, the test results show that, for instance, CSS 3 functionalities for the automatic generation of content like paginations, footnotes, running heads and numbered cross-references are

already supported; which cannot be said, however, for the automatic creation of catchword indexes or tables of contents.

Analysed new features of CSS 3 for formatting print layouts		Full S	upport	Partial Sup- port No Suppo		oport	
		PR	AH	PR	AH	PR	AH
Automatic generated content	13	6	6	6	6	1	1
Page settings	19	7	7	3	4	9	8
Paragraph, word and charac- ter formatting	44	9	15	5	8	20 ⁽¹⁾	20 ⁽²⁾
Fragmentation options	5	0	2	1	1	4	2
Settings for printing and distri- bution	4	2	3	1	1	1	0
Selectors	16	16	16	0	0	0	0

Table 1: Tested categories and excerpt of results (for more detailed descriptions cf. Götz, 2014)

Legend: PR: Rendering Engine YesLogic Prince v9 rev5.0, AH: Rendering Engine Antenna House Formatter V6.2 Evaluation, (1) 10 of the analysed features could not be tested, (2) 1 of the analysed features could not be tested, because the missing support of more basic CSS functions made a rendering impossible

As for page settings, the results show that about 20 percent of these new CSS 3 features are supported, for instance the controlling of page, section, paragraph and character breaks in static layouts. Furthermore, it has become apparent that the rendering engines also support additional proprietary, non-standard CSS properties to control fragmentation, page breaks and hyphenation.

The results for paragraph, word and character formatting are equally mixed. CSS 3 functionality defining color models and controlling image resolution, edge trim, corner marks and bleed is mostly supported, except for the definition of CMYK colors (the rendering engine *YesLogic Prince* uses a proprietary CSS command instead of the one specified by the W3C).

Regarding the new CSS 3 selectors, that can be used, for instance, to format content based on the logical structure of the document tree (such as selecting each third item or bullet in a list etc.) all are fully supported by both rendering engines in the test. This, obviously, simplifies the formatting of specific elements, thus guaranteeing a stricter separation of content and layout between the database file and the CSS document.

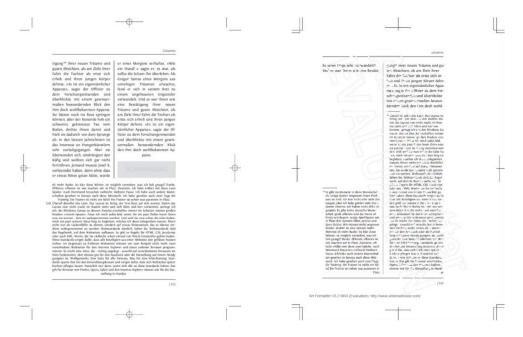


Figure 1: Rendering results for columns of the footnotes area (left: YesLogic Prince; right: Antenna House Formatter)

Apart from that, clear differences between the two rendering engines in the test can be detected. Not only that both support only a limited amount of the new CSS 3 features, but the test results also show that the supported features are quite different. In some cases, certain features are supported by both renderers, but the rendering process is nonetheless different and the same CSS instruction could result in differing print layouts. One example is the rendering of the footnotes area: While the column layout of footnotes rendered using the *Antenna House Formatter* automatically corresponds with the column layout of the main text, the rendering engine *YesLogic Prince* always renders footnotes as a single column, even if the main text has a multi-column layout (cf. Figure 1).

4. Conclusions

The possibility to generate and format both print and digital layouts using only XML (or HTTML) and CSS 3 could be the key to a significant simplification of cross media publishing: the re-use or reprocessing of media-independent data in order to automatically generate websites, e-books and well-designed and complex print layouts from the same media-independent data in a shorter time, with less effort and at reduced costs is an attractive prospect. The W3C standard CSS 3 has the potential to make this vision come true. At present, however, our tests show that only few of the corresponding paged media features are currently supported by the leading rendering engines. For the time being, CSS 3 can already be used to generate works of fiction with a very simple semantic structure, but is not yet suited for the production of complex non-fiction and text books. Somewhat better results can be achieved by using non-standard, proprietary features of the respective rendering engines, thus adjusting the layout settings. But this, of course, contradicts the principle of a standards-compliant, media-independent and flexible production.

Acknowledgments

The authors would like to thank Franziska Bröckl and Nadia Nikolaus for fruitful discussions and proof reading.

References

Ertel, Andrea, & Laborenz, Kai. (2014). Responsive Webdesign (1 ed.). Bonn: Galileo Press (Galileo Computing), p. 58

Götz, Christin. (2014). CSS 3 zur Formatierung für XML-basierte Printlayouts. (Master), Hochschule für Technik, Wirtschaft und Kultur Leipzig, Leipzig, pp. 42

Graham, Tony. (2014, February 14-16). *Formatting from XML*. Paper presented at the XML Prague 2014, University of Economics, Prague, Czech Republic, p. 265

Harold, Elliotte Rusty, & Means, W. Scott. (2004). XML in a Nutshell (3 ed.). Sebastopol: O'Reilly Media, Inc., p. 221

Kleinfeld, Sanders. (2013, August 6-9). *The Case for Authoring and Producing Books in (X)HTML5*. Paper presented at the Balisage: The Markup Conference 2013, Montréal, Canada.

Lie, Håkon Wium, & Day, Michael. (2005, 2005-01-19). *Printing XML. Why CSS Is Better than XSL*. Retrieved 2014-03-28, from http://www.xml.com/pub/a/2005/01/19/print.html

McKesson, Nellie. (2012, 2012-06-12). *Building Books with CSS3*. Retrieved 2014-02-06, from http://alistapart.com/article/building-books-with-css3

Meyer, Eric A. (2012). CSS and Documents. Sebastopol: O'Reilly Media, Inc., pp. 1

Ott, Tobias. (2014). Crossmediales Publizieren im Verlag. Berlin/Boston: Walter de Gruyter GmbH, p. 3, 117

Quin, Liam. (2013, 2013-11-02). *Re: [xsl] xsl 2.0?*. Retrieved 2014-09-27, from http://www.biglist.com/lists/ lists.mulberrytech.com/xsl-list/archives/201311/msg00014.html

Quin, Liam. (2014, February 14-16). *Publishing in Style with XML. Or, Why It's Not XSL-FO*. Paper presented at the XML Prague 2014, University of Economics, Prague, Czech Republic, p. 253

W3C. (2011a, 2011-04-12). CSS Multi-column Layout Module. W3C Candidate Recommendation 12 April 2011. Retrieved 2014-06-11, from http://www.w3.org/TR/2011/CR-css3-multicol-20110412/

W3C. (2011b, 2011-06-07). CSS Color Module Level 3. W3C Recommendation 07 June 2011. Retrieved 2014-06-12, from http://www.w3.org/TR/2011/REC-css3-color-20110607

W3C. (2013a, 2013-03-14). CSS Paged Media Module Level 3. W3C Working Draft 14 March 2013. Retrieved 2014-03-18, from http://www.w3.org/TR/2013/WD-css3-page-20130314/

W3C. (2013b, 2013-10-03). CSS Fonts Module Level 3. W3C Candidate Recommendation 3 October 2013. Retrieved 2014-06-07, from http://www.w3.org/TR/2013/CR-css-fonts-3-20131003/

W3C. (2013c, 2013-10-10). CSS Text Module Level 3. W3C Last Call Working Draft 10 October 2013. Retrieved 2014-06-09, from http://www.w3.org/TR/2013/WD-css-text-3-20131010/

W3C. (2014a, 2014-05-05). CSS Color Module Level 4. Editor's Draft, 5 May 2014. Retrieved 2014-06-12, from http://dev.w3.org/csswg/css-color/

W3C. (2014b, 2014-05-13). CSS Generated Content for Paged Media Module. W3C Working Draft, 13 May 2014. Retrieved 2014-05-19, from http://www.w3.org/TR/2014/WD-css-gcpm-3-20140513/

W3C. (2014c, 2014-01-16). CSS Fragmentation Module Level 3. Breaking the Web, one fragment at a time. W3C Working Draft 16 January 2014. Retrieved 2014-05-29, from http://www.w3.org/TR/2014/WD-css3-break-20140116/

The influence of location-related Factors on the Perception of Billboard Advertising

Janine Spieß, Ulrich Nikolaus^{*}

Leipzig University of Applied Sciences (HTWK Leipzig), Faculty of Media, Gustav-Freytag-Straße 42, 04277 Leipzig, Germany

E-mail: janine.spiess@me.com; ulrich.nikolaus@htwk-leipzig.de

Short Abstract

The impact that printed out-of-home advertisements have on the consumer's perception is dependent on several location-based factors. For a long time, however, this influence was only indirectly gathered by measuring the relative performance of different locations of billboard advertising: Advertising agencies were traditionally only measuring consumer movement patterns and estimating "exposure opportunities", but no real visual contacts. In this paper, the influence of several location-based factors such as distances, viewing angles, etc., used with increasing frequency to improve those estimates, on billboard perception is analyzed using eye tracking technology. The results show that these factors affect human attention in different ways and allow for a more precise comparison of the relative importance of these location-based factors on the consumer's perception.

Keywords: billboard advertising; eye tracking; visual perception; location-related factors

1. Introduction and background

Printed advertisements play an important role in marketing today. The oldest written advertisements in existence date back to the ancient Egypt (Unger et al., 2007, p. 281). Nowadays printed advertisements can be found in a huge variety of forms – as newspaper advertisement or billboard poster, advertising supplement, leaflet or prospectus, catalogue, flyer, sticker and so on; all existing in a huge variety of sizes and formats (Esch, 2015). For consumers, printed advertisements are today ubiquitous, and can be found both at home (in newspapers, journals or as advertising mail) and outside – as posters, lamppost banners or billboards near bus stops or on the side of the road.

In today's world of fragmented media, outdoor advertisement plays an important role in reaching an increasingly elusive consumer (Wilson and Till, 2011, p. 909). Out of home advertising is one of the fastest growing media segments (OAAA, 2015, p. 1), and especially billboards or posters that can often be found in high-traffic areas on the side of the road account for 65 % of the industry revenue in the U.S. (l.c.). In Germany, backlit billboard posters represent more than a quarter of the overall market for outdoor advertising today (Meffert, Burmann and Kirchgeorg, 2012, p.637). Billboard advertising provides high levels of reach and frequency at a lower cost than other media (Lane, King and Reichert, 2010, p. 371). The contact opportunities are higher than those of other classic advertising media because the majority of the population leaves their house at least once a day (Anspach, 2004, p. 7), so that even consumers that are very mobile and exposed less frequently to traditional forms of advertising, billboard posters cannot be switched, turned off or skipped (Anspach, 2004, p. 7) or interfere with any editorial content – which is probably why it enjoys a comparatively high level of users' acceptance (l.c.; Meffert, Burmann and Kirchgeorg, 2012, p. 651).

^{*} Corresponding Author

Apart from the advertising message and the graphical design of an advertising poster, the location of the billboard on which the poster is presented has important implications for the success of an advertisement as well: While the poster contains the actual information, the backlit billboard has to carry the message on to the target group the advertiser wants to reach (Schweiger and Schrattenecker, 2009, p. 300). Thus, advertising media bridge the gap of space and time between sending and receiving of an advertising message (Pepels, 2005, p. 21); they form the context of the advertisement (Felser, 1997, p. 267). How efficient this can be done may depend on multiple factors, such as the environment in which the billboard partly hidden by any obstacles? What is the viewing angle or distance the advertisement is looked at?

Factors like these might influence the impact of out-of-home advertisements, and consequently, the advertising providers assign different performance/price categories depending on the respective placement of their billboards (cf. Bloom, 2000, p. 396; APG|SGA, 2015; Prosser, 2013). In Germany, for instance, the outdoor advertising trade association (Fachverband Außenwerbung, cf. FAW e.V., 2012) introducedas of January 1, 2013 a method of measurement called PpS ("Plakatseher pro Stelle"; meaning "poster viewers per billboard location" in German). PpS uses GPS data from a representative sample of pedestrians to identify consumer movement patterns (this data is counterchecked by a Last Day Recall via telephone interviews). This value is weighted using seven location-related factors, namely the period of exposure, the linear and lateral position of the billboard in relation to the passing traffic, the degree of occlusion of the billboard's position, the number of competing nearby billboards, the overall complexity of situation and environment and the lighting conditions (l.c., p. 5). Based on these values, the quality of the different billboard locations are determined – and, consequently, also the price.

Sophisticated as this measuring method may be, it still basically determines how many consumers *walk past* a billboard poster, but not how many people actually *look* at it. Even if these *advertisement exposure opportunities* are later weighted using the above-mentioned location-related factors – they are still basically estimates, not real visual contacts. The objective of this paper, therefore, is a more thorough analysis of the perceptual impact that the location-related factors have on the human perception of billboard advertising – to find out if the said weighting really leads to an improved visibility assessment. In order to do this, eye tracking technology has been used, and a test scenario was developed that simulated two slightly different walks through the city of Leipzig. Both walks (recreated using two different slideshows) featured out-of-home campaigning at various locations, from different distances, viewing angles, etc. These two slideshows were presented to two different experimental groups, and their gaze behavior was recorded using eye tracking technology. The eye tracking results allowed a direct comparison of both walks as well as a more detailed assessment of the impact these parameters have on human cognition.

2. Materials and Methods

The study described in this paper consisted of two parts. In the first part, the effects of location-related factors on the test viewers' perception and information processing of billboard advertising were analyzed using eye tracking technology. In the second part of the test, the unaided recall, aided recall and recognition of the selected billboard advertising was tested for each of the test participants using printed questionnaires.

In preparation of the eye tracking part of the test, the locations of all 119 x 175 cm backlit billboards (also known as city light posters; cf. Unger et al., 2007, p. 285; Ströer, 2015) in Leipzig's city center were determined and marked on a Leipzig city map. Next, a walking route through the city was chosen, which provided the basis for the eye tracking test scenario. The route started at Leipzig central station, followed some prominent arterial roads (passing nine city light billboards on the way) and ended at a certain restaurant which was given as the intended destination to the test viewers.

In order to create a low-involvement situation that is typical for the quite unintentional and casual contact with out-of-home advertising (Bloom, 2000, p. 395), the test subjects were left unaware of the real purpose of the investigation. Instead, they were told to memorize the way from the central station to the restaurant in order to be able to describe it later to a friend. In reality, however, the perception of the city light billboards and the influence of the above mentioned location-based factors were tested. In order to do this, two slightly different versions of the walking route were simulated using two series of 65 photographs each that were taken under identic lightning conditions from a pedestrians' perspective (cf. Figure 1). 56 of these photos are neutral stimuli showing ordinary street sceneries that are identical in both versions of the walk. The nine others, however, differ in both versions according to the characteristics of the location-related factors named above, e.g. a high or a low sight distance, frontal or lateral viewing angle, and were each displayed to only one of the groups.

3.1 Subjects

Sixty test participants (28 males, 32 females) were recruited for the experiments and divided into two groups. Their age was 24.5 on average, ranging from 17 to 33. Almost all had normal or corrected vision, except for two individuals with a slight cataract and a red-green deficiency, respectively (which, however, had no negative impact on the results of the study). Although more than three-quarters of the participants attended courses related to media, none of them had special knowledge in outdoor marketing or billboard advertising.

3.2 Stimuli

As the eye tracking system used for this test was computer-based, two series of photos were used to simulate the walking route mentioned above. Each of these slideshows contained 65 photographs (9 of the relevant city light posters and 56 neutral ones of streets), taken from the perspective of pedestrians (cf. Figure 1). The neutral stimuli were identical, whereas the billboard views varied according to the location-related factors, the linear and lateral distance, the degree of occlusion of the billboard's position, the number of competing nearby billboards and the overall complexity of situation and environment. Each slideshow was presented to the test subjects, one picture at a time, each display lasting two point five seconds. The distribution of billboard advertising corresponded to their actual placement in the city, and the order and perspective of the photos also reflected reality.



Series 1:



Figure 1: Samples of the two series of photos used in the eye tracking test. The blue frames (samples (b & f) and (d & h)) mark neutral stimuli that were identical in both series. Green frames test several location-based factors, showing backlit billboards in varying distances (a & e) or number of competing nearby billboards (c & g).

3.3 Apparatus and procedure

The stimuli were presented on a monocular, desktop based NYAN 2 XT/EDGE eye tracking system. User reactions were recorded and their visual scan paths were analyzed. In order to assess the relative importance of the main visual components on the billboard advertising, Areas of Interest (AOI) were defined beforehand to compare hit rates, the time to first fixation, gaze durations, etc.

After the test, the participants were asked to complete a questionnaire, asking them at the outset to name those locations of billboards and all advertising posters shown that they could recall without the aid of a cue or prompt. After that, a list of brand names was given to the test participants. They were now asked to name those brands they thought they had seen earlier. Finally, the advertising posters in the test (and some others that had not been shown) were presented to the test participants once again, and they had to decide for each of them if they had been part of the test or not.

The statements of the survey were analyzed in order to find tendencies and patterns as to which advertising posters could be remembered and to what extent, and how these recognition values were influenced by location-related factors.

3. Results and Discussion

As for the general distribution of attention between the billboard advertising and their environment, the results were largely as expected. In direct competition, the surrounding street life was (on average) not only viewed 4 times longer than the billboards, but it was also fixated 0.5 s faster and over 4 times as often. Of course, it has to be taken into account that the environment occupies much more space than the billboards on most of the images. That supports previous statements that outdoor advertising is a glance medium (Pant, 2007, p. 199) with usually only a very brief exposure (van Meurs and Aristoff, 2009, p. 82), where contact is only casual and quite unintentional (Bloom, 2000, p. 395).

However, 61.5 % of the test participants *did* look at the billboard advertising at any time on average. This is also in line with the statements attributing high levels of reach and frequency (Lane, King and Reichert, 2010, p. 371) and a repeated exposure rate (Donthu, Cherian and Bhargava, 1993, p. 70) to billboard advertising, producing a major impact (Prasad, 2009, p. 15).

Furthermore, test results showed that location-related factors like linear and lateral distance, the number of competing nearby billboards, the overall complexity of the environment and the degree of occlusion do indeed have an impact on the perception of billboard advertising (cf. Figure 2).



(1)



(m)

(n)

Figure 2: Sample heat maps from location-related factors analysis. Those areas of the stimuli that received most attention from the test participants are marked with colored spots (red means a high level of attention, followed by yellow and green). Areas that got minor attention are darkened.

For the two factors occlusion and environmental complexity, results were more or less as expected. Billboards that were partly hidden by other objects such as, in this case, a street sign or pedestrians, got a considerably lower level of attention (cf. Figure 2 k). The corresponding ad was perceived by much fewer participants (on average only 64 % of the subjects fixated partly hidden billboards at least once, compared to 90 % fixations for the same ads without occlusion), and both the gaze duration and the fixation count decreased by over 50 % on average. Considering the direct influence on physical visibility, this was hardly surprising.

As for environmental complexity, it was to be expected that billboards positioned in low complexity situations would get higher levels of attention than those with a higher environmental complexity. Fittingly, the results showed a positive effect on the attentional values for billboards placed in a low environmental complexity. On the other hand, an increased environmental complexity seems to have a lower impact on the level of attention than might be assumed. Based on the eye tracking data, only a comparatively small difference between high and low complexity situations was noticeable, and the additional visual stimuli didn't seem to distract people from their main point of attention. Only approaching persons or objects tended to get a bit more attention, probably because they could cause a collision in the case of real locomotion. Apart from that, most of the test participants appeared to be mostly focused on their primary task, which in this case was to focus on the route. The impact of the other location-related factors on eye tracking results, however, was lower or different than assumed. Here, the viewing patterns and eye movement parameters recorded via eye tracking showed diverging results. This could be explained by the fact that the analyzed location-related factors may differ in

terms of importance for the perception of billboard advertising and supersede or superimpose themselves on factors of lower relevance.

Regarding the linear distance from the point of first visibility, it was assumed that billboards located close to the participants would receive higher levels of attention than far-distant ones. For the situation depicted in Figure 2 (i) and (l), respectively, this assumption was confirmed by the results (cf. Figure 3 Position (a); numeric data will be explained in greater detail in the full paper). However, the analysis of the second situation (cf. Figure 2 (j) and (m), respectively) yielded opposite results: the hit rate of 67 %, the gaze duration of 0.32 s as well as the number of fixations of 1.13 on average were higher for the far-distant billboard than the same parameters were for the closer billboard (47 %, 0.23 s and 0.97, respectively; cf. Figure 3, Position (b)). As for the influence of competition by nearby billboards, the eye tracking results in one case showed lower attentional values for a single advertising poster than for the same ad in direct competition, and diverging outcomes in the second one.

Looking at the general distribution of attention between the billboards and the surrounding environment, it can be stated that the visual attention is usually focused either on the center of the image or on the vicinity of the expected route. In those scenes mentioned above with the unexpected results, the billboards with higher attentional values were always nearer to the main focus point of attention, and those at a shorter distance or with less competition but lower attentional values were further away. One possible explanation for these results might be that the proximity to the main point of interest plays a more important role than other location-related factors such as linear distance or competition (more details will be discussed in the full paper.)

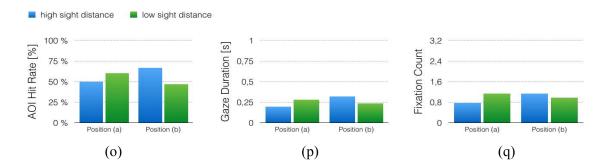


Figure 3: Eye tracking results for linear distance. (o): The hit rate (in percent) shows the number of participants that fixated the billboard advertising at least once. (p): The gaze duration (in seconds) indicates the average viewing time. (q): The fixation count states the number of fixations.

Another interesting result is that no considerable influence of the lateral distance could be detected. Due to the fact that a frontal view provides optimal visibility of the advertising poster, it was to be expected that billboards positioned directly in front of the test participants would get higher levels of attention than those at a more acute angle. Based on the eye tracking data, however, the viewing angle does not seem to have much influence on gaining and retaining attention, as the values for hit rate, time to first fixation, gaze duration and fixation count do not differ significantly. The unaided and aided recall test conducted subsequent to the eye tracking test showed that the recall performance was low in spite of the high percentage of viewers that had looked at the billboard ads according to the eye tracking results. The analysis demonstrated that both unaided and aided recall (a list of brand names was given to the test participants as *aide-mémoire*) of the advertisements shown was only successful for less than one-sixth of the test participants on average. Especially during the unaided recall, recollection was often diffuse and, in some cases, inaccurate. Test viewers, for instance, remembered only the main color of advertising posters and erroneously linked them to previous viewing experiences with (outdoor) advertising (e. g. the color red to Vodafone ads, although no such advertisement was included in the test). In the same way, many viewers thought that the advertising of

a telecommunications provider, which advertised a smartphone, belonged to a well-known technology company like Sony or Panasonic.

In contrast, the direct visual recognition of the advertising posters shown in the test was at a relatively high level. When the seven advertisement posters that were used during both tests were presented to the test participants along with three others that were not, the advertisements shown were recognized by 42 % of the participants on average. Compared to that, the number of false positives was twice comparatively low (5 %). In one case, however, an additional poster had a coloring similar to an advertising poster actually shown – and immediately, the number of false positive rose to 18 %. This is another indication of the fact that billboard advertising is indeed perceived only fragmentary and at an unconscious level. However, which and how many advertising posters were remembered differed widely amongst test participants (possibly dependent of the viewers' level of interest or their likes and dislikes regarding certain products or brands).

4. Conclusions

Although there has been an increasing research interest in outdoor advertising, it still remains an underresearched topic (cf. van Meurs and Aristoff, 2009, p. 83). Consequently, outdoor advertising is frequently criticized for a lack of verified audience measurement research (King and Tinkham, 1989, p. 47), and there is still very little known about the effectiveness of outdoor advertising (Donthu, Cherian and Bhargava, 1993, p. 70).

The impact of location-related factors on the performance or billboard advertising has for a long time been judged rather than measured (cf. Bloom, 2000, p. 406), and as a result, poster audience research systems differ widely from country to country (Bloom, 2000, p. 396). The usage of eye tracking technology to analyze the impact of billboard advertising on human attentionand perception, therefore, might be very helpful to identify those factors that influence the recall and overall effectiveness of outdoor advertising (cf. Donthu, Cherian and Bhargava, 1993, p. 70).

The results of this study seem to confirm some basic assumptions on billboard advertising, namely that it can achieve a large coverage and high levels of reach but that billboard ads are at the same time perceived only fleetingly and without conscious attention. The influence of a variety of location-related factors on the attentional impact of billboard advertisements could equally be confirmed. These factors include, for instance, the environmental complexity and the degree of occlusion of the billboard. The influence of other factors, such as the linear distance to the billboard or the number of competing nearby billboards, was however lower than expected; rather, a position near the viewers' main point of attention seems to be decisive. Thus a central billboard position could compensate for the negative impact of other location-related factors.

This might imply that common weighting procedures used to determine the influence of locationrelated factors on the effectiveness of outdoor advertising should be reconsidered. Currently (at least in the German rating system), all location-related factors are equally involved and deficiencies in one area cannot be compensated by more favorable values in another (FAW e. V, 2012, p. 5). Therefore, these results might help to better understand consumer reactions on billboard advertising and help to improve existing weighting systems that describe the impact of location-related factors.

Acknowledgments

The authors would like to thank Christian Findling and André Göhlich for their invaluable help during the preparation of the test, Klaus Stiller and Frank Roch for fruitful discussions and Nadia Nikolaus for additional advice and proof reading.

References

Anspach, S., 2004. Plakat- und Verkehrsmittelwerbung: Gestaltung, Planung und Wirkungsweise von Außenwerbung. Göttingen: Business Village.

APG | SGA, 2015. *Pricing 2015: Efficient ads – analog, digital, innovative*. [online] Available at: <http://www.apgsga.ch/media/filer_public/94/6c/946cf7a8-a2c8-4feb-8a9ed850da54e4f0/ 2015_taschenplaner_english_preise_e.pdf> [Accessed 22 January 2015].

Bloom, D., 2000. Measuring the audience to poster advertising. *International Journal of Market Research*, 42(4), pp. 395–412.

Donthu, N., Cherian, J. and Bhargava, M., 1993. Factors influencing recall of outdoor advertising. *Journal of Advertising Research*, 33(3), pp. 64–72.

Esch, F.R., 2015. *Gabler Wirtschaftslexikon: Printwerbung*. [online] Available at: <<u>http://wirtschaftslexikon.gabler.de/Archiv/81071/printwerbung-v7.html></u>[Accessed 23 January 2015].

FAW e.V., 2012. *Eine für alles: Leitwährung PpS*. [online] Available at: http://www.schwarz-aw.de/de_media/aussenwerbung/mediaforschung/pps/visuals/Broschuere_PpS.pdf [Accessed 12 December 2014].

Felser, G., 1997. Werbe- und Konsumentenpsychologie: Eine Einführung. Stuttgart: Schäffer-Poeschel.

Francese, P., 2003. More home less. American Demographics, 25(8), pp. 40-41.

King, K.W. and Tinkham, S.F., 1989. The learning and retention of outdoor advertising. *Journal of Advertising Research*, 29(6), pp. 47–51.

Lane, R., King, K. and Reichert, T., 2010. *Kleppner's Advertising Procedure* (18th Edition). Upper Saddle River, NJ: Prentice Hall.

Meffert, H., Burmann, C. and Kirchgeorg, M., 2012. *Marketing: Grundlagen marktorientierter Unternehmensführung. Konzepte – Instrumente – Praxisbeispiele* (11th Edition). Wiesbaden: Gabler.

OAAA, 2015. *Out Of Home Advertising Today*. [online] Available at: <http://www.oaaa.org/ Portals/0/Public% 20PDFs/Outdoor%20Advertising%20Today_2015.pdf> [Accessed 22 January 2015].

Pant, H., 2007. Advertising and Media. New Delhi: ABD Publishers.

Pepels, W., 2005. Marketing-Kommunikation: Werbung - Marken - Medien. Rinteln: Merkur.

Prasad, R.R., 2009. Advertising. New Delhi: Swastik Publishers & Distributors.

Prosser, M., 2013. *Should You Re-Purpose Your Newspaper Ad For A Billboard?* [online] Available at: [Accessed 22 January 2015]">http://fitsmallbusiness.com/jcdecaux/>[Accessed 22 January 2015].

Schweiger, G. and Schrattenecker, G., 2009. Werbung. Eine Einführung (7th Edition). Stuttgart: Lucius & Lucius.

Ströer, 2015. *Ströer Media Data: Campaign media – Poster. City-Light-Poster*. [online] Available at: http://www.stroeer.de/uploads/media/2015_MDB_City-Light-Poster_Englisch_01_15.pdf [Accessed 23 January 2015].

Unger, F., Durante, N.V., Gabrys, E., Koch, R. and Wailersbacher, R., 2007. *Mediaplanung: Methodische Grundlagen und praktische Anwendungen* (5th Edition). Berlin: Springer.

Van Meurs, L. and Aristoff, M., 2009. Split—Second Recognition: What Makes Outdoor Advertising Work? *Journal of Advertising Research*, 49(1), pp. 82–92.

Wilson, R.T. and Till, B.D., 2011. Effects of outdoor advertising: Does location matter? *Psychology & Marketing*, 28(9), pp. 909–933.

RocReadar - a system for collaborative transmedia publishing

Yujue Wang¹, Saunil Desai², Elena Fedorovskaya¹

¹School of Media Sciences, College of Imaging Arts and Sciences, Rochester Institute of Technology ²Department of Computer Science, College of Science, Rochester Institute of Technology

E-mail: yw1766@rit.edu; saunildesai@gmail.com; elena.fedorovskaya@rit.edu

Short Abstract

We designed and tested RocReadar - a prototype of a system for a collaborative publishing method across different media platforms, in the form of an application for smart personal Android-based devices. This system includes and uses a printed document (e.g. a magazine publication) as an entry point. The printed document is seamlessly linked to digital web-based material and collaboratively augmented by the users in an interactive fashion using personal computing devices. While the printed document stays the same, the associated virtual digital multimedia part is updated by the community of authors and readers and discovered by using image-based tags embedded in the printed document via augmented reality technology. To demonstrate the feasibility and utility of the system we asked participants to use the system while reading a printed magazine and provide their evaluation of the quality of experience. The results of the experiments can be used to formulate design recommendations for implementing transmedia publishing system in practical real-life applications and to improve our prototype.

Keywords: collaborative transmedia publishing; augmented reality, social media

1. Introduction and background

It is widely acknowledged that media communications, and the printing and publishing industry are undergoing tremendous changes, evoked by rapid digital technology developments, accumulation of knowledge and data on the web, and a ubiquitous use of personal electronic devices. During the last several years the place and role of web and digital media in publishing and communication grew exponentially, spurred by such attributes as instant access to the web-based vast data sources, peer networks and social media, availability of video and multimedia material, interactivity and real-time modifications of digital media. That growth has come at the expense of printed media. Companies like Google even ran "go paperless" campaigns. Yet, many scholars and industry leaders acknowledge specific advantages of the print medium: physicality/tangibility, portability, unique sensory qualities, its association with more effective reading comprehension, and information retaining; emphasizing therefore, its significance in the media communication realm (Davidson et al., 2010; Mangen, Walgermo, and Bronnick, 2013; Jabr, 2013). Unique affordances of different media and their distribution channels are being successfully exploited in cross-media marketing campaigns and transmedia "entertainment supersystems" (Kinder, 1991). The need for transmedia communication in journalism and news reporting has been noted by Veglis (Veglis, 2009; Veglis, 2012). It is essential to understand how to design a transmedia publishing method that capitalizes on the advantages of each media communication channel and provides superb and engaging user experience.

Recently, augmented reality (AR) technology received a significant attention as a tool to integrate digital and tangible media forms. Unlike in the early stages of AR development, when head- mounted displays were used to overlay virtual information over real world environment, the new versions of AR software for smartphone and tablets allow to easily link various digital media items with physical objects, including prints. Several companies (e.g. Metaio, Aurasma, Wikitude, Vuforia) released their implementations of AR software, and made SDKs available for custom application development. In the previous study we have used Aurasma software for the experiment, where we asked participants to

read short fictional stories produced and presented in several ways, including in electronic form, print, and paper with AR. We found that the latter method of reading, which integrates paper and electronic mediums, significantly enhances user experience and interest level as opposed to just plain reading from print (Fedorovskaya, Yu, 2014), potentially leading to better comprehension and retention of information. This method of reading was on par and even slightly better, in comparison with the interactive reading from the screen.

To explore further we decided to design a prototype of a collaborative transmedia communication system and test its feasibility and usefulness, specifically, for news and information media, by creating and evaluating a limited implementation of the method as an application for the personal smart device. Transmedia communication with collaborative features can be attractive to young audiences and may help to reduce a decline of reading, particularly with respect to news publications. As studies have shown, exposure to news correlates with higher political knowledge and civic and political engagement (e.g. Livingstone, and Markham, 2008), which, in turn, enhance psychosocial well-being (Flanagan and Bundick, 2011).

The envisioned system links three different media channels: printed publication, digital web-based information and social media with its collaborative capability of sharing and adding content.

The proposed system of a collaborative transmedia news publishing is a novel concept. It can be characterized by several key features:

- a) it is print-centric -printed information is an entry point to the process of communication;
- b) it integrates print and digital information by linking digital data through tags or triggers, making it a transmedia document;
- c) it is interactive users can navigate to digital information associated with the print via personal devices (a smartphone, tablet or a wearable augmented reality device, where a Google Glass is an example);
- d) it is collaborative users can contribute to the document creation by adding material, leaving their comments, images, etc.;
- e) it transforms the printed document into one of the channels of social media;
- f) it enables a fluent and seamless switching between print and digital; between reading and contributing.

Many of these features separately (with the exception of d) - f)) are known and have been applied in publishing and communication. The concept with all these components in combination is novel and does not yet exist. Features d), e) and f) are novel by themselves in application to publishing in a printed form.

To build the prototype with the functionality that we deemed important, we used Wikitude SDK. Existing ready-to-use AR software systems that we evaluated (Aurasma, Metaio, Layar) did not provide multiple channel media integration and user interaction experience suitable for news publishing.

We also interviewed 3 local publishers representing a newspaper, monthly life style magazine for women, and quarterly trade publication to obtain the input from the publisher side. As a result we developed a mobile Android and Google Glass prototype applications called RocReadar. These applications modify the reading experience by adding augmented reality function into normal reading process to access digital information from the printed magazine and provides a possibility to share the users own relevant content and a feedback. By taking advantages of RocReadar, readers can view pictures, videos, and listen to audio and music when they scan the magazines or books, as well as upload their own material, including photos, video and text.

In the present study we address the following research question: Does this method of publishing provide a desirable utility and an enhanced quality of reading experience? At this stage we used only one version of the system available on the smartphone.

225

We also aim to gauge the necessary workflow and usability requirements for the field trial of the application with an on-campus magazine publication to understand whether transmedia publishing can, in fact, promote news reading.

2. Related Work

Kinder (1991) extensively studied children's media and introduced the term "transmedia," to refer to the "entertainment supersystem" with examples of characters that move across media platforms and encourage their fans to follow them wherever they appeared. Jenkins (2006) extended the concept to transmedia storytelling, which refers to the systematic unfolding of elements of a story world across multiple media platforms, with each platform making a unique and original contribution to the experience as a whole. Davidson et al. (2010) elaborated on evolution and novel usage of different media in entertainment and communication denoting it as crossmedia communication. Veglis (2012) focused on the potential use of transmedia reporting in news articles.

Clearly, scholars have realized and explored potential benefits of crossmedia and transmedia storytelling and communication. They describe these methods of crossmedia communication as delivering and communicating information using various media channels, where each medium is used to engage users and deliver content that is beneficial for a particular channel or a medium. However, their work focuses mostly on the user engagement and entertainment side of communication. Moreover, none of these concepts consider a printed document as a portal for collaborative publishing, practically limiting the function of printed communication to one static channel that exists in parallel to other media, or completely eliminating printed material from the media assembly.

In contrast, we propose to conceptualize printed communication as an entry point, a portal to information that is accessed through print and is augmented and updated using connected computing devices. This approach makes the printed document a living document, and at the same time connects the reader with other readers. As mentioned earlier, these features can be particularly appealing to young audiences that are accustomed to social media. At the same time, inherently valuable characteristics of tangible print media would continue to play their role in supporting reading.

The goal of the current study is to test the feasibility of the concept by creating the prototype of the system and allowing participants to use it while reading articles in the magazine and provide the evaluation of their reading experience. We also compare reading experience with and without the system by randomly assigning participants into different groups, as well as trying to gauge their interest in using the devices.

3. Methodology

3.1 Study Design and Procedure

The study was conducted in the Discover Lab at the School of Media Sciences, Rochester Institute of Technology. Participants were seated behind a desk with a magazine placed on the desk. After introducing the study with the orientation script, which contains information about the purpose of the study and the usage of smart devices, each participant was asked to fill out a consent form and a background information questionnaire. Afterwards, the participant read the magazine, page by page, and after finishing reading the magazine, answered the main questionnaire. The questionnaire was designed to evaluate the quality of experience (Geerts, De Moor, et al, 2010) and consisted of Likert-type 7-point scales to rate both instrumental qualities: utility, efficiency, functionality, effectiveness, usefulness and ease of use; and non-instrumental qualities, such as aesthetic qualities, and motivational qualities (Mahlke et al., 2007).

Participants were pseudo-randomly assigned into two groups ensuring the equal number of both genders in each group.

1. The first group reads a selected article in the magazine without any device in a standard way. Then they are asked to read a second article, this time using a smartphone (Google Nexus smartphone) with RocReadar application, which is placed on the desk near the magazine. Participants are explained how to use the device prior to reading via a printed instruction sheet and asked to explore the system. The availability of additional digital content is designated by printed icons on the margins of the article. After each reading condition the participants fill a questionnaire.

2. In the second group, the sequence of reading conditions is switched. The participants are asked to use RocReadar on the smartphone while reading a first article. Subsequently, they read a second article without any device. Similarly to the first group, they fill a questionnaire after each reading.

In the smartphone reading condition, participants are asked to use RocReadar whenever there is additional digital content. The experimenter suggests trying out different functions of RocReadar, including sharing comments and media content using social media, to gauge the participant opinion about the usefulness and desirability of these options.

At the end of the experiment, after the reading tasks and questionnaires are completed, we interview participants for their comments, suggestions and insights.

3.2 Participants

Twelve male and twelve female RIT students in the same age group (18-26 years old) participate in the experiment. The participants are screened prior to the experiment to ensure the same level of familiarity with the augmented reality concept.

3.3 System Prototype

A prototype of the system was created as a smartphone application for an Android Nexus 5 smartphone. We utilized Wikitude free trial Android SDK (Wikitude.com) to link digital content with the printed material. To inform about the availability and to access the digital content we implemented touch icons on the user interface for video, audio, web information, and 3D animation. In addition, a comment icon and share icon were provided. The comment icon, when touched, allowed the user to upload their own multimedia content (photos or video) or type in their comments as text. It also provided access to the comments submitted by other readers. The share icon allowed the user to post and share their comments on social media sites: Google+, Facebook or Twitter. We also developed a second prototype for a wearable smart device application using Google Glass, not tested in this experiment.

The digital content was placed on the School of Media Sciences server. We used a recent issue (Spring/ Summer 2014) of the Research at RIT magazine as a printed publication and redesigned it for the experiment by choosing 3 four-page articles. We assembled relevant digital content for each page of the articles. This digital content varied across pages and included different combinations of video, audio, a 3D animation, and a supplemental web material for different pages. We randomized articles assignments for reading to control for their potential differences in interestingness.

Two versions of the redesigned magazine were printed. In the first version of the magazine we provide icons that designate the available digital content on the margins of the corresponding pages. This version is intended for the use with the digital devices. The second version does not contain any such icons and is used for the traditional reading. The printed icons have the same appearance as the user interface touch icons on the smart device applications. Figure 1 illustrates the pages of the redesigned magazine with the printed icons (Fig.1a) and a screen of the RocReadar application for the smartphone prototype (Fig. 1b).



Figure 1: (a) An open page of the redesigned Research at RIT magazine with the printed buttons on the margins designating digital content. The buttons for available content are printed in orange color (b) The Nexus 5 smartphone with the open RocReadar application. The buttons designate available digital content matching the orange buttons on the printed page. Clicking on the buttons provides easy access to the digital content.

4. Results and Discussion

The questionnaire responses were aggregated in groups pertaining to the following evaluation categories:

- 1. Reading performance;
- 2. Usability, and
- 3. Satisfaction with the current prototype;
- 4. Projected satisfaction with the ideal case including Net promoter score.
- 5. Aesthetics of the interface and design elements;
- 6. Utility and usefulness of an ideal implementation of the system (e.g. when all the shortcoming were eliminated);
- 7. Motivational quality of the system for reading news media;

Each category contained several questions (3-4), responses for which we averaged for the analysis for every participant and across participants. We treated ratings as the interval scale data, based on the findings of our previous experiment (Fedorovskaya, et. al., 2014) and available literature. We also used the participants' comments to better understand the results.

There was no significant difference between the articles in two conditions for reading performance in terms of comprehension, the effort of focusing attention, the interest level of the material. The participants could describe the content of the articles in both cases. However, in a few instances in the RocReadar condition, when asked to recall a specific detail the participants recalled the content they learned from digital media.

Overall the users were satisfied with the prototype (ratings higher than 4, which we consider as neutral) and were even more satisfied for the ideal case. Their average rating for the questions in categories 3 and 4 was statistically higher than neutral (p<0.05). For a few participants, if their response was below neutral, it was often in relationship to this particular magazine, or an Android platform. Here is a quotation of one of the participant's comments to the question whether they will re-use the system: "If it's an app for an Apple device, and if I read more RIT magazines, yes. I would use it. I think it's very cool, but personally I read novels over magazines, while this app is for magazines".

The usability ratings were rather neutral or slightly below neutral. The main concerns were associated with the speed of the application, performance stability, configuration and the display quality of additional media items. The users wanted to have more feedback to know what the system is doing, for example, whether the page scanning is completed or the video clip is going to last more than a minute, etc. If the access to the media items was not instantaneous, users commented about losing track of reading. They also pointed out that a more intimate link with the media would be desirable. For example, if there were a video complementing a specific sentence or a paragraph in the text, then underscoring a relevant word, or marking a sentence with a symbol would aid in explaining or illustrating the text. These comments suggest that image recognition- based AR may not be sufficient to support transmedia publishing and other technologies, such as optical character recognition or invisible marking may be required.

The significant results, which we were able to obtain, concern the perceived utility and motivational quality of the prototype as well as the way the printed magazine conveys availability of additional content. The majority of users rated highly the perceived usefulness of the application for learning more information to supplement and expand the printed content, for making information more interesting and easy to remember. They acknowledged that providing easy to recognize, familiar icons on the articles' margins of the printed magazine motivated them to use the system and explore content further.

The sharing feature was deemed very desirable and helpful to share interesting reads, and to engage friends by sending links, photos, notes or even captured pages. Participants suggested to implement an instant "read" feature, so users can share views and content, if they happen to read the same material in synchronicity.

The study results led us to conclude that RocReadar is perceived to be a useful tool for providing more engaging reading experiences, additional in-depth information to support learning, social sharing, and a cool factor that can help motivate news reading and reading, in general. In order to confirm whether the benefits of the system can translate beyond the lab into the real world, we plan to conduct a field study in collaboration with local Rochester publishers.

Acknowledgments

We wish to acknowledge the contribution of computer science students: Pranav Dadlani, Yogesh Jagadeesan and Mustafa Bharmal for help with improving the prototype of the applications, and Fanyi Cheng for help with interviewing participants.

References

Davidson, D. (2010). Cross-Media Communications: An Introduction to the Art of Creating Integrated Media Experiences. ETC Press.

Fedorovskaya, E.A. and Yu, L. (2014). "Investigating the Effects of Publishing Approaches Using Print, Electronic and AR Media for Storytelling on User Experience". Advances in Printing and Media Technology. Vol. XLI(I) . p. 162.

Flanagan, C., and Bundick, M. (2011). "Civic Engagement and Psychosocial Well-Being in College Students." *Liberal Education* 97 (2), 20–27.

Geerts, D., De Moor, K., Ketykó, I., et al. (2010). Linking an integrated framework with appropriate methods for measuring QOE. IEEE, QoMEX, 158-163.

Jabr, F. (2013). The Reading Brain in the Digital Age: The Science of Paper versus Screens. Scientific American, April 11, 2013.

Jenkins, H. (2006). Convergence Culture: Where Old and New Media Collide. NYU Press.

Kinder, M. (1991). Playing with Power in Movies, Television, and Video Games: From Muppet Babies to Teenage Mutant Ninja Turtles. Berkeley and Los Angeles, California: University of California Press, 38-119.

Livingstone, S. and Markham, T. (2008). "The contribution of media consumption to civic participation." *British Journal of Sociology*, 59 (2), 351-371.

Mangen, A., Walgermo, B.R., Bronnick, K. (2013). Reading linear texts on paper versus computer screen: Effects on reading comprehension. International Journal of Educational Research, 58, 61-68.

Mahlke, S., Lemke, I., and Thüring, M. (2007). The diversity of non- instrumental qualities in human-technology interaction. mmi. Interactive, 13, 55-64.

Veglis, A. (2009). Cross media communication in newspaper organizations. MCIS 2009 Proceedings. Paper 37.

Veglis, A. (2012). From Cross Media to Transmedia Reporting in Newspaper Articles. Publishing research quarterly, 28 (4), 313-324.

Library Patrons' Preferences for Social Reading of ebooks

Olli Nurmi

VTT Technical Research Centre of Finland, P.O. Box 1000, FIN-02044 VTT, Espoo, Finland olli.nurmi@vtt.fi

Short Abstract

Social reading includes a range of networked functions that are related to reading. By collaborating with other readers the satisfaction and reading experience can be improved. There is a rage of ebook reading software that supports various social reading functions like discussions, information sharing and recommendations.

The purpose of this paper is to define social reading and to study the user preferences for the different social reading dimensions in the public library context. The user feedback was gathered by focus group discussions and by web survey.

Keywords: eBooks, social innovations, social reading, public library

1. Introduction

Ebook is transforming the idea of the book and way we read books. In the new concept, rather than book as an artefact, an ebook may be seen as a mean to be connected with other people enabling a range of social activities. In addition to the traditional reading together and discussing the book social reading includes a large number of networked functions like sharing and receiving shared information.

The social reading has a longer history than solitary reading. Book historians agree that reading was originally done in groups and by reading aloud (Kilgour, 1998). Digital social services enable some of those features to re-emerge. For the shared marginalia and underlining, for example, the networked technology offers new opportunities.

Typically some of the social reading functions are integrated into the ereading software enabling the readers to interact with each other concurrent reading. However most readers use separate platforms like Facebook or Goodreads for social reading functions like sharing ratings and sharing their reading archive.

By social reading libraries and publishers are able to create a network of partnerships to provide engagement and stimulate community-based reading activities. In the world of libraries the use of social networks are widely used as a communication tool, and alongside with the library's website and its profiles on Facebook and Twitter. However, the use of social and collaborative reading is still not widespread in libraries worldwide (Nuria, 2012).

In the empirical part of this paper, readers' preferences for social reading features were tracked. The survey and focus group interviews were carried out with the patrons of the Helsinki Metropolitan Library.

Results show that the usefulness of social reading features depend on a number of factors, first of all the different types of literature and reading modes prefer different features. Shared underlining, quotations and discussions are more in place in the goal-oriented reading. In reading for leisure most relevant features are possibilities to share ratings and receive recommendations.

Secondly, the usefulness depends on the implementation of the social reading features in the ereading software. Prerequisite is that the social reading functions are integrated into the ebook reading service or application as an option so that it does not interfere with the normal linear reading. In addition,

ecosystems where networked features reside have to be so large that the benefits of crowdsourcing will materialise.

2. What is social reading?

Cordón-García and García-Figuerol (2012) are convinced that evolution of reading will eventually follow the internal logic of the network, towards connection with others and towards non-linear content. This path leads towards a book, which is more social and collaborative, more decontextualized and more heterogeneous.

Stein argues that reading and writing have always been social; the paper-medium has just covered that (2010). Stein sees an inevitable development where we will confront "many levels of reader engagement from the simple acknowledgement of the presence of others presence to a very active engagement with authors and fellow readers" – because of Internet.

In addition to the solitary time spent with a book, reading can be connected to a wide variation of social activities. For example, people loan, share and give books as a gift. People talk about what they are reading, make and receive recommendations, publish and read reviews, identify material that others might be interested in, help people explore mutual reading interests, read annotations made by others, as well as read to each other's. Reading together can be a bonding activity, which provides entertainment and education. Besides these readings or library can also act as a social symbol of an identity and a status.

In the digital world, the reading artefacts can include intentional records, such as annotations, clippings, bookmarks, notes, and other purposeful things the reader has created while she was reading. In addition, there are implicit records of reading that have been recorded by the e-reading software including logged events such as page turns, scrolls, opening and closing books, reading times, mouse clicks and so on.

Modern model sees learning more as a social process where learning happens together, by interacting, by sharing, by doing. This creates a direct demand for collaborative and network-savvy post-artefactbook, where co-creation and exporting and receiving information in the form of annotations, links and notes is common practice.

3. Categorising social reading based on reading motivation

Kamil et al. summarize the diverse depiction of engaged reader as a reader that has wants and intentions that enable reading processes to occur. That is, a person reads not only because she can to it, but because she is motivated to do it. Kamil et al. proposes that engaged readers coordinate their strategies and knowledge (cognition) within a community of literacy (social) in order to fulfil their personal goals, desires, and intentions (motivation) (Kamil et al, 2009).

Wentzel suggest that the reading motivation can be divided into intrinsic and extrinsic motivation, self-efficacy and social motivation. Intrinsic reading motivation refers to and individual's enjoyment of reading activities that is performed for their own sake. Extrinsic motivation is the desire to receive external recognition, rewards, or incentives. Bandura (1997) defined self-efficacy as "people's judgments of their capabilities to organize and execute reading. Social motivation for reading relates to reads interpersonal and community activities. Sharing books with peers and participating in community of learners leads to increased amount of reading (Wentzel, 1996).

Strommen and Mates highlights in their study that the fact that if reading is to become a lifelong habit then people must see themselves as participants in a community that views reading as a significant and enjoyable activity (2004). Indeed, the gift of reading can best be given by another reader who models what it is like to get pleasure from reading (Sheldrick-Ross, McKechnie and Rothbauer, 2005).

Based on this theory of engagement and motivation in reading the social reading tools can be grouped in the following four categories:

1. Factors that influence in intrinsic motivation and lower the barrier to read. Examples of this kind of tools are ereading software that enables to read anywhere anytime. Wigfield and

Guthrie (1997) divide the aspects of intrinsic motivation into importance, curiosity, involvement and challenge. In social reading context the importance, the belief that reading is valuable, is of relevance. Social reading can visualize that there are other readers too and the reading activity itself if of importance. Also recommendation services that allow reader to find good books to read can be included into this category.

- 2. Factors that influence in extrinsic motivation. According to Wigfield (1997), extrinsic motivation include following aspects:
 - Reading for recognition which is the pleasure in receiving a tangible form of recognition for success
 - Reading for grades which refers to the desire to be favourably evaluated
 - Competition in reading referring the desire to outperform others in reading.

Examples are tools that enable the reader to and receive comments from others that encourage her to read further.

- 3. Factors that influence in self-efficacy, for example tools that enable the reader to follow his personal achievements like achieve of reading history and statistics of reading.
- 4. Factors that influence in social motivation, for example tools enabling the reader to share his reading history and annotations allowing him to participate in social interactions and receive social recognition.

4. Methods and experimental setup

The library patrons' preferences for social reading were studied using both focus group discussions and web questionnaire as methods.

Two focus group discussions were arranged with avid readers to find out the social reading features that the Finnish library users appreciate. The events were arranged in June 2013 in two library buildings in Helsinki and Espoo. Altogether 10 persons of different age and reading habits participated into the focus group. The participants were selected to ensure that they were motivated to develop digital library services.

The discussions in the focus groups were guided by the researcher to cover following topics related to social reading: discussions, ratings and recommendations, annotations and notes, own reading archive, sharing of reading information and networking with other readers. After a brief introduction, the participants were asked to write positive and negative comments on post-it notes for each of the functions. The notes were grouped for the final discussion.

Data was also collected in the web questionnaire during autumn 2013 among the users of digital library service. Users were asked to indicate their level of agreement with several statements concerning the perceived properties of a social reading service. Altogether 210 digital library users answered the questionnaire.

5. Results

5.1 Focus group

The participants pointed out that it is crucial that the focus of the discussion in the social reading services should reside within the topic of the book and comments should deal with a particular page or chapter of the books. This rather specific requirement is understandable because the Internet is full of unfocused discussion groups covering all kinds of issues.

The focus group members saw value in discussion and they identified also a need to record the comments and come back to it. This requires a mechanism to link the discussion to the relevant page or chapter even after the book has been returned to the library. A systematic way to present one's own

comments was requested. Comments could be presented and managed for example using personal library functionality.

The focus group saw a need for a person that moderates the discussion. Less moderation may be needed if log in is required. It was regarded important that the people could participate anonymously into the discussion.

Some participants argued that the discussion could mislead the reader to pay attention to the minor issues. The main focus should always be reading the book itself not the social functions. People read books also offline and social functions must not be seen as a requirement. The quality of the discussion should be high enough. However some of the books may not initiate any discussion.

There were both negative and positive views concerning rankings and recommendations. Some people saw them useful and helping them to find interesting books to read. Some augmented that the rankings or recommendations may be too dominant or cover only too narrow area of literature disturbing or limiting the readers own choices.

Annotations and notes were considered relevant only when studying. The idea of making notes or annotations while reading a novel was considered as strange. People were not used to make annotations while reading an ebook.

People in the focus groups thought that the reading history should be private and it should be possible to manage privacy settings. Some people want to post it to everyone; others are more comfortable sharing with a smaller group, like just their friends. It is essential that the privacy settings are visible and clearly explained.

In focus groups, the concerns of privacy occurred, it is important that the status of sharing state is clearly expressed in the service. The need to have visible options and preferences in program level to indicate of what particular action is doing obvious also based implementations on existing services. User has to be sure if his/hers annotations are personal or not.

5.2 Web questionnaire

The result of the web questionnaire is shown in the figure 2. The five most appreciated social reading related functionalities were own book achieve, bookmarks, magazine and newspaper articles about the books, personal book recommendations and book ranking.

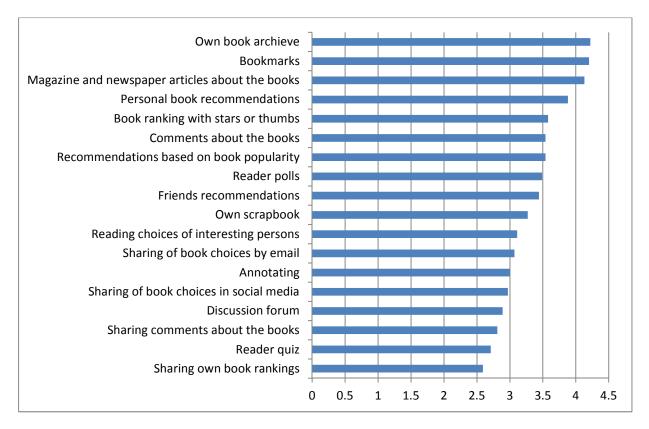


Figure 2 Result of web survey. The library patrons were asked to rank different social reading functionalities (scale 1-5).

Based on the survey most readers find it important to have a possibility to organize and to bookmark books for themselves and to receive book ratings and recommendations what to read from others. Functions that involved sharing to others were of lesser importance.

6. Conclusions

The social reading features may be needed only occasionally depending on reading purpose and reader. According to the experimental part of this study the relevant social reading features can be categorised into two groups supporting either goal oriented reading or free reading.

The first group includes features like forming new discussion or studying group and annotation features like highlighting. These features will be supported in digital learning products and services.

The features in the second group were mostly related into receiving relevant information from others that help readers to orient and get inspiration for reading itself. These features should always lay on background letting space to the readers own imagination and choices not to jeopardize the pleasure of reading.

The discussion forum was not considered the most requested feature in web polling, but yet it was considered only "somewhat important". Commenting and discussion were seen as interesting, though library patrons emphasized that discussion has to be moderated, kept to the point and to the place.

Another way to categorise the results is group them into social reading features related to the extrinsic motivation, self-efficacy and social motivation. Factors related to intrinsic motivation were not directly mentioned.

The features affecting to library patrons' extrinsic motivation included magazine and newspaper articles about the book, personal book recommendation, book ranking, comments about the book, recommendations based on book popularity, reader polls, showing reading choices of interesting persons and reader quiz. For some readers sharing of own book achieve can be a motivating factor.

Most of the features affecting to library patrons' self-efficacy improving the readers' performance were related to learning. Example of mentioned functions were own book achieve, bookmarks, own scrapbook and annotations.

The functions affecting readers' social motivation included friends recommendations, sharing book choices by email, sharing book choices in social media, discussion forum, sharing comments about the books and sharing own book rakings.

7. Discussion

The interaction that social reading services enable is based on social needs that has existed before and reformed through technology available.

Because functions of social reading are in the process of forming and the terminology is not established, some caution in detailed interpretation of these results is necessary: we cannot be sure, if participants fully understood the meaning and difference between separate functions, like discussing and commenting, between categories of annotating and between sharing and receiving shared information, even these issues were addressed in set up of the test.

Results can be interpreted so that e-reading is still generally undeveloped and possibilities in social reading are difficult to perceive without a priori knowledge.

In Finland, where the ecosystems of e-reading are weakly developed partly because the relative small linguistic area, the library could act as a unifying factor. Libraries have a large enough user base and wide publisher independent selection of Finnish literature. Social reading would suit the objectives of the library; it would allow the better discovery of literature, as well as a new kind of reading experience, which would perhaps help libraries to gain new user groups.

These findings underscore the importance of social reading to encourage children and young people to read for pleasure.

Unfortunately despite the enormous potential of the practice of social reading at present, a serious problem has to be faced: the quagmire of closed and mutually incompatible devices, platforms and formats.

Since the study HelMet libraries have continued to promote social reading together with the leading Finnish ebook electronic commerce company. The library has formed social reading club that discusses about the ebooks and reading experiences in the public libraries. Anyone interested can participate into these sessions. The social reading club has also private and public Facebook groups.

Also the digital learning environments and learning materials have evolved after 2013. Many of the social reading functions have been integrated into these services. It seems that the role of the libraries in the digital learning environments will be minor.

References

Cordón-García, J. A., Cascón, F. C., & Alonso-Arévalo, J. 2012. Libros electrónicos y contenidos digitales en la sociedad del conocimiento: mercado, servicios y derechos. Madrid: Pirámide.

Bandura, A. (1997). Self-efficacy. The exercise of control. New York, W. H. Freeman.

Heikkilä, H. Nurmi O., Laine J., Social reading in e-books and libraries, Helsinki: Next Media, [online] Available at: http://virtual/nextmediaDeliverables-2013/D1.3.7.4_eReading%20Social%20reading%20in%20e-books%20and%20Libraries.pdf [Accessed 25 June 2014].

Kamil M., Mosenthal P., Pearson D., Barr R., Handbook of Reading Research , Digital Printing 2009, New York

Marshall, C. 2009. Reading and Writing the Electronic Book. San Rafael: Morgan and Claypool.

Nuria R., 2012, Social reading and the creation of customer loyalty clubs or communities to improve communication with our users and reduce costs in marketing and advertising, The Bottom Line: Managing library finances, Vol. 25 No. 2, pp. 63-67

Kilgour, F. G. 1998. The evolution of the book. New York: Oxford University Press.

Krashen, S. (1993). The Power of Reading. Englewood, Col.: Libraries Unlimited, Inc

Laurel, B. 2013. Computers as theatre: A dramatic theory of interactive experience. Upper Saddle River, NJ: Addison-Wesley.

Sanacore, J. (1999). Encouraging children to make choices about their literacy learning. Intervention in School and Clinic, 35, 38–42.

Stein, B. 2010. A Taxonomy of Social Reading: a proposal, [online]. Available at: http://futureofthebook.org/social-reading/categories-1-2-3/index.html [Accessed 25 June 2014].

Strommen, L. and Mates, B. (2004). Learning to love reading: Interviews with older children and teens. Journal of Adolescent and Adult Literacy, 48, 188-200.

Sheldrick-Ross, C., McCechnie, L. and Rothbauer, P.M. (2005). Reading Matters: What the research reveals about reading, libraries and community. Oxford: Libraries Unlimited.

Turkle, S. 1997. Life on the Screen: identity in the age of the internet. London: Phoenix.

Wigfield, A., and Guthrie, J. T. (1997). Relations of children's motivation for reading to the amount and breadth of their reading. Journal of Educational Psychology, 89, 420-432.

Wigfield, A. (1997). Children's motivation for reading and reading engagement. In J. T. Guthrie and A. Wigfield (Eds.). Reading engagement: Motivating readers through integrated instruction. Newark, DE: International Reading Association.

Studies on the social Judgment of paper-based Communication in a special Environment

Csaba Horváth^{1*}, Erzsébet Novotny²

¹Óbuda University, Institute of Media Technology and Light Industry Engineering, Doberdó u. 6.
 H-1034 Budapest, Hungary. Tel +36 30 355840
 ²ANY Security Printing Company, Halom u. 5. H-1102 Budapest, Hungary

E-mail: horvath.csaba@rkk.uni-obuda.hu, novotny@any.hu *corresponding author

Short Abstract

At the Óbuda University, the Faculty of Light Industry and Environmental Engineering has a distinctive educational feature. They concurrently educate environmental engineers, as well as light industry engineers with specialization in printing and paper-related technologies. The researchers have examined how in this environment the opinions and attitudes of the environmental engineers studying at the faculty change in connection with paper-based communication. Conclusions concerning the marketing actions that are capable of efficiently influencing changes in the attitude of the society have also been drawn for the popularization of printed communication.

Keywords: paper-based communication, electronic communication, sustainability, Twosides

1. Introduction and background

Printed communication (the Gutenberg Galaxy) has been the key vehicle of culture and information exchange for centuries. Its social role and significance have far surpassed the economic power that it has represented. The first devices of electronic communication, the radio and television did influence this role, yet could not suppress its significance. On the other hand, the appearance of the Internet has brought revolutionary changes in the structure of printed communication and its role as an information conveyor in society. The Gutenberg Galaxy has lost its principal leading role, and become a part of the communications business just within two decades. This process has been further aggravated by the emerging and strengthening, rather unjustified indisposition against paper-based communication in the society for its erroneously alleged environmentally impairing effects. "Paperless" campaigns for the popularization of electronic communication and the strongly biased approach of environmental organizations, movements for saving rainforests tend to reinforce each other to shape adverse opinions (Macro, 2013).

The responses of the actors that are interested in the sustenance of paper-based communication encompass various measures for the introduction of sustainable and responsible forest management, environmentally friendly printing, as well as the associated popularizing campaigns. Most of them target the customers of printed products, and also have the goal of exercising positive influence on social judgment.

It was in 2011 when in representation of Hungary the Federation of Hungarian Printers and Paper Makers joined the Twosides¹ initiative that is a joint positive marketing campaign in protection and for the popularization of paper-based communication.

¹ Twosides an initiative by companies from the Graphic Communications Supply Chain including forestry, pulp, paper, inks and chemicals, pre-press, press, finishing, publishing, printing, envelopes and postal operators. Their common goal is to promote the sustainability of the Graphics Communication Supply chain and dispel common environmental misconceptions by providing users with verifiable information on why print and paper is an attractive, practical and sustainable communications medium.

To create and implement a domestic campaign, this organization has offered financial support and the use of a communications tool kit. In these activities, the Federation has relied on the "Institute of Media Technology and Light Industry Engineering" operated at the Óbuda University as the Hungarian knowledge centre of the print and paper industry. Our institute used certain tools provided by Twosides as experimental means for the provision of information to students. Our faculty, i.e. the Faculty of Light Industry and Environmental Engineering at the Óbuda University has a distinctive educational feature. We concurrently train environmental engineers, as well as light industry engineers with specialization in printing and paper-related technologies.

Another specialty is that the students belonging to various majors within the Faculty share basic subjects of science, economics and technology, and therefore they frequently meet each other in person, as a result of which they have a number of common programs, regular personal communication.

Furthermore, during their studies students of environmental engineering can become familiarized with light industry technologies, as well as paper and printing technologies in Semester 6.

This special situation has been exploited to observe whether the attitude of students of environmental engineering being particularly committed to environmental protection changes, and if yes, how during the five semesters when they are targeted by the campaign popularizing paper-based communication, but they are not in possession of technological knowledge of the topic.

2. Research methods

The students of environmental engineering who start to study paper and printing technology as a separate subject in Semester 6 are requested to complete 3 questionnaires at the beginning of the first lecture in order to unveil their opinions and knowledge of paper-based communication.

The first questionnaire focuses on their opinions and attitudes, whereas the second is based on Twosides' "Myths and Facts" publication to assess as to what extent they have adopted the false views that are widespread in public opinion. The third questionnaire reveals from what sources they have derived information that positively influences their opinions.

Since February 2010, the first questionnaire has been completed by all the students from all years, while the second and third questionnaire was introduced a year later, in 2011, and therefore the results cover this period of time.

At our faculty, we began to popularize paper-based communication in 2011, partly with the use of Twosides' campaign tools and materials, and partly by disseminating the works of light industry engineering students specialized in printing technologies.

Thus, we assessed the opinions of those students of environmental engineering (as basic information) who still not had been "infected" by the marketing effects of printed communication, and how and to what extent such a campaign could exercise its influence, and which tools proved to be effective. The following three questionnaires (Table 1–3) were compiled for the assessments. It is apparent that we worked with multiple choice tests so that the answers could be unambiguously processed.

Table 1: Opinion-requesting test

	Test questions
1	Do you like reading?
2	Do you read any book in the last month?
3	Have you ever been to a printing plant or digital printshop?
4	What do you prefer? Printed materials to read or digital data carriers?
5	Is the operation of the Internet and use of computers detrimental in any way to the
	environment?
6	Is the print technology sustainable?
7	Which of them causes larger environmental loading: electronic or graphic communication?
8	Do you have an e-book reader (or tablet)?

	Myths	Facts	True?	False?
1	Making paper always destroys forests.	Paper production supports sustainable		
		forest management.		
2	Well managed planted forests are essential	Planted forests are bad for the		
	to meet increased demand for forest goods,	environment		
3	Paper is bad for the environment	Paper is one of the few truly		
		sustainable products		
4	Most energy used renewable carbon	Making paper uses a lot of non-		
	intensity is surprisingly low.	renewable energy and has a high		
		carbon footprint.		
5	Only recycled paper should be used.	Paper made from sustainable forests is		
		needed to start the paper cycle.		
6	Paper is one of the most recycled products	Print and paper is wasteful product.		
	in the world.			
7	Electronic communication is more	Not necessarily e-media also has		
	environmentally friendly than print and	environmental impacts.		
	paper.			
8	Many consumers value paper based	Digital is now the preferred means of		
	communications	communication.		
9	Paper based packaging protects goods,	Packaging is wasteful and unnecessary.		
	reduces waste and is recyclable.			

Table 2: Myths and facts (Twosides, 2014)

Table 3: Positive influence

	What have you perceived as positive influence on your opinion in relation to paper-based communication at the university?
1	Exhibitions of students of printing technology
2	"Open Door" events of the Institute of Media Technology and Light Industry Engineering
3	Conversations with students of printing and paper engineering
4	Publications and notices given or hanged out in the building of the Faculty in connection with the paper industry and printing
5	Paper making practices in the framework of the open laboratories campaign
6	Demonstrations of printing engineering students held in the aula for the popularization of their profession
7	Influences from other sources
8	Has your opinion changed positively in relation to paper-based communication?

3. Results and discussion

The dates of the completion of the tests and questionnaires described in the previous chapter, as well as the numbers of students asked are summarized in Table 4. (The tests were completed by all the students of environmental engineering from the given Semester 6.)

	Year	Date	Number of students
1	2009/2010	February 2010	37
2	2010/2011	February 2011	31
3	2011/2012	February 2012	35
4	2012/2013	February 2013	40
5	2013/2014	February 2014	51
6	2014/2015	February 2015	42

 Table 4: Number of students of environmental engineering having completed

 the questionnaires and tests

Figure 1–3 demonstrates how responses given to the individual questions changed during the years. Percentage ratios as a specific dimension have been used to adjust the different numbers of respondents from the individual years for the purpose of the analysis.

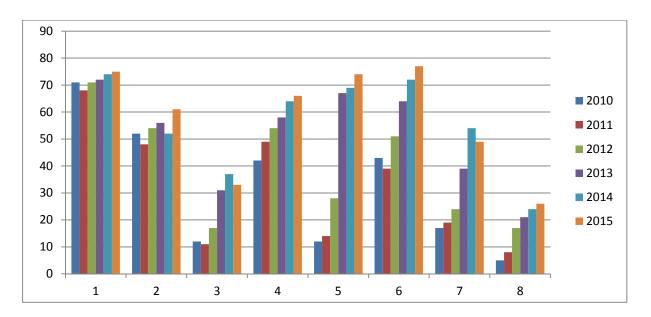


Figure 1: Percentage rate of the positive answers given to the questions, in summary of the students' opinions

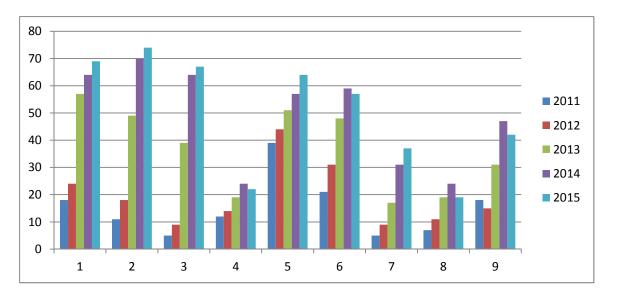


Figure 2: Myths and facts Percentage rate of correct answers concerning Print & Paper

Trends shown by the responses of students clearly reflect that the factors of positive influence presented in Table 3 have had increasing influence on the students over the years. Especially the assessment in February 2013 and thereafter suggest such changes.

Correlations are even more clearly revealed when the diagrams for the time and characteristics of the campaign that was commenced at the Faculty in 2011 - for the popularization of paper-based communication – are compared with the results collected from the answers.

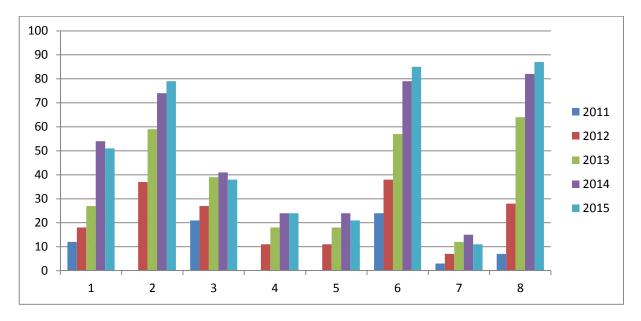


Figure 3: Percentage rate of the factors of positive influence

From among the results of positive influence – at the same time it can be taken as notable experience – it is to be highlighted that respondents were affected to the largest extent by events that demanded activeness on their part.

This experience has been exploited by the Hungary the Federation of Hungarian Printers and Paper Makers in its own campaigns organized to reach the entire society.

4. Conclusions

Summarized in this extended abstract, the results of the assessment evidence that considerable influence can be exercised on the opinions of the students of environmental engineering – who are the most committed to environmental protection – in the environmental impacts of printed communication in case they find themselves in a developing environment that well represents real facts. These results clearly show that students from the years that have not or have just shortly encountered the campaign have nurtured social prejudices. On the other hand, those who have spent more time with the campaign at the Faculty could more easily accept true facts. Finally, the views of the most recent years on printed communication have changed to a convincing extent.

References

Macro, K.: The future of print sales, Printing Industries Press, Sewickley, PA, 2013, p.101, ISBN 978-0-88362-770-9

Print and Paper Myths and Facts, Twosides, Daventry, UK, 2014, September, p. 22

42nd International Research Conference, 06 to 09 September 2015



Session 5A

Coatings

Tuesday, 8 September 2015 11:05 – 12:30

Chair: Patrick Gane

Modelling Reflectance Spectra for Special Effect Pigment Coatings

Nina Rogelj¹, Marta Klanjšek Gunde²

¹University of Eastern Finland, Institute of Photonics, Joensuu, P.O.Box 111, Finland ²National Institute of Chemistry, Ljubljana, Hajdrihova 19, Slovenia

E-mail: nina.rogelj@uef.fi; marta.k.gunde@ki.si

Short Abstract

A good calculation model for goniometric reflectance for effect coatings with interference flakes was developed and analysed here. For this purpose, reflectance spectra for coatings were applied with Iriodin 4504 from Merck, a metal lustre coating using interference mica-based pigments coated with iron (III) oxide. The numerical model used for computation incorporates three parts of the light scattering behaviour; scattering from the front surface of the coating, scattering from pigments inside the coating and scattering from the coating substrate. In order to validate the model, reflectance spectra were measured at an incident angle of 45° and at reflected angles of 60°, 30°, 20°, 0°, -20° and -65° using a commercial multi-angle spectrometer. The thickness of the interference coating applied on the flakes was adjusted to obtain good matches of interference features between modelled and measured reflectance. The influence of surface coverage by pigments and the pigment slope distribution on the resulting spectra was analysed. The first parameter represents the fraction of surface area covered with pigments and the second tells us how well the pigments inside the coatings are oriented.

Keywords: appearance, scattering, spectrum, multi-angle reflectance, effect coatings

1. Introduction and background

Surfaces that change their appearance significantly with illumination and viewing directions are becoming increasingly important in several applications, ranging from the purely decorative up to providing various functional purposes. The unique optical impressions of such surfaces give eyecatching effects, angle-dependent interference colours, pearl lustre, or multiple reflection, which characterize the appearance of the so-called gonioapparent effect. A large variety of samples falls into this class, giving angle-dependent effects due to (a) topography of the micro- and macro textures of surfaces, such as leather, textile and other microtextured surfaces, and (b) optical effects coming from metallic, interference and surface-structured flaky pigments applied in coatings, plastics and printing inks, which are mostly used in automotive, decorative and security coatings. The optical properties of such samples spread well beyond solid colour and cannot be described by any straightforward colorimetric measurement. Advanced applications demand that the appearance of objects so treated be repeatable, and so must be predictable and controllable in production to become feasible. This requires several conditions to be fulfilled; one major amongst them being the possibility to measure the appearance, predict it theoretically and to write all the details in a suitably convenient way. We have shown already that goniospectrophotometric space curves, a special representation of the corresponding bidirectional reflectance distribution function (BRDF), could serve as an appearance fingerprint of several types of gonioapparent samples (Klanjšek Gunde 2013). However, theoretical consideration of this methodology was made originally only on diffraction gratings (Rogelj 2013). This research is continued in order to provide a good model that will enable the BRDF of effect coatings to be reliably predicted, i.e. of paints or inks containing interference flakes.

BRDF contains spectral radiance coefficients for all possible illumination and viewing directions which could be measured by gonioreflectometry. Most research made so far has been for metal-effect coatings, interference coatings, and more complex effect coatings. Three different geometries are good enough for metallic coating (ASTM 2001), whereas at least five or six are required for coatings

adopting the interference effect (Takagi 2005). Most complex effect coatings require 1 485 geometries (Takagi 2007). The goniospectrophotometers currently available on the market have 6 (BYC-Mac[®], BYK Gardner), 19 (MA98[®], X-Rite) and 98 geometries (GK311/M[®], Zeiss) (Kirchner 2012).

Effect pigments can be classified into two groups, (i) pigments that consist of only one optically homogeneous material (substrate-free pigments, e.g. metallic and pearlescent flakes) and (ii) pigments that have a layered structure and consist of at least two optically different layered material (pigments with layer-substrate structure or multilayered pigments with or without a substrate, e.g. interference flakes). The class (i) pigments give rise to reflection and/or partial refraction from flakes, which in the corresponding coating make metallic or pearlescent effects, respectively. Interference flakes add the effect of interference in thin film layers, which contributes strong angle-dependent colour, which is added to the lustre and brilliance (Pfaff 1999, Maile 2005).

The objective set for this research is to make a good calculation model for BRDF of effect coatings with interference flakes, and to verify it by comparison with goniospectrometric measurements. Most attention was devoted to build the theoretical model that would enable one to vary the optical constituents of flakes (in terms of refractive index and absorption coefficient), their pigment-volume concentration, size, average inclination angle, and variance of these angle and the optical properties of the substrate. There are some attempts in the literature for such an approach (Germer 2001), which were regarded as input knowledge and were developed further to apply them for the purpose of the application in hand. The first successful and promising results are shown here together with the plans for future research.

2. Materials and Methods

2.1 Samples

Effect coatings and substrate samples were taken from the Merck test chart (Merck-Gruppe, Darmstadt, Germany). The product is one from a series of metal lustre pigments from Merck's effect pigments colour card. Interference pigments are mica-based covered with iron (III) oxide (Fe2O3) (Merck 2012). Specifications of the coating are shown in Table 1. This is a useful sample source while enough data can be provided for the flakes.

2.2 Numerical model

The numerical model describes the scattering of light impinging onto a special effect pigment coating (Fig. 1) at an angle θ from the surface normal. Reflected light is described using polar angl θ and azimuthal angle ϕ relative to the normal to the flat surface. Pigments are assumed to be tilted at an angle γ' , the angles are distributed randomly under the defined slope distribution function, i.e. a normal distribution using γ' as mean value and a variance of σ^{12} . Other angles describing the geometry are shown in Figs. 1 and 3.

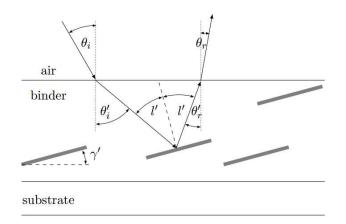


Figure 1: Simplified schematic of light scattering on special effect pigment coating.

There are three parts contributing to the scattering that are incorporated in the model:

- scattering from front surface of the coating facet model
- scattering from special effect pigments inside the coating flake model
- scattering from substrate of the coating base model

The Stokes vector for scattering from the whole sample is, thus, described as:

$$S = (F^{facet} + F^{flake}C + F^{base})S_0$$
^[1]

where S_0 is the Stokes vector of the incident light, F^{model} is the Mueller matrix containing bidirectional reflections for the specific model and the *C* parameter is the fraction of the surface area which is covered by pigments, going from 0 (no pigments) to 1 (pigments covering the whole sample area). The facet and flake models are treated in similar fashion and described in detail in [Germer 2001, Germer 2014]. Here we describe the base model in the subsection 2.1.1, whereby the facet model treats the scattering from the front surface as specular reflection from aligned facets which have their slopes distributed according to a slope distribution function. The flake model treats pigments as aligned facets and also takes into account the refraction into and out of the contacting, surrounding binder.

2.2.1 Base model

The base model incorporates scattering from the substrate. According to this model, light can take 4 different paths on its way through the coating to the substrate and back. The paths, along with their respective angle notations are schematically represented in Figures 2-3, and expressed as:

- 1. transmission through pigments, reflection from substrate, transmission returning through pigments
- 2. transmission through binder (no crossing of pigments), reflection from substrate, transmission returning through pigments
- 3. transmission through pigments, reflection from substrate, transmission returning through binder (no crossing of pigments)
- 4. transmission through binder (no crossing of pigments), reflection from substrate, transmission returning through binder (no crossing of pigments)

The first option will always occur to some extent, being the only possible path in the case of complete coverage, whereas the other three can occur only if surface coverage is less than 1.

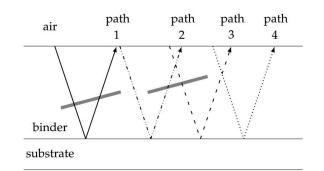


Figure 2: Schematic representing the 4 different paths the light can follow when undergoing scattering from the substrate. For simplicity only two pigment particles are drawn.

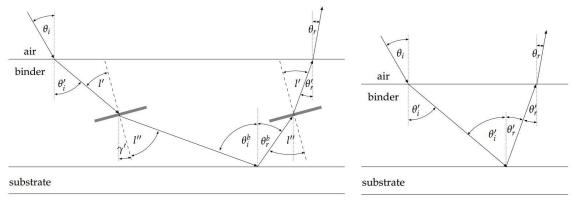


Figure 3: Schematic of base model – path 1 (left) and path 4 (right).

2.3 Numerical modelling of the samples

A numerical model is used to compute the pathway interactions for the special effect pigment coating Iriodin 4504 (Fig. 4). Values in italic font were adjusted accordingly to match the calculated spectra with the measured values as best as possible.

Table 1: Specifications of the analysed interference effect pigment coating - data for binder, substrate and flakes. γ' is the flake's tilt angle and σ'^2 is the variance for the slope distribution of flakes. Mica (Muscovite $KAl_2(Si_3Al)O_{10}(OH,F)_2$) is here represented with the parameters of silicate (SiO₂).

binder	material	acrylic pitch	
binder	thickness $[\mu m]$	600	
substrate	material	white paper	
substrate	thickness $[\mu m]$	100	
	materials	Fe_2O_3 +mica(SiO_2)+ Fe_2O_3	
special	thicknesses [µm]	0.076+0.125+0.076	
effect	mean diameter [µm]	5-50	
pigments	С	1	
(flakes)	γ´ [°]	0	
	$\sigma^{\prime 2}$	0.08	

Sector Constants			

Figure 4: Appearance of Iriodin 4504 under aspecular angles 60°, 30°, 20°, 0°, -30° and -65° (from left to right).

3. Results and Discussion

The comparison between calculated reflectance and the observed goniospectrophotometric values is now summarised.

3.1 Measured vs. modelled reflectance

The coating was measured with the commercial multi-angle spectrophotometer X-Rite MA98[®] in the wavelength range 400-700 nm with an interval step of 10 nm. Incident angle was set to 45°, and only in-plane reflection was measured with aspecular angles 60° , 30° , 20° , 0° , -30° and -65° . The same angles were used when modelling the multi-angle reflectance of the coating. All calculations were made for the 440-700 nm spectral region. The reason for choosing a narrow region is related to the optical data available, i.e. the published values for refractive index and absorption coefficient, which are limited to this region, and no extrapolation was used.

Fig. 5 left shows a comparison between measured and modelled reflectance, where computational parameters are those from Table 1. Fig. 5 right shows CIE a^*b^* colour coordinates at aspecular angles for measured reflectance values. Measured spectra have a slight increase at 440 nm, a valley in region between 500 and 550 nm, a peak around 630 nm and a decrease at 700 nm. Modelled values exhibit the same features. The position of peaks and valleys is greatly determined by the pigment layer thickness. Since thickness data were not available for Iriodin 4504, the pigment layer thickness was adjusted in each case to minimize the CIE76 colour-difference formula. Table 2 shows CIE76 colour difference and Root Mean Squared Error (RMSE) between colours determined from measured and modelled reflectance spectra. The obtained results show an acceptable match.

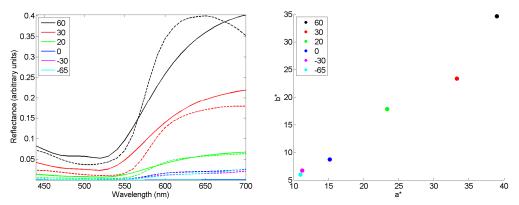


Figure 5: Left figure shows measured (dashed line) and modelled (solid line) reflectance values. Computational parameters are those from Table 1. Right figure shows CIELAB a*b* colour values for the measured reflectance spectra (see also Figure 4 and Table 2). Aspecular angles are specified in the corresponding legend.

aspecular angle	CIE76	RMSE
60°	11.72	0.0409
30°	9.2	0.0238
20°	7.22	0.0048
0°	17.96	0.0134
-30°	13.89	0.0107
-65°	13.17	0.0119

Table 2: CIE76 colour difference and RMSE between measured and modelled reflectance values.

3.2 Influence of surface coverage

Changing volume concentration of pigments or their diameter means changing surface coverage, C in equation (1). Smaller surface coverage means that less of the area is covered with pigments and the substrate has a greater influence on overall reflectance. To show this influence, we have taken different values of surface coverage and computed the reflectance values.

We changed the surface coverage from 0.1, 0.5 to 0.9 (Fig. 6). We notice that smaller surface coverage results in a bigger influence of the substrate, as expected. Reflectance curves for all angles converge toward one value, but their relative separation stays the same. The shape of the reflectance also changes with surface coverage. For decreasing coverage, the reflectance curves become more similar to that for the refractive index of the coating substrate alone.

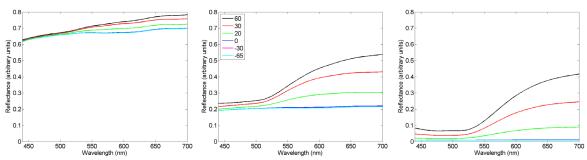


Figure 6: Changing surface coverage from 0.1 (left), 0.5 (middle) to 0.9 (right). Legend in middle graph is applicable to all three graphs. For easier comparison, all graphs have the same scale.

3.3 Influence of slope distribution of flakes

The slope distribution of flakes in the model is assumed to be a normal distribution with its mean value and variance. Both parameters influence the results, but we will only take a look at the influence of the variance and assume that the mean value is 0°, i.e. the mean value is parallel to the plane of the substrate. Fig. 7 shows Iriodin 4504 using variance values of 0.04 and 0.16. The same figure also shows the normal distribution for variances 0.02, 0.08 and 0.3. From the graphs we see that smaller variance results in significant reflection only at angles close to specular reflection, 30° and 60°. Other angles have relatively low reflectance values. When the variance gets bigger, reflectance over a wider range of angles becomes significant. A small variance means that the reflection is highly directional, in the specular direction. Large variance means that the pigments are randomly oriented in the coating and its appearance becomes more matt, meaning that a wider range of reflection directions reflect similarly.

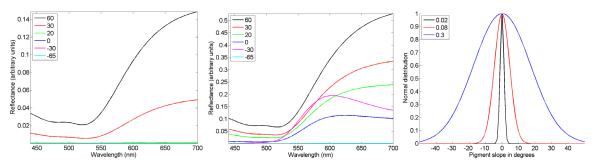


Figure 7: Iriodin 4504 using variance 0.04 (left) and 0.16 (middle). Legend in left graph is applicable to both left and middle graph. Normal distribution using three different variances is shown in right graph.

To have a better picture of how the variance affects different angles, Fig. 8 shows the relative reflectance at 650 nm for all angles in respect to reflectance at angle 60° . Relative reflectance is plotted against variance going from 0.02 to 0.3. The angles closer to the specular reflection have a quicker rise and then slowly converge to a constant value. The angles furthest away from specular reflection, -30° and -65° , start their rise rather late, but then rise more quickly than the other angles. This behaviour is in accordance with the assumption applied of a normal distribution; until variance becomes large enough to cover all angles, the ones farthest away from specular reflection have very low probability and consequently very low reflectance.

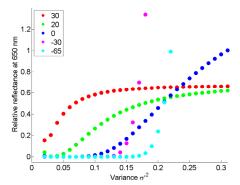


Figure 8: Relative reflectance (reflectance at 650 nm for specific angle divided by reflectance at angle 60°) versus variance.

4. Conclusions

The focus of this research was on modelling reflectance spectra of a selected special effect coating with interference pigments, having an Fe_2O_3 layer on a mica core. The modelled reflectance was validated with measurements of the selected sample at the same goniometric angles that were used in the model. The influence of two important parameters on goniometric reflectance was analysed, namely surface coverage and pigment orientation slope distribution. Surface coverage is directly related to diameter and volume concentration of pigments for a given pigment thickness, i.e. aspect ratio, and both can be varied in the ink or paint manufacturing process. Depending on this factor, the coating substrate will affect the reflectance and become incorporated in the appearance of the coating. The pigment orientation slope distribution is an important factor when applying the ink or paint. Different application processes can distribute pigments with different orientation variance, and, as we show, this parameter can have great influence on the reflectance.

The preliminary results of our research confirm that the model provides useful results and it will be developed further. Other types of flakes are currently being considered, e.g. metallic, pearlescent and interference with more layers. Other interesting substrates will be introduced, such as black and

coloured paper, and metallic sheet. Furthermore, the pigment orientation slope will be varied to obtain goniometric reflectance of effect coatings with well oriented flakes. The entire data will be used to check the possibility for appearance fingerprinting and to answer how many measurement geometries are required to describe it in sufficient detail.

The entire research has an ambition to provide a generalized view on the need for high spatial resolution of goniometric measurements for effect coatings. While all currently known special effects will be taken into account, the model will provide the reflection spectrum for arbitrary illumination-viewing directions for any combinations of ink/paint preparation and application. Such data are important for several purposes, being for decorative, security or any other purpose.

Acknowledgments

The authors acknowledge X-Rite and their country representative, Roman Habicht, for the opportunity to apply the software of the MA98 multi-angle spectrophotometer, and to Hana Erznožnik and Frenk Šemen at Helios d.d., Slovenia, for measurements on their apparatus.

References

ASTM E2195.01., 2001, Standard practice for specifying the geometry of multiangle spectrophotometers, Tech. Rep. ASTM E2194.01

Germer, T.A. and Nadal, M.E., 2001, Modeling the appearance of special effect pigment coatings SPIE Proceedings Vol. 4447, pp. 77-86

Germer, T.A., Zwinkels, J.C. and Tsai, B.K., 2014, Accurate Measurement of Optical Properties of Materials (Experimental Methods in the Physical Sciences), Spectrophotometry, Volume 46.

Kirchner, E., Cramer, W., 2012, Making sense of measurement geometries for multi-angle spectrophotometers, *Color Res. Appl.* 37, pp. 186–198

Klanjšek Gunde M., Rogelj, N., 2013, Suitability of goniospectrophotometric space curves as appearance fingerprints, *Appl. Opt.* 52, pp. 2718–2728

Maile, F. J., Pfaff, G., and Reynders, P., 2005, Effect pigments-past, present and future, *Prog. Org. Coat.*, 54, pp. 150–163

Merck, 2012, Safety data sheet, Iriodin 4504 Lava Red

Pfaff, G. and Reynders, P., 1999, Angle-dependent optical effects deriving from submicron structures of films and pigments, *Chem. Rev.*, 99, pp. 1963–1982

Rogelj, N., Poberaj I., and Klanjšek Gunde, M., 2013, Goniospectrophotometric space curves of diffraction gratings and their applicability as appearance fingerprints, *Appl. Opt.*, 52, pp. 8355-8362

Takagi, A., Watanabe, A., Baba, G., 2005, Prediction of spectral reflectance factor distribution of automotive paint finishes, *Color Res. Appl.*, pp. 30, 275–282

Takagi, A., Watanabe, A., Baba, G., 2007, Prediction of spectral reflectance factor distribution of color-shift paint finishes, *Color Res. Appl.*, 32, pp. 378–387

Eveliina Jutila¹, Risto Koivunen¹, Roger Bollström², Patrick Gane^{1,2}

¹Aalto University, School of Chemical Technology, Department of Forest Products Technology, 00076 Aalto, Helsinki, Finland ²Omya International AG, Baslerstrasse 42, CH-4665 Oftringen, Switzerland

E-mail: eveliina.jutila@aalto.fi; risto.koivunen@aalto.fi; roger.bollstroem@omya.com; patrick.gane@omya.com

Corresponding author: Eveliina Jutila, Tel.: +358 50 3841753, Fax: +358 9 47124259.

Short Abstract

The applicability of highly porous functionalised calcium carbonate (FCC) as ab and adsorbent in thin layer chromatography (TLC) is investigated to provide functionality in printed microfluidics. Five different FCCs were prepared to illustrate effects of surface area and charge distribution. In its natural state, FCC displays anionicity within the particle pores and cationicity on the particle extremities (Lamminmäki et al., 2012). Two polymeric agents, cationic high molecular weight polyDADMAC and anionic sodium polyacrylate dispersant, were additionally used to control overall surface charge of the FCC particles. A silica gel TLC plate was used as reference. Three colorants, Sudan red, soluble in ethanol, and Gardenia blue and Amaranth red, each soluble in water, were chosen as optically traceable analytes. Ethanol and deionised water were subsequently applied, respectively, as eluents. The choice of the latter two dyes was specifically made to differentiate between an insoluble and soluble analyte in respect to mixture separation in the two wicking solvents. Eluent wicking time was fast within the higher surface area FCC coatings, and relatively low amounts of analyte could be used due to the high separation resolution capabilities of FCC. Sudan red with ethanol eluent in FCC coatings displayed a higher in-solvent retention factor, $R_{\rm f}$, than on the reference TLC plate, indicating that the dye shows no physisorption to FCC. Amaranth red and Gardenia blue remained strongly fixed against ethanol eluent. With deionised water as eluent, the Sudan red remained fixed due to its insolubility, whereas the Amaranth red and Gardenia blue, being strongly polar, either became progressively adsorbed on FCC, undergoing charge attraction to cationised surface, or became fixed after a short distance, suggesting coagulation and subsequent size exclusion. By converting the surface to anionic charge, the anionic Amaranth red flowed freely with both the ethanol and aqueous solvents. The control properties support future microfluidic applications.

Keywords: thin layer chromatography, functionalised calcium carbonate, specific surface area

1. Introduction and background

Thin layer chromatography (TLC) is an inexpensive, simple and versatile chromatographic method, which can be used to separate and identify compounds in a solvent-borne mixture. TLC has a wide range of applications including pharmaceutical analysis (Attimarad *et al.*, 2011; Ferenczi-Fodor *et al.*, 2006), drug analysis (Pyka, 2014), food and agricultural analyses (Sherma, 2000), and environmental analysis (Agarwal and Behari, 2007). TLC plates comprise of a supporting substrate and a thin absorbent layer. Plastic, glass or metal are typically used as substrate material and the typical coating choices include silica gel, cellulose, polyamide and aluminium oxide. The plates are prepared by marking the origin position for initial analyte placement, and the final position to be reached by the solvent(s) wicking front line. The sample is dissolved and applied as a spot onto the TLC plate using a pipette or a micropipette and allowed to dry. A chosen solvent is poured into a chromatographic chamber with a lid and the solvent is allowed to saturate the chamber before the plate is placed inside it. The solvent, acting as eluent, wicks through the absorbing coating layer by capillary action and the

components of the sample migrate at different rates and thus separate from each other. The plate is removed from the chamber when the solvent front has nearly reached the top of the plate and the plate is allowed to dry. The samples are visualized usually by the naked eye or by examining the sample under UV light if a UV tracer is used (Wall, 2005).

The TLC separation process can be evaluated by calculating the retention factor, $R_{\rm f}$, for each compound using Equation 1:

$$R_{\rm f} = \frac{a}{b}$$
 [1]

where a is the distance travelled by the compound and b the distance travelled by the solvent. The retention factor is dependent upon the type of absorbent, thickness of the absorbent, solvent, amount of analyte and the environmental conditions including temperature and humidity (Ferenczi-Fodor *et al.*, 2006; Heftman, 1983).

The TLC technique has been previously applied to precipitated calcium carbonate (PCC) by Donigian *et al.* (1999), and to modified industrial quality calcium carbonates, which were used in the study by Lamminmäki *et al.* (2011). The objective of this study was to clarify whether coatings consisting of functionalized calcium carbonate (FCC), formed similarly by a selected acid dissolution of the carbonate and reprecipitation, could be used as thin absorbent layers in TLC. This study focuses on comparing FCC coatings with different properties such as specific surface area, porosity and surface charge with commonly used silica gel coated TLC plates using simple colorant and solvent systems at first. Here the coatings were made completely anionic or cationic by adding the dispersant directly into the coating mixture. In future work anionic and cationic areas are printed onto the FCC coated samples using an inkjet printer. Further studies will also include examination of different binders, colorants and analytes, with a view to establishing their suitability for microfluidic diagnostics. For example, Printed anionic and cationic areas in microfluidic channels could be used in separation of compounds or to control the flow of components in microfluidic channels.

2. Materials and Methods

2.1 Coating Materials and Formulations

Table 1 shows the formulations and properties of the coatings which were used as thin absorbent layers in the TLC experiments. The chosen coating pigments are highly porous forms of functionalised calcium carbonate (FCC), a refined form of surface modified carbonate, with specific surface areas of 40 m²g⁻¹, 105 m²g⁻¹ and 160 m²g⁻¹ (BET, ISO 9277), respectively, provided by Omya International AG, Oftringen, Switzerland. Figure 1 shows scanning electron microscope (SEM) images from two FCC pigment coatings having specific surface area of 40 m²g⁻¹ (left) and 105 m²g⁻¹ (right), respectively.

The intrinisic charge distribution on the FCC particles displays two charge regions, cationic on the extremities of the particle surface structure and anionic within the particle pores. Two different water soluble polymeric agents, a cationic high molecular weight polyDADMAC (Cat) (Sigma-Aldrich, product code 409030) and an anionic sodium polyacrylate dispersant (An) (GXN, Coatex S.A., France) were used to modify the average surface charge of the FCC particles. The coatings were made in this first trial series without binder.

Coating	Specific surface area / m²g⁻¹	Polymer/Dispersant	Solids content /%	Thickness /µm
FCC(40)	40	-	27.1	51.8 ± 3.8
FCC(105)	105	-	14.7	70.7 ± 3.8
FCC(160)	160	-	18.4	70.6 ± 2.5
FCC-An	105	An (5 pph)	15.5	53.0 ± 8.5
FCC-Cat	105	Cat (5 pph)	14.5	63.9 ± 0.8

Table 1: Coating recipes for substrate coatings

Microscope glass slides (25×75 mm², VWR International BVBA, Leuven, Belgium) were coated with the various test formulations using a K202 Control Coater (RK PrintCoat Instruments Ltd., Herts, UK) adopting the blue labelled wire-wound rod, which applies a 100 μ m thick wet film onto the substrate, with a speed setting of 6 m.min⁻¹. Silica gel 60 F₂₅₄ coated aluminium TLC plates (50×75 mm², Merck Millipore) were used as a reference substrate. The TLC plates have a pore diameter of 6 nm, pore volume (N₂-isotherm) of 0.74 – 0.84 cm³g⁻¹, specific surface area (BET) of 480 – 540 m²g⁻¹ and coating thickness of 165 – 235 μ m.

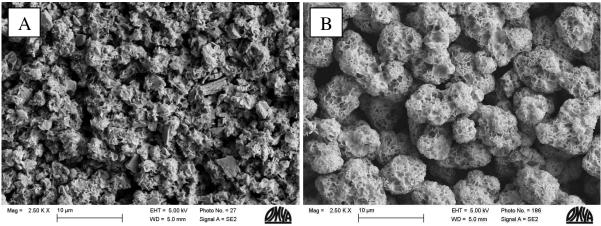


Figure 1: SEM images of FCC coated samples: a) FCC(40), and b) FCC(105).

2.1 Colorants and solvents

The colorants used for the TLC experiments were Sudan Red G colorant (Sigma-Aldrich, product code 17373), water soluble Gardenia Blue powder (Omya Hamburg GmbH, product number OP0154) and water soluble Amaranth red (Fluka, product code 06409). Additionally, water insoluble Sudan red was used for comparison. Sudan red is an industrial azo-dye, which is used to dye multiple materials including plastic, fats, oils, polystyrene and cellulose (Hayenga, 2011). Gardenia blue is a natural dye, which is exploited in coloring food, cosmetics and fabrics (Oda, 2011). Amaranth red is a synthetic anionic azo dye (Gong *et al.*, 2005; Minioti *et al.*, 2007), which is utilised in dyeing foods, drinks, drugs and cosmetics (Mpountoukas, P. *et al.*, 2010). The Sudan red was dissolved into denatured ethanol (0.1 %) (Industol PE 2, Altia Oyj, Rajamäki, Finland), and Gardenia blue and Amaranth red were dissolved in deionised water (0.1 %). Other colorants and commonly used analytes will be tested in the future.

A simple test, in which Amaranth red and Gardenia blue were dissolved into ethanol (0.1 %), revealed that Amaranth is somewhat soluble in ethanol whereas Gardenia blue is not. In this initial study, it was therefore possible to investigate the various chromatographic responses using denatured ethanol and deionised water in turn to study the transport properties of each dye through the FCC structures during wicking. Other solvents will be explored in future tests.

2.3 Characterization methods

The solids content of the coating slurries was determined with a Precisa HA 300 Moisture Balance (Precisa Gravimetrics AG, Dietikon, Switzerland). Thickness of the coated samples (Table 1) was measured using a Dektak 6M stylus profilometer (Veeco, Plainview, NY, USA), by moving the coated sample beneath a diamond-tipped stylus that moves along the surface of the sample. A tip with a radius of 12.5 μ m was used at a scan speed of 500 μ ms⁻¹ applying 1.00 mg equivalent force to the coated samples. The background gradient was levelled using the machine software and the average step height values for each sample were obtained using the interfaced software.

SEM micrographs were obtained with a Sigma VP field emission scanning electron microscope (Carl Zeiss AG, Germany), stereo light microscopy imaging was carried out with a Leica MZ16A (Leica Microsystems Ltd., Heerbrugg, Switzerland) and surface profiling made with a confocal laser scanning microscope, Zeiss LSM 5 Pascal (Carl Zeiss Micro Imaging GmbH, Göttingen, Germany).

The properties of FCC coatings, such as particle size distribution of the pigments, viscosity of the coating, zeta potential and pH of the coatings as well as coating and particle pore volume were determined and shall be presented in future work.

2.3 Thin layer chromatography (TLC)

Thin layer chromatography (TLC) experiments were performed under standard laboratory conditions: 23.0 °C \pm 1.0 °C and 50 % \pm 2.0 % RH (Jokio and Hiertner, 1999). Due to the fragility of the coated samples, only small markings were made on the sides of the slides 1 cm from top and bottom (Figure 1 a)). In the first test series three 0.5 μ l spots of Sudan red in ethanol (0.1 %), Gardenia blue in water (0.1 %) and a mixture of these colorants were applied to the bottom line of the samples (Figure 2a)). Amaranth red in water (0.1 %), Gardenia blue in water (0.1 %) and a mixture of test series. The samples were allowed to dry for 72 hours. Glass jars with a plastic petri dish as a lid were used as chromatographic chambers. The glass jars were filled with 15 cm³ of either denaturated ethanol or deionised water, which corresponds to approximately 3 – 4 mm initial solvent front height, to act as the wicking solvent. Each solvent, was allowed to saturate the chambers for 15 minutes, after which the samples were placed into the chamber, as can be seen in Figure 2b). The samples were positioned on top of a light table during wicking until the solvent eluent line reached the top 1 cm mark.

The solvent front wicking time was recorded, the samples were photographed with a digital camera (Sony Cyber-shot DSC-HX20V, Sony Europe Limited, Espoo, Finland) and viewed under a dual wavelength UV lamp (254 nm and 366 nm) (CAMAG, Muttenz, Switzerland). The retention factors, R_f , for each colorant on each sample were calculated using Equation 1. Figure 2c) shows a schematic of determining the retention factor from a developed TLC sample.

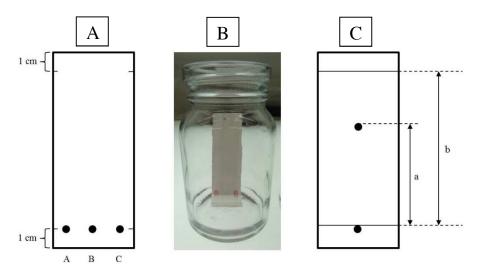


Figure 2: TLC experiments: a) preparation of coated slides, b) TLC test set-up and c) determining the retention factor R_{f_i}

3. Results and Discussion

The two eluent solvents, ethanol and water, respectively, are now reported individually.

3.1 Denatured ethanol as eluent

The results for the ethanol solvent front wicking time from approximately 3 - 4 mm from the bottom of each sample to the 1 cm upper limit, and the retention factors, are presented in Table 3. The fastest wicking time was provided by the reference TLC plates. Of the three different FCC pigments, similar wicking times were observed for FCC(105) and FCC(160). FCC(40) has a significantly lower surface area than the other two pigments, partially contributing to the slower wicking. The higher the surface area, the smaller are the pigment particles (Gane, 2001). Therefore, the coating structures have different pore structures linked with surface area, which, for constant surface chemistry, primarily affect the wicking speed (Koivula *et al.*, 2012). FCC(40) has less wicking surface compared to FCC(105) and FCC(106), which in turn differ in respect to pore size and connectivity. This will be confirmed in later published work.

Table 3: Ethanol solvent front wicking time and retention factors (R_f) *of TLC tests.*

			R _f	
Sample	Solvent front wicking /min	Sudan red in ethanol (0.1 %)	Gardenia blue in water (0.1 %)	Mixture (Sudan red/Gardenia blue)
FCC(40)	120	0.83	0.00	0.83/0.00
FCC(105)	34	0.95	0.00	0.96/0.00
FCC(160)	33	0.91	0.00	0.93/0.00
FCC-An	43	0.95	0.00	0.95/0.00
FCC-Cat	34	0.96	0.00	0.95/0.00
TLC (silica gel)	20	0.79	0.00	0.80/0.00
Sample	Solvent front wicking /min	Amaranth red in water (0.1 %)	Gardenia blue in water (0.1 %)	Mixture (Amaranth red/Gardenia blue)
FCC(40)	127	0.00	0.00	0.00/0.00
FCC(105)	27	0.00	0.00	0.00/0.00
FCC(160)	25	0.00	0.00	0.00/0.00
FCC-An	37	0.16	0.00	0.15/0.00
FCC-Cat	30	0.00	0.00	0.00/0.00
TLC (silica gel)	19	0.64	0.00	0.63/0.00

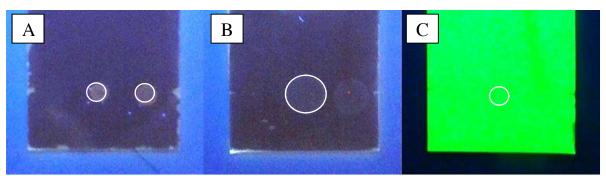


Figure 3: Images of the samples under UV light (254 nm) showing Gardenia blue fixation a) FCC(160), b) FCC-An, showing some spreading on initial drop placement, and c) TLC (silica gel with fluorescence indicator). The spot positions are shown artificially by the white rings.

The retention factors in Table 3 show that Sudan red moves along the coated structures, but that neither Gardenia blue nor Amaranth travel with the ethanol solvent through the coatings, except in the case of the Amaranth red in the anionic coating (Figure 3). Amaranth red, being an anionic dye is not adsorbed onto the surface of the anionic coating because of charge repulsion, but instead wicks with the solvent. On a cationic coating, the dye becomes fixed to the surface, because of electrostatic interactions (Lamminmäki *et al.*, 2011). Examination of samples under UV light (254 nm) confirmed that the Amaranth red and Gardenia blue remained fixed in their original spots (Figure 2).

Figure 4 shows the examples of this same ethanol solvent test series under ambient light, in which once again drops of Sudan red in ethanol, Gardenia blue in deionised water and a mixture of these were applied onto the samples. Figure 5, in turn, shows the second test series with Amaranth red, Gardenia blue both applied in aqueous solution and a mixture of the two. With ethanol as the transport solvent, Gardenia blue is barely visible due to its poor light fastness (Oda, 2011), but is visible on the samples under UV light (Figure 3). Sudan red, fully soluble in ethanol, was the only colorant which travelled through the coatings, except for Amaranth red, weakly soluble in ethanol, on the anionic coating.

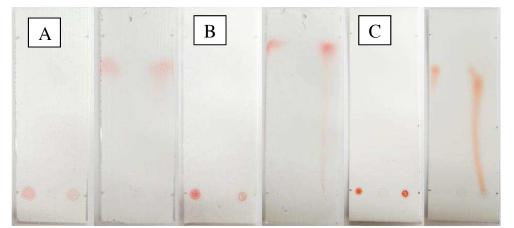


Figure 4: TLC experiments with Sudan red and Gardenia blue: the sample before the experiment (left) and the developed sample (right): a) FCC(40), b) FCC-An and c) TLC (silica gel).

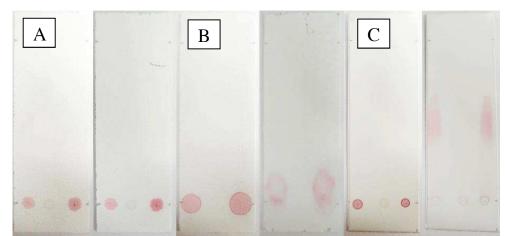


Figure 5: TLC experiments with Amaranth red and Gardenia blue: the sample before the experiment (left) and the developed sample (right) a) FCC(40), b) FCC-An and c) TLC (silica gel).

3.2 Deionised water as eluent

The results for the solvent front wicking time and the retention factor with water as eluent are presented in Table 4. Sudan red remains fixed due to its insolubility. Gardenia blue is mobile on the TLC plates, but barely retainable in the eluent, suggesting surface adsorption and/or ionic coagulation and size exclusion - to be reported in later work. Amaranth red, in contrast, travels with the water through the coatings, as can be seen in Figure 6. The wicking of Amaranth red is greatly influenced by the surface area of the coating, as well as the charge interaction between the dye and the coating. On the anionic coating, for example, the Amaranth dye flows freely and travels to the sides of the slides where a coffee ring effect takes place as the solvent evaporates, whereas on the cationised coating the Amaranth red is rapidly adsorbed. The influence of excess cationic polymer on coagulation will also be reported in later work. The pore structure and charge of the coatings will be determined to explain the differences in Amaranth dye travel between FCC(105) and FCC(160).

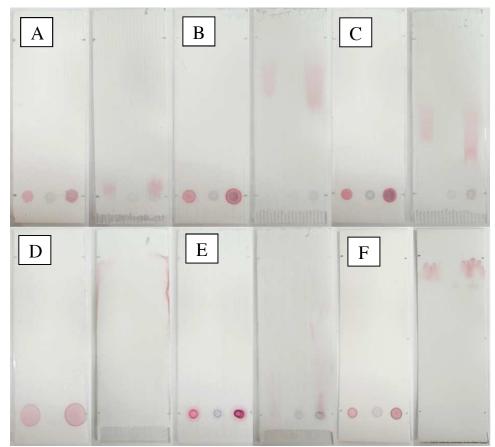


Figure 6: TLC experiments with Amaranth red and Gardenia blue with deionised water as solvent: the sample before the experiment (left) and the developed sample (right): a) FCC(40), b) FCC(105), c) FCC(160) d) FCC-An, e) FCC-Cat and f) TLC (silica gel).

Wicking was much faster with deionised water than with ethanol, because water has a much higher surface tension than ethanol. The fastest wicking time of the water solvent was provided by FCC(105) and FCC(160). FCC(40) showed much slower solvent and dye wicking and further experiments are required to determine the cause.

			$R_{\rm f}$	
Sample	Solvent front wicking /min	Sudan red in ethanol (0.1 %)	Gardenia blue in water (0.1 %)	Mixture (Sudan red/Gardenia blue)
FCC(40)	47	0.00	0.00	0.02/0.02
FCC(105)	10	0.01	0.14	0.02/0.07
FCC(160)	10	0.05	0.14	0.00/0.06
FCC-An	13	0.00	_*	0.00/0.00
FCC-Cat	21	0.00	0.06	0.01/0.04
TLC (silica gel)	77	0.01	0.73	0.00/0.73
Sample	Solvent front wicking /min	Amaranth red in water (0.1 %)	Gardenia blue in water (0.1 %)	Mixture (Amaranth red/Gardenia blue)
FCC(40)	52	0.07	0.01	0.07/0.01
FCC(105)	11	0.80	0.13	0.80/0.11
FCC(160)	10	0.55	0.08	0.51/0.06
FCC-An	13	0.93	_*	0.92/-*
FCC-Cat	21	0.24	0.06	0.53/0.04
TLC (silica gel)	75	0.93	0.80	0.95/0.79

Table 4: Deionised water solvent front wicking time and retention factors (R_f) *.*

^{*}Gardenia blue wicked with the solvent and became invisible due to dilution and background brightness.

4. Conclusions

The preliminary results of tests where three optically traceable analytes and two eluents were used show that FCC coatings could potentially be used as a chromatographic medium. The tests demonstrate that surface area and surface charge of the coatings can be used to control the separation of compounds and eluent wicking time, with no evidence of physisorption in the cases studied.

High surface area coatings provide fast eluent wicking and high separation potential. Furthermore, the eluent wicking time of deionised water was much faster on FCC coatings than that of ethanol, because of the highly ionic surface nature of the coatings. These properties are ideally suited for the separation of analytes in microfluidic devices, with particular application in printed diagnostics. The ability to control local charge by the potential of printing gives high resolution on these coated surfaces. Cellulose based filter paper is used currently in such applications, but FCC coatings provide better resolution and enable the use of lower sample and reagent volumes (Jutila *et al.*, 2015). An additional benefit, therefore, is that relatively small amounts of analyte can be used due to the high separation resolution capabilities of FCC.

The properties of FCC coatings, such as the zeta potential and porosimetry, as well as the introduction of binders, will be further elucidated, once again with ultimate focus on pharmacy and microfluidic diagnostic applications.

Acknowledgments

Funding for the project was solely supported by Omya International AG. Silvan Fischer (Omya International AG) is acknowledged for the SEM analysis and Janne Halme (Aalto University School of Science, Department of Applied Physics) for assisting with profilometry.

References

Agarwal, R. and Behari, J.R., 2007. Screening for mercury in aqueous environmental samples and urine samples using thin layer chromatography. *Water Environment Research*, 79(12), pp. 2457-2463.

Attimarad, M., Muaan Ahmed, K.K., Aldhubaib, B.E. and Harsha, S., 2011. High-performance thin layer chromatography: A powerful analytical technique in pharmaceutical drug discovery. *Pharmaceutical Methods*, 2(2), pp. 71-75.

Donigian, D.W., Wernett, P.C., McFadden, M.G. and McKay, J.J., 1999 Ink jet dye fixation and coating pigments. *Tappi Journal* 82(8), pp- 175–182.

Ferenczi-Fodor, K., Végh, Z. and Renger, B., 2006. Thin-layer chromatography in testing the purity of pharmaceuticals. *Trends in Analytical Chemistry*, 25(8), pp. 778-789.

Gane, P.A.C., 2001. Mineral pigments for paper: Structure, function and development potential. *Wochenblatt für Papierfabrikation*, 129(3), pp. 110-116.

Gong, R., Ding, Y., Li, M., Yang, C., Liu, H. and Sun, Y., 2005. Utilization of powdered peanut hull as biosorbent for removal of anionic dyes from aqueous solution. *Dyes and Pigments* 64(3), pp. 187-192.

Hayenka, I., 2011 Sudan red dye standards. AnalytiX 8(4).

Heftman, E., 1983. *Chromatography: fundamentals and applications of chromatographic and electrophoretic methods*. Part A: Fundamentals and techniques. New York: Elsevier Scientific Publishing Company.

Jokio, O. and Hiertner, M., 1999. *Reliability of results in physical testing of pulp and paper*. Pulp and Paper Testing. Papermaking Science and Technology, Book 17. Second Edition. Ed. Levlin, J-E. & Söderhjelm, L.; Fapet Oy, Helsinki, pp. 256-275.

Jutila, E., Koivunen, R., Gane, P.A.C.,2015. Effect of coating pigment, binder type and binder amount on planar liquid wicking on coated substrates. Revised and resubmitted to *Journal of Print and Media Technology Research*.

Koivula, H., Toivakka, M. and Gane, P., 2012. Short time spreading and wetting of offset printing liquids on model calcium carbonate coating structures. *Journal of Colloid and Interface Science*, 369(1), pp. 426-434.

Lamminmäki, T.T., Kettle, J.P., Puukko, P.J.T. and Gane, P.A.C., 2011. The chromatographic separation of anionic dye in inkjet coating structures. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 377(1-3), pp. 304-311.

Lamminmäki, T., Kettle, J.P., Gane, P.A.C., 2012. The electrostatic and hydrophobic interactions with inkjet ink colorant during adsorption and their effect on the print quality of inkjet printed coating layers", *8th International Paper and Coating Chemistry Symposium*, Stockholm, pp. 81-84.

Minioti, K.S., Sakellariou, C.F. and Thomaidis, N.S., 2007. Determination of 13 synthetic food colorants in water-soluble foods by reversed-phase high-performance liquid chromatography coupled with diode-array detector. *Analytica Chimica Acta* 583(1), pp. 103-110.

Mpountoukas, P., Pantazaki, A., Kostareli, E. Christodoulou, P., Kareli, D., Poliliou, S., Mourelatos, C., Lambropoulou, V. and Lialiaris, T., 2010. Cytogenetic evaluation and DNA interaction studies of the food colorants amaranth, erythrosine and tartrazine. *Food and Chemical Toxicology* 48(10), pp. 2934-2944.

Oda, H., 2011. Improving light fastness of natural dye: photostabilisation of gardenia blue. *Colorant Technology* 128(1), pp. 68-73.

Pyka, A., 2014. Detection Progress of Selected Drugs in TLC. *BioMed Research International*, 2014:732078, 19p.

Sherma, J., 2000. Thin-layer chromatography in food and agricultural analysis. *Journal of Chromatography A*, 880(1-2), pp. 129-147.

Wall, P.E., 2005. *Thin-layer Chromatography: A Modern Practical Approach*. RSC Chromatography Monographs. Cambridge: The Royal Society of Chemistry.

Yellowing of Thick-Film Coatings printed with UV-Curable Inkjet Varnishes

Manfred Schär¹, Tjaša Krivec², Gorazd Golob², Karl-Heinz Selbmann¹

¹ Institute of Print Technology, Bern University of Applied Sciences, Berne, Switzerland ² Faculty of Natural Sciences and Engineering, University of Ljubljana, Ljubljana, Slovenia

E-mail: manfred.schaer@bfh.ch

Short Abstract

Photo induced yellowing of UV cured clear varnish begins increasing immediately after UV light exposure. The effect grows proportionally with the intensity of radiation exposure and the concentration of photo initiators in the varnish. Thicker layers of varnish, as applied for spot or structure varnish, are affected disproportionately. Although initial photo yellowing is partly reversible, quality control and color matching become difficult under such conditions. Varnish layers of up to 250-500 μ m are needed to produce readable Braille letters. In the context of the production of a small series of Braille books for blind and visually impaired children, several UV inkjet varnishes were investigated by means of artificial aging, with the goal of improving quality and readability. The measurements confirmed the hypothesis of a two-step process of initial and long-term yellowing. The causal factors contributing to yellowing are also discussed.

Keywords: yellowing, UV ink, UV inkjet print, artificial aging, tactile images

1 Introduction and Background

UV curing chemistry has been used in the printing industry for many years, and as a result of recent investment in inkjet print head and fluid formulation R&D, ink-jetting is now an established and robust deposition tool for such UV curable fluids (Hudd, 2010).

Besides conventional multi-color UV printing, clear UV varnish is becoming more important in so called "up-value" production steps such as the application of eye catching localized varnish on packaging. In this case the varnishes optical effect, intensifying the brilliance and gloss of the underlying printed media, is the goal. A layer thickness of a few μ m is sufficient for this finishing process. To provide tactile impressions of a substrates structure, varnish is also used. The creation of distinct haptic effects similar to embossed texture requires layer thickness in the range of 20 to 200 μ m.

One very specific application of structured varnish is the printing of Braille letters for visually impaired readers. This idea has even been extended into the creative sphere, with raised imaging used for illustrations in Braille books as well as museum and gallery signage and descriptive labels (Eccles, 2014). The objects and artefacts exhibited in museums for people with visual impairments are typically made available either through directly touching the original or a 3D reproduction. Paintings, on the other hand, require a different approach. In this case a tactile adaption is typically created. A process for the creation of such a tactile adaption of the painting is described in Krivec et al., 2014. Therein, they describe using a Roland UV LEC-330 inkjet printer to reproduce the haptic effect of the painting "*Portrait of Empress Elisabeth of Habsburg*" in a multi-step, layered printing process.

The tactile elements in books for the visually impaired people should typically have $250-500 \,\mu\text{m}$ of relief texture to ensure good tactile recognition. Usually, embossing machines are used to punch Braille letters into white paperboard. Recently, having seen the proven feasibility of printing raised images with UV cured inkjet processes, several manufacturers, such as Roland, Mimaki and Scodix,

have integrated the capability to process such raised textures into their industrial inkjet printers and Raster Image Processors.

Since UV ink can be cured instantly after printing, it is possible to print multiple layers onto the same spot to raise the image if single pass printing cannot achieve the required height.

UV curing ink is known for yellowing, which is due to initial photo yellowing and subsequent aging. Also, a layer with an ink film thickness in the range of sub-microns to microns will show considerably less color change when compared to a layer with an ink film thickness of 500 μ m. This may be attributable to the Beer-Lambert law (Studer et al., 2001).

In the framework of a thesis project, a small series publication for blind children was produced using typical industrial printing equipment. Textual and graphical elements in the book were produced using digital printing equipment. Following the application of color, tactile and braille elements were overprinted onto the pages using ink jet technology. Potential inks for the tactile elements were limited to materials which presented no health risks to the reader. The printed tactile elements were transparent, such that the visual effect of the book for sighted individuals was not compromised; the tactile elements had to preserve the color and clarity of the base printed material, such that any yellowing or color shifting could not be tolerated. Additionally, the effect of dark or light background colors upon any yellowing that might occur was explored.

Hence, the target of this work was to study the yellowing of UV curable inkjet inks due to curing with UV light and artificial aging in sun-like conditions. Further, it was concluded which ink was suitable for printing raised images up to 500 μ m with minimal yellowing. Finally, an explanation for the physical and chemical process behind yellowing was investigated.

2 Materials and Methods

2.1 Safety issues of UV curing inks

UV cured ink can be hazardous to human health. Special attention was paid to the ink's suitability for use in printing tactile images, especially since the printed product will be handled by children. European Union directives and safety measures pursuant to toys were applied where applicable.

Standard	Description
Directive 2009/48/EC	Toys are defined as "products designed or intended, whether or not exclusively, for use in play by children under 14 years of
	age."
EN 712: 2011 Safety of toys – Part 2: Flammability;	Spread of flame and flame speed on paperboard
EN 719, 10 and 11: Organic Chemical Compounds;	With varnish layers higher than 500 μ m tests, as described in this standard, should be made.
The Chemicals in Toys; A General Methodology for	Special considerations for products which are in close dermal
assessment of chemical safety toys with focus elements report 320003001/2008.	contact.

Table 1: EU Directives and standards for toy safety

2.2 Measurement of color change (yellowing) after artificial aging

The color difference ΔE_{ab}^* is calculated according to the equation [1]:

$$\Delta E_{ab}^* = \sqrt{\left(L_b^* - L_a^*\right)^2 + \left(a_b^* - a_a^*\right)^2 + \left(b_b^* - b_a^*\right)^2}$$
[1]

Where $(L_b^* - L_a^*)$; $(a_b^* - a_a^*)$; $(b_b^* - b_a^*)$ are the differences of the measured $L^*a^*b^*$ values of the aged coating _b and the original coating _a.

2.3 Paperboard

In Table 2 the paperboards used for the accelerated aging tests are shown.

Properties	Iggesund Invercote Creato	Storaenso Ensocoat	Standards
Weight (g/m2)	260	250	ISO 536
Thickness (µm)	290	300	ISO 534
Color L*	96.5	97.7	ISO 5631-2
a*	2.3	2.5	ISO 5631-2
b*	-7.8	-7.1	ISO 5631-2
Whiteness (%)	127	125	ISO 11475
ISO brightness (%)	94	95	ISO 2470
Customization	none	one color offset printing	
		(black) on printing side	

Table 2: Paperboard used

2.4 Ink formulations

The dominant chemistry for both UV and EB curing is the radical polymerization of unsaturated monomers and oligomers. Monomers and oligomers containing unsaturated acrylate are the most commonly utilized in UV and EB initiated radical polymerization due to their higher reactivity compared to methacrylate, allyl or unsaturated vinyl (Arceneaux et al., 2012).

Table 3: UV cur	able inkiet va	rnishes used to	coat the pa	per samples
10010 01 01 011	ciore initiger rei	involves used to	cour nic pu	ber beimpreb

SCODIX varnish		ROLAND varnish		TEST varnish		FLINT varnish		
Scodix PolySense 100	Scodix PolySense 100		Roland EUV-GL		Test Overprint Varnish		Flint Overprint Varnish	
							•	
Ink composition	% by	Ink composition	% by	Ink composition	% by	Ink composition	% by	
	weight		weight		weight		weight	
Acrylic ester	30-40	Acrylated amine	10-20	Dipropylenglycol	25 - 50	No information		
		synergist		diacrylate		retrievable		
Trimethylbenzoyldip	1-2	hexamethylene	30-40	2,2-	10 - 2,5			
henylphosphine oxide		diacrylate hexane-		bis(acryloyloxym				
		1,6-diol diacrylate		ethyl)butyl				
		-		acrylate				
Acrylic acid ester	10-20	Acrylic esters	30-40	-				
1,6-	1-2	Other photo	10-20					
Hexanedioldiacrylate		sensitive						
		monomers						
		Phosphine oxide	5-15]				
		derivative						
		Others	0-1	1				

2.5 Printing of the samples, accelerated aging process and evaluation of the color change

The coating was applied manually with a profiled rod in one layer onto the paperboard sample and immediately UV cured after this application. Next, the initial color measurement of the sample was made. The samples were then put together into the accelerated aging chamber and, at scheduled intervals, the color measurement was retaken. Finally, the coating thickness was measured with digital micrometers. In Table 4 the process steps are described. In Table 5 the measurement equipment is shown. It must be mentioned that in order to protect the spectrophotometer from ink residues there

were no color measurements made on the wet coatings. Consequently, the color change due to the curing process was not evaluated.

Nr.	Process step	Parameter / unit		Devices used
1	Coating application	Ink film thickness in μ m		VisCone Instruments Profiled rods 24, 80, 140, 220 μ m
2	UV curing	Curing conditions: 100 % UV power, belt speed 15 m/min		Hönle UVAPRINT 300 HPL ACM G 6.0 kW Fe doped Hg-lamp
3	Initial Color measurement (a)	Measuring conditions: ABS/D65/2/DIN/POL		x-rite SpectroEye spectrophotometer
4	Accelerated aging		70 W/m2 @ 340 nm, C ℃	Q-SUN 3100 HS Xenon test chamber
5	Interval Colour measurement (b)	Measuring conditions: ABS/D65/2/DIN/POL		x-rite SpectroEye spectrophotometer
6	Coating Thickness measurement	coating thickness in mm		Digital micrometer and Digital indicator on a gage stand

Table 4: Process steps to apply the ink and to measure the colour difference of the accelerated aging

Table 5: Measuring devices to measure the colour difference and the coating thickness of the varnish

Nr.	Device	Accuracy	Calibration
1	x-rite SpectroEye spectrophotometer 3.264 - 30106	Inter instrument agreement: maximally $0.8 \Delta E^*_{CIElab}$ (D50, 2°) on 12 BCRA tiles Short term repeatability: $0.02 \Delta E^*_{CIElab}$ (D50, 2°), mean value of 10 measurements every 10 seconds on white	White reference: ceramic tile traceable to NIST CS-10
2	Digital Micrometer Sylvac S_Mike Pro 0-30mm, Typ 903.0300.10, Serial Nr 810002	±0.001 mm	Calibrated with a 1 mm gauge block
3	Digital indicator Mitutoyo ID- C112B, Code 543-250B, Serial No 01866, 12.7 0.001 mm, mounted on a gage stand	±0.001 mm	Calibrated with a 1 mm gauge block

2.6 Theory on yellowing: understanding the physical and chemical processes

The yellowing effect can be divided into two stages. Yellowing begins with radiation exposure during the curing process. This yellowing is partially reversed during the first hours after curing, and is followed by a much slower yellowing during the aging period. The extent to which the yellowing caused by curing disappears during the critical period after curing depends upon the storage temperature. Storage atmosphere (Air vs. N_2) has less of an impact on the samples (Schwalm, 2006). Measurements showed (Studer, 2001) that yellowing increases with higher concentration of initiators up to a certain limit after which it begins to slowly decline. One other yellowing factor could be the binder, or bulk material. Studer and Königer mixed identical photo initiators with different binders and

reached different degrees of yellowing under otherwise identical conditions. This shows that both photo initiators and the varnish matrix affect yellowing.

Furthermore, investigations of e-Beam cured (without photo initiator) and UV-Light cured (with photo initiator) resins show that e-beam cured resins starts with higher color values than by UV resins without photo initiators bleeding faster and more completely.

It seems the radical concentration has an intrinsic effect on the first stage. (Studer, 2001) Although the yellowing is a function of the e-beam dose respectively the photo initiator concentration. For low irradiation doses the yellowing increases, however at very high doses it correlates reversely.

3. Results and Discussion

Figure 1 shows the printed and aged test samples after 6 hours (top) and 180 hours (bottom) of accelerated aging. The substrates used were black Ensocoat and white Invercote paperboard. Samples of both substrate types with no varnish and samples coated with ROLAND varnish, TEST varnish, FLINT varnish and SKODIX varnish were prepared. The FLINT varnish and the SCODIX varnish samples show rather severe warping due to the shrinking of the thick ink film during the curing process. Each sample was measured at three locations with the spectrophotometer. The spectrophotometer crosshairs and the sample sequence numbers indicate the measurement locations on the sample surface.

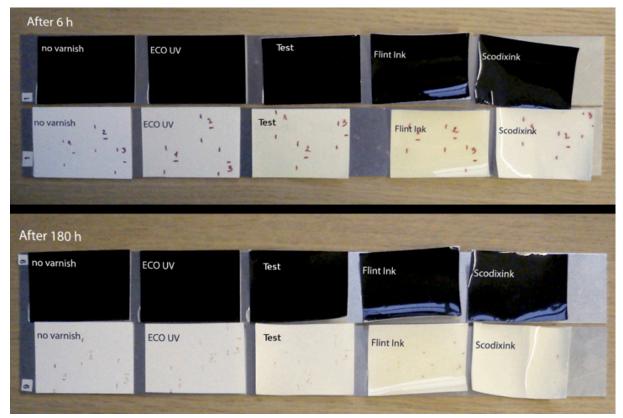


Figure 1: Samples of black and white paperboard, one without coating and four coatings with different inks after 6 hours and 180 hours of accelerated aging.

Figure 2 shows the measuring results for the coating thickness of the varnishes on **white** paper board. The biggest error is the uniformity of the coatings caused by the coating process steps itself, firstly from unreproducible handling of the profiled rod and secondly from the shrinkage and warping of the varnish during the curing process. Measurement uncertainties caused by the measuring devices (digital micrometer Sylvac S_Mike, digital indicator Mitutoyo ID-C112B) is ± 0.001 mm, compression of the paperboard due to the measuring force is ± 0.010 mm and sample warping is ± 0.010 mm.

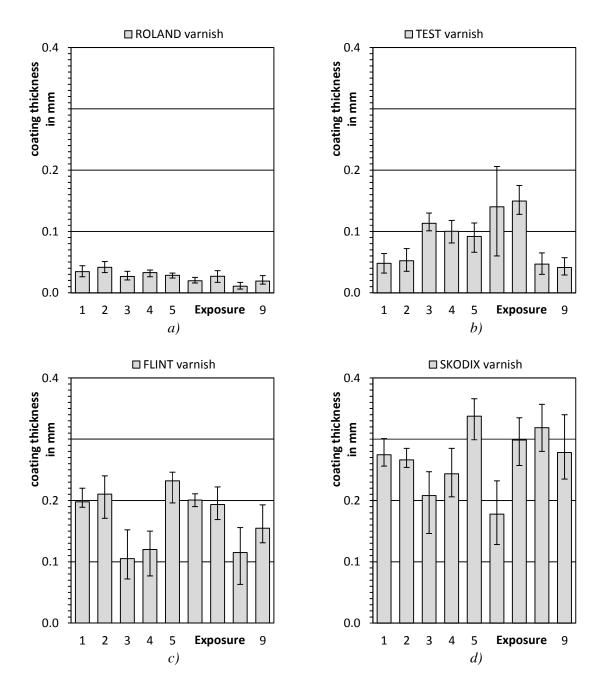


Figure 2: Graphs a) to d) plot the measuring results for the coating thickness of the varnishes on white paper board. Bars plot the mean value of seven measurements on each exposure sample. Three measurements are identically located with the spectrophotometer as illustrated in Figure 1. Error bars show minimal and maximal values of seven measurements. Bigger widths of error bars indicate more uneven coatings.

Graph	Varnish	Thickness mean value	Profiled rod	
<i>a</i>)	ROLAND varnish	0.027 mm	24 µm	
<i>b</i>)	TEST varnish	0.087 mm	80 µ m	
<i>c</i>)	FLINT varnish	0.170 mm	140 µm	
<i>d</i>)	SKODIX varnish	0.267 mm	220 µm	

Table 6: Mean values of the coating thicknesses

Figure 3 shows the spectrophotometer measuring results of varnish on white paperboard. Measurement uncertainties of the spectrophotometer are $0.02 \Delta E^*_{ab}$ (D50, 2°) for short term repeatability and maximally 0.8 ΔE^*_{ab} (D50, 2°) for inter instrument agreement. However, the warped samples prevented proper placement of the spectrophotometer which resulted in partly poor reading of the color values.

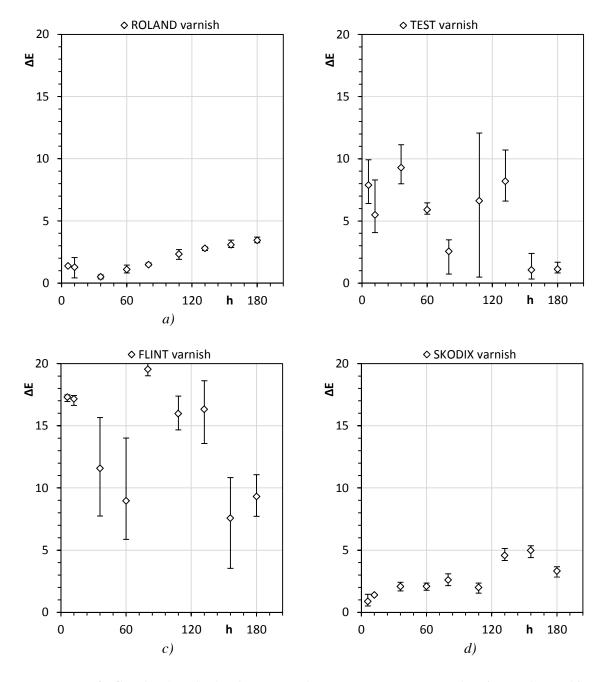


Figure 3: Graphs a) to d) plot the spectrophotometer measuring results of varnish on white paperboard. Error bars show minimal and maximal values of three measurements on each exposure sample. Widths of error bars are bigger on warped samples, a) coating with ROLAND varnish and reasonable results, b) coating with TEST varnish resulted in poor readings due to warped samples, c) coatings with FLINT varnish resulted in poor readings due to warped samples, d) coatings with SKODIX varnish and reasonable results.

The colour difference ΔE^{*ab} was correlated with the coating thickness *d* to study how the colour would changes over time due to accelerated ageing. Calculations were done according to equation [2] and results are shown in Figure 4.

$$\frac{\Delta E_{ab}^*}{d} \qquad [2]$$

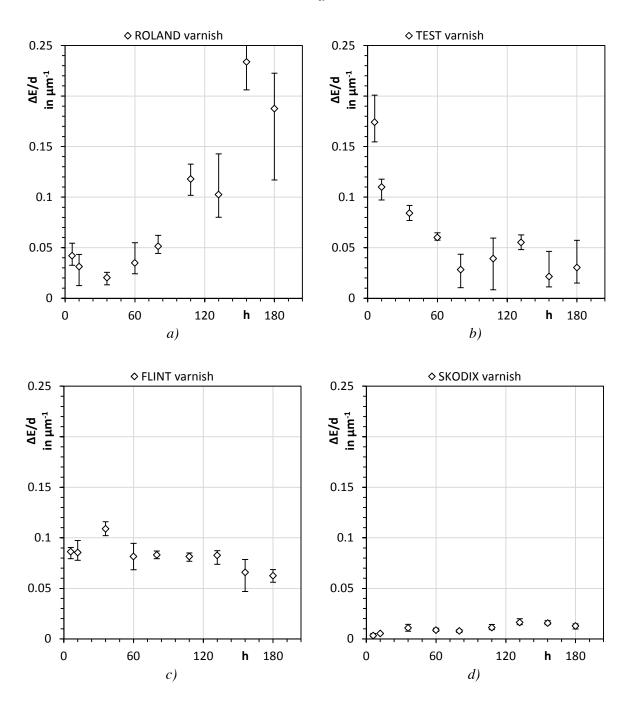


Figure 4: Graphs a) to d) plot the color difference correlating with coating thickness on white paperboard. a) ROLAND varnish bleached out in the first 36 h and increased yellowing afterwards, b) TEST varnish bleached out in the first 60 h and constant afterwards, c) FLINT varnish with strong yellowing at first then bleaching out slightly, d) SKODIX varnish with low yellowing at first then slightly increasing yellowing.

In Figure 5 the accelerated aging color changes of the uncoated white and black paperboard are plotted. The ΔE^*ab is less than one unit in both cases, indicating that there was almost no visible color change caused by the aging process.

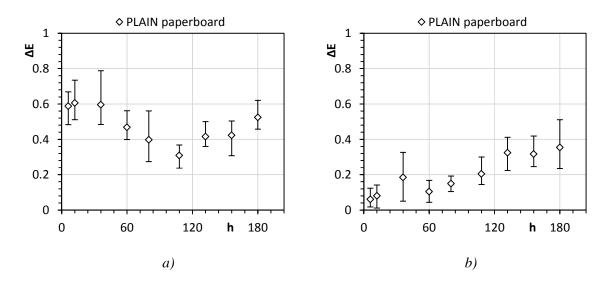


Figure 5: Graphs a) to b) plot the spectrophotometer measuring results of plain paperboard. Error bars show minimal and maximal value of three measurements on each exposure sample.
a) White paperboard without varnish. b) Black paperboard without varnish.

Figures 6 shows the color changes of each varnish type during aging on black paperboard. The ΔE^*_{ab} measurements indicate that there was almost no visible color change.

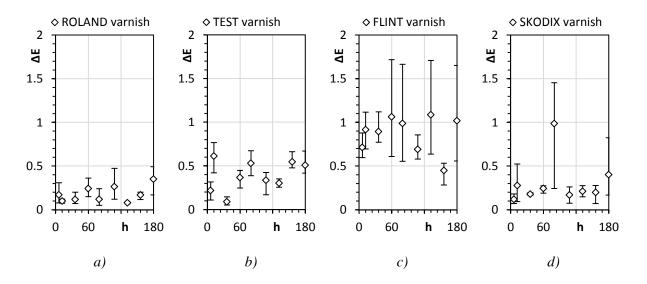


Figure 6: Graphs a) to d) plot the spectrophotometer measuring results of varnish on **black** paperboard. Error bars show minimal and maximal value of three measurements on each exposure sample. Widths of the error bars are much bigger on warped sample. The results show that even thick coatings on black paperboard will not produce a noticeable difference except for FLINT varnish with a just noticeable difference, a) coating with ROLAND varnish, b) coating with TEST varnish, c) coatings with FLINT varnish, d) coatings with SKODIX varnish.

4. Conclusion

The work shows that thick clear coatings with UV curable inkjet varnish undergo a significant yellowing during accelerated aging processes. Some varnishes demonstrate increased yellowing with age while others indicate signs of bleaching during aging. At the outset of the project, it was hypothesized that thick varnish layers printed onto a black background would cause noticeable yellowing effects. Ultimately, tests showed that this hypothesis was incorrect; while the yellowing is clearly visible on white substrates, it is not noticeable on black substrates.

For the small series publication for blind children, as mentioned in the introduction, ultimately, the SKODIX varnish and a dark gray background was chosen for the majority of the tactile elements in the book. This ensured that the yellowing would be unnoticeable without giving the book an overly dark appearance to sighted readers.

It is also hypothesized that the hygroscopic nature of cellulose causes applied UV varnishes to be partially absorbed into substrate materials. These fibers are then able to interfere with the activation of the photo initiators, leading to incomplete crosslinking of the monomer material. This may also factor into yellowing processes, and is an area for ongoing study.

References

Arceneaux, J. and Willard, K, 2012. UV&EB Chemistry and Technology. RatTech Printer's Guide. [Online] Available from: http://radtech.org/index.php?option=com_content&view=article&id=196&Itemid=141 [Accessed 14th February 2015].

Directive 2009/48/EC of the European parliament and of the council of 18 June 2009 on the safety on toys. [Online] Available from: http://eurlex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2009:170:0001:0037:en:PDF

Eccles, S., 2015. How to print 3D with a UV digital inkjet. [Online] Available from: http://www.fespa.com/ news/features/how-to-print-3d-with-a-uv-digital-inkjet.html [Accessed 13th February 2015].

Hudd, A., 2010. Inkjet Printing Technologies. In: Magdassi, S., eds. *The chemistry of Inkjet Inks*, World Scientific Publishing, pp. 11.

Ink ingredients according to the material safety data sheets of Scodix PolySense 100, Roland EUV-GL, Schmid Rhyner Overprint Varnish, Flint Overprint Varnish

Krivec, T., Muck, T., Fugger Germadnik, R., Majnarić, I. and Golob, G., 2014. Adapting Artworks for People Who Are Blind or Visually Impaired Using Raised Printing. *Journal of Visual Impairment & Blindness, January-February 2014*, pp. 68-76

Schwalm, R., 2006. UV Coatings Basics, Recent Developments and New Applications; Elsevier Science (December 21, 2006)

Seng, H.-P., 2000. Coatings 5, 199

Studer, K. and Königer, R., 2001. Initial photoyellowing of photocrosslinked coatings. *European Coatings Journal*, 2001(1-2), pp. 26-33

Effect of Coating Structure and Cationic Charge Level on Inkjet Printability

Frank Gref^{1,2}, Päivi Miettinen¹, Jaana Heinonen¹, Mika Sundquist³, Martti Toivakka²

¹CH-Polymers Oy, Raisio, Finland ²Laboratory of Paper Coating and Converting and Center for Functional Materials, Åbo Akademi, Turku, Finland

³ UPM R&D, Lappeenranta, Finland

E-mail: frank.gref@ch-polymers.com; paivi.miettinen@ch-polymers.com; jaana.heinonen@ch-polymers.com; mika.sundquist@upm.com; martti.toivakka@abo.fi

Short Abstract

In order to achieve acceptable high speed inkjet runnability with low smearing tendency, a paper with high absorptivity is needed. This is normally achieved with highly porous and absorbent silica pigments and hydrophilic additives such as polyvinyl alcohol. In addition to absorptivity, cationic charges on paper surface are advantageous for fixing the ink on paper.

In this study, the effect of coating colour cationic character and porosity of the coating layer on inkjet printability was investigated. Based on a laboratory scale pre-study, coating colours for pilot scale coating trials were selected considering the rheological stability and inkjet printability with dye ink office printer. The cationic character of the coating layer was controlled with cationic additives, pigments and polymers. The pore volume of the paper coating was adjusted by choice of pigment and vinyl acetate acrylate (VAcA) latex, and coverage by using different coating methods. The pilot-scale coated papers were printed with a high speed inkjet printing machine with dye-based inks at two printing speeds.

The cationic character of the coating improved significantly water fastness and wet rub resistance of the inkjet printed papers, but did not influence print-through or smearing on a high speed printing machine. Porosity of the coated paper was found not to contribute to ink adhesion, but it had a significant effect on printing machine runnability (smearing), intercolor bleeding, mottling and print-through properties of the printed paper. Film transfer coating appeared, when compared to blade coating, to be advantageous in respect to ink wet rub resistance and print non-uniformity.

As a summary, high cationic charges are needed for good ink adhesion. High porosity reduces smearing in high speed printing, but may cause unevenness of the ink layer due to insufficient ink spreading. High porosity may also cause print-through problems. Inkjet printability can be controlled by the variables tested in the study to obtain a well-behaving compromise between coating colour stability, ink adhesion and ink holdout properties.

Keywords: inkjet, printability, porosity, cationic charges, smearing, water fastness, intercolor bleeding

1. Introduction and background

In inkjet printing ink droplets are ejected directly on to media from a jetting device controlled by an electronic signal. The inkjet printer is the dominating color printer in offices and homes because of its low cost. The inkjet technology is also becoming more common for industrial applications, both due to its ability to print on a variety of substrate types and, especially in the package printing industry, due to its print-on-demand capability.

Previous studies, by e.g. Rigdway et al. 2006, 2011a, 2011b, Lamminmäki 2012, have highlighted the roles of coating layer porosity and surface chemistry on inkjet print quality. Uncoated paper absorbs ink too fast and one may get problems with the print-through. Pigment coating can be used to increase ink holdout and to reduce print-through. The ink spreading and absorption can be controlled by

adjusting coating layer porosity with a proper choice of coating pigment, binder and other coating components. Besides these structural properties ionic interactions between the coating color components and the dye-based ink can be used to ensure adequate ink adhesion. The coating layer surface should have suitable chemical groups in relation to the ink being used.

Inkjet printing on coated paper can be described as a step-wise process. The first step is wetting of paper surface by an ink droplet. Here the droplet begins to fill in the voids between the coating layer pigments and spreads in the x-y plane of the surface. In the second step, ink liquid is absorbed into the porous coating layer by capillary forces. The pore structure of the coating layer has a major effect on how fast the ink is absorbed. If there is insufficient void volume between the pigments, the droplet will spread excessively on the surface causing so-called bleeding. The third step is diffusive transport or differential separation of colorant and carrier fluid. The diffusion can consist of bulk diffusion, Knudsen diffusion or surface diffusion depending on surface chemistry and sizes of the pores in coating layer in relation to the diffusing species (Lamminmäki, 2012).

Common problems in high speed inkjet printing are related slow ink absorption by paper, which can cause smearing during printing and poor print quality from intercolor bleeding. However, excessive absorption can create print-through issues. High quality inkjet-printed papers should also have high print uniformity, water fastness and wet rub resistance. The objective of the current work was to understand how inkjet printability is influenced by coating layer porosity together with its cationic charge. Different levels and types of cationic charge were created by using different cationic components in the coating color.

2. Materials and methods

2.1 Laboratory scale pre-study

Laboratory scale rheology testing, coating and printing with Canon Pro 100 dye ink office printer were used to select coating colour formulations for pilot scale coating trials. The pre-study aimed at identifying those combinations of raw materials that were expected to both have sufficient runnability on coater and improve inkjet print quality. In laboratory scale, positive results in water fastness and wet rub resistance tests were obtained when using various cationic additives. As expected, the cationic charges on paper surface clearly improved the adhesion of anionic dye-based inks. A new generation vinyl acetate acrylate (VAcA) binder was also tested and found to improve ink holdout and print-through properties. Overall, closed surface structure and the cationic charges of the coated paper appeared to improve print-through and ink adhesion.

2.2 Materials

In the pilot scale study, porosity, pore size distribution and the role of coating colour cationic charge were in focus. Table 1 has the materials used in the pilot scale trials. The material parameters studied were:

- Cationic polymers and additives
- Pigment composition: carbonate vs. structured cationic specialty pigment
- VAcA-latex
- Type of coater: film transfer vs. blade coating
- Coat weight

The coating colour formulations used in the pilot scale coating trials are in Table 2. Coatings were done both with film transfer and blade coater on woodfree pre-coated base paper. Target coat weight was 7 gsm, with the exception of results in Figures 10 and 11, in which 9 gsm and 1 gsm were also used. Papers were lightly supercalandered after coating at fixed calendering conditions.

Material	Description		
Calcium carbonate 1	PCC		
Calcium carbonate 2	GCC, 60% < 2 ym		
Cationic additive 1	CH IQ 1001 ¹		
Cationic additive 2	CH IQ 1002 ¹		
VAcA	CHP 1000i ²		
Cationic starch			
PVA	PVA 4-98		
Cationic polymer	FL 28405 ¹		
Cationic pigment	FP 300 CS^3		
¹ Oy Chemec Ab			
² CH-Polymers Oy			

Table 1. Materials used in the laboratory and pilot coating trials

³FP Pigments Oy

	Coating colour			
	1	2	3	4
Cationic charge	low	medium	high	high
Porosity	low	medium	medium	high
Calcium carbonate 1	70pph	70pph	70pph	
Calcium carbonate 2	30pph	30pph	30pph	
Structured cationic specialty pigment				100pph
CaCl ₂ addition	Х		х	
VAcA (CHP 1000i)		6pph	9pph	5pph
Cationic additive 1		30pph		••
Cationic additive 2				7.5pph
Cationic starch amount	high	low	medium	
PVA	0.5pph	0.5pph	0.5pph	
Cationic polymer			1pph	

Table 2. The pilot scale coating colours

pph = parts per hundred parts pigment by weight

2.3 Analysis of the inkjet printability

Pilot coated papers were printed with a high speed inkjet printing machine (Ricoh Infoprint 5000) using dye-based inks. Printing was done at two different speeds 32 m/min and 64 m/min. The following inkjet printability properties were measured:

- Water fastness
- Wet rub resistance
- Print-through
- Visual evaluation of ink evenness (mottling) and intercolor ink bleeding
- Ink penetration with light microscopy from paper cross-sections
- Porosity with Hg-porosimetry from pressure-filtrated coating colour tablets

2.3.1 Water fastness

Water fastness was quantified by measuring maintained color density of 100% cyan area after placing the sample in deionized water for one minute and letting it dry vertically in standard conditioned room overnight. The result is given as percentage; 100% signifying perfect water fastness.

2.3.2 Wet rub resistance

An automatic abrasion apparatus (Byk abrasion tester) was used for the wet rub resistance test in order to produce repeatable rubbing. Color density of a black printed 4x4 cm sample was measured before the rubbing test was started. Prior to rubbing, three drops of deionized water were placed onto the sample and allowed to absorb for 10 seconds. After 10x rubbing motions, the sample was left to dry overnight and the color density of the rubbed off ink at 1 cm distance from the edge of the black print was measured. Since the method appeared to produce unreliable results at times, a visual ranking was used to complement the measurement.

2.3.3 Print-through

Print-through was measured as whiteness of printed black area through the paper, when placed on white background. The result is given as percentage; 100% corresponding to no print-through.

3. Results and discussion

3.1 The effect of cationic charge

The cationic charge of the coating colours was varied from low to high by using different types of cationic additives and polymers. The porosity of the most cationic coating colour was also increased with a structured specialty pigment. The charge level of the coated paper improved significantly ink adhesion properties as shown in Figures 1 and 2, which show water fastness and wet rub resistance.

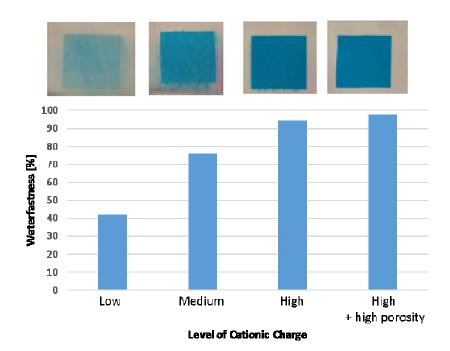


Figure 1. Effect of the cationic charge level on water fastness of dye-based ink.

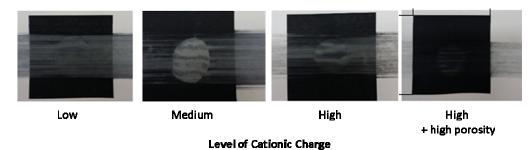
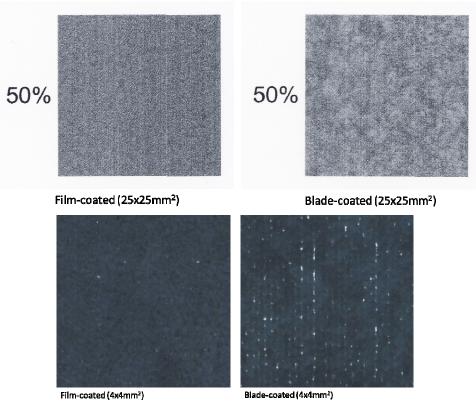


Figure 2. The effect of cationic charge on wet rub resistance of dye-based ink.

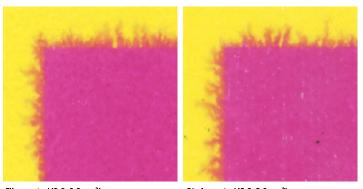
3.2 The effect of coating method

A clear difference in print quality was observed between the papers coated with film transfer coating and blade coating. Majority of the blade coated samples showed unacceptable print unevenness (mottle) as shown to the top in Figure 3. The length scale of the non-uniformity suggests this to be due to non-uniform coating layer thickness distribution caused by blade metering. Ink leveling on the blade coated samples seemed to be lower than on the film coated samples. This can be seen as white dots in the bottom right, in figure 3. However, no significant differences in line widths or line edge blurriness in black printed lines (Figure 4 top) or in intercolor bleeding (Figure 4 bottom) could be seen.





Film-coated (3.5x3.5mm²) Blade-coated (3.5x3.5mm²) Figure 3. The effect of coating method on print unevenness.



Film-coated (3.2x3.2mm²) Blade-coated (3.2x3.2mm²) Figure 4. The effect of coating method on line edge blurriness (top) and intercolor bleeding (bottom).

The film transfer coated samples had higher wet rub resistance than the blade coated ones (Figure 5). Furthermore, film transfer coated papers produced less smearing during high speed inkjet printing. According to X-ray photoelectron spectroscopy analysis the paper coated with the film transfer technique had more VAcA latex at the coating top layer compared to the blade coated paper. This may explain the differences in smearing and ink hold out due to the characteristic properties of this type of latex. The reasons for the differences in inkjet printability observed between the coating methods can also be related to coating coverage and surface roughness.

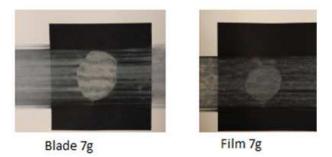


Figure 5. The effect of coating method wet rub resistance

3.3 The effect of coating porosity

Hg-porosimetry was used to characterize porosity and poresize distribution of the coatings. Direct measurement of the coated papers is complicated by the pre-coating and, therefore, pressure-filtered samples of the coating colors were used instead. The approach assumes that the coating structure is similar when coated on paper.

Clear differences in porosity and poresize distributions between the coatings can be seen (Figure 6.) The addition of VAcA-latex increased the porosity of the coating layer (samples 2 & 3) and the structured specialty pigment (sample 4) gave the highest pore volume.

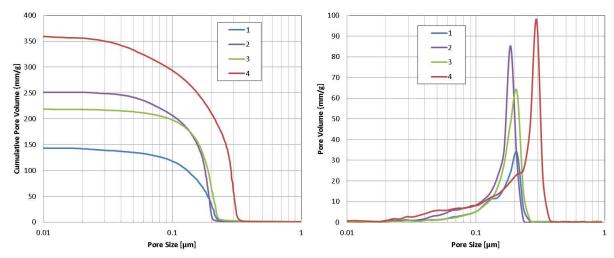


Figure 6. Poresize distributions of coatings used in pilot coating trial.

The porosity of the coating layers influenced the high speed inkjet printability. The most porous coating, sample 4, could be printed at the highest speed, 64 m/min, whereas the coating with lowest porosity (sample 1), could not be printed without problems, not even at 32 m/min. The effect of coating layer porosity on ink smearing during printing can be seen in Figure 7.

Open pore structure with a high number of large pores improves inkjet printing machine runnability (reduces smearing) due to the high permeability of the coating layer. The high porosity also reduces intercolor bleeding as shown in figure 8. However, a too rapid z-directional ink absorption into a porous coating can also reduce the ink levelling and cause unevenness in the printed image. This is evident in Sample 4 in Figure 8. Permeable coating can also lead to poor print-through performance (Figure 9).

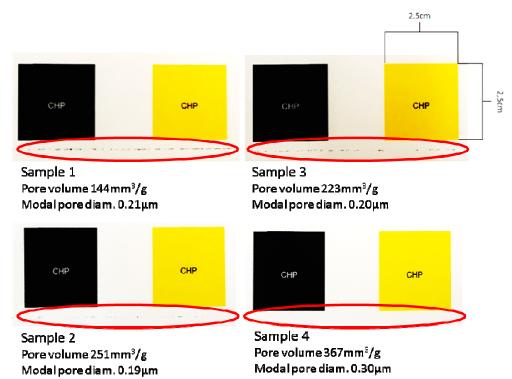


Figure 7. Too low porosity coating causes smearing during high speed printing even at low printing speeds of 32 m/min (sample 1). Paper with the highest porosity (sample 4) could be printed at 64 m/min speed with no smearing.

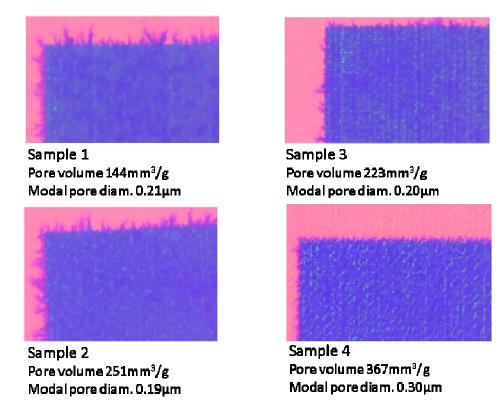


Figure 8. The effect of coating pore structure on intercolor bleeding.

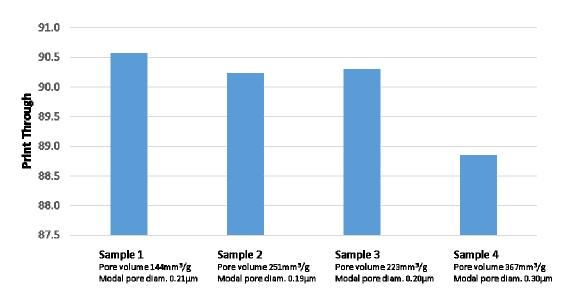


Figure 9. The effect of coating pore structure on print-through. Higher values indicate better print-through performance.

Coating layer pore volume available of ink absorption can also be increased by increasing the coat weight. Figure 10 shows how intercolor bleeding reduces when the coat weight is increased from 7 gsm to 9 gsm and further to 11 gsm. Previously printed ink is able to absorb into the paper, thereby reducing the spreading of the ink on top. For the same reason, the water fastness improves as well (Figure 11).

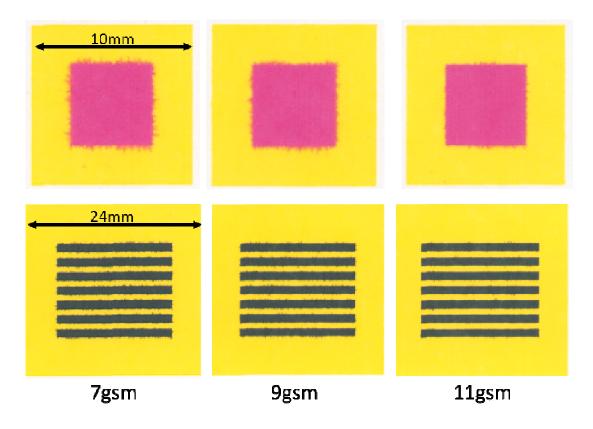


Figure 10. The effect of coat weight on intercolor bleeding. Blade coated samples.

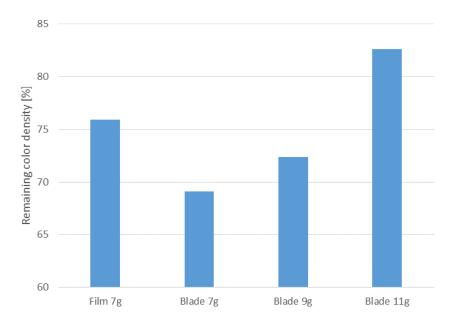


Figure 11. The effect of coat weight on water fastness.

4. Conclusions

A cationic charge of the paper coating was clearly a necessity for good ink adhesion when printing with anionic dye-based inks. The higher the cationic charge of the coated paper was, the higher water fastness and wet rub resistance were. The pore structure of the coated paper correlated well with the inkjet printability. High number of large pores increased the ink penetration in z-direction (higher permeability) providing low intercolor bleeding but causing print-through problems and high unevenness in print. The most porous coating had best high speed inkjet runnability even at high printing speeds. The print-through caused by a permeable coating can potentially be improved by closing the base paper surface for example by precoating. In the current study, film transfer coating produced papers that had better inkjet print quality than those coated with a blade coater. Possible explanations for this might be the differences in coating coverage, surface roughness and the location of the latex.

References

Lamminmäki, T., 2012. The comparative dynamics of bulk liquid flow and interpolymer diffusion during inkjet ink imbibition in porous coating structures (Doctoral dissertation), Aalto University School of Chemical Technology (Espoo, Finland)

Ridgway, C. J., Gane, P. A. C., J. Schoelkopf, J., 2006. Achieving Rapid Absorption and Extensive Liquid Uptake Capacity in Porous Structures by Decoupling Capillarity and Permeability: Nanoporous Modified Calcium Carbonate, Transport in Porous Media 63: 239–259

Ridgway, C.J., Kukkamo, V., Gane, P.A.C, 2011. Effects of Binders and Additives on Inkjet Coating Pigment Pore Structures. TAPPI PaperCon, TAPPI Press, Atlanta.

Ridgway, C. J., Schoelkopf, J., Gane, P. A C., 2011. Competitive Absorption of Polar and Non-Polar Liquids into Latex Bound Porous Structures of Fine Ground Calcium Carbonate, Transport in Porous Media 86:945–964

42nd International Research Conference, 06 to 09 September 2015



Session 5B

User and audience experience 2

Tuesday, 8 September 2015 11:05 – 12:30

Chair: Claire Gauzente

Why do certain Consumers avoid new Media Developments? An Investigation of three prudent users' Subjectivity

Stéphanie Gauttier, Claire Gauzente

LEMNA, University of Nantes

E-mail: stephanie.gauttier@gmail.com; claire.gauzente@univ-nantes.fr

Short Abstract

New media development constantly challenges consumer's habits. While innovations are supposed to bring new facilities to users, a certain number of them still remain reluctant in accepting, adopting and using new media offers. The aim of this research is to review the theoretical frameworks that are available for understanding such consumers' attitudes and behaviours and to confront theoretical analysis with in-depth subjective investigation of three prudent users facing a set of different mobile media offers. The empirical analysis is conducted using Q-method. Preliminary results offer a nuanced view of the representational space of non- and partial acceptation, adoption and use of new media developments.

Keywords: media and technology adoption, rejection attitude, non-use, partial use, Q-method

1. Introduction and background

New media developments are part of pervasive technologies. They completely permeate our everyday life as they grow mobile and, yet, a certain number of people still avoid, resist and even refuse to use them as much as possible. While a lot of studies have been dedicated to the digital divide that tend to explain IT and new medias non-use as the result of either economic, cognitive or socio-cultural deficiencies, researchers have begun to refine the study of IT and media non-use by integrating progressively attitude, technophobia and ideology (Selwyn, 2006). In organisational contexts, the use of technologies has been studied as a process of acceptance with quantitative models (Davies, 1989). These reference models, although becoming more and more sophisticated, fail to explain 100% of use/non-use and, more fundamentally lack qualitative nuances. Trying to bridge the two literatures and research traditions, the one directed at non-use and digital divide and the one dedicated to technology acceptance and use, we suggest that Q-method studies can document effectively the viewpoint of reluctant consumers and unveil to what extent non-use is actual, partial, arbitrary, paradoxical, and far from a binary variable.

Trying to map the theoretical and empirical contributions to the understanding of the use or non-use of media technologies leads us to discover a fragmented, although very rich, literature. Different disciplines have been looking at technology and media use/adoption/acceptance or non-use and include economic and management sciences, education, psychology, sociology, ergonomics and human-machine (or computer)-interaction.

Bringing these contributions together, it should be noticed that although literature dedicated to usage and literature dedicated to non-usage should have mirror-like logic, the underlying perspectives are indeed different. The stance of non-usage studies follows more or less from the digital divide research tradition while the usage-centred studies can be split into two sub-categories identified by Brangier, Hammes-Adelé and Bastien (2010): the operational acceptation on one hand and the social acceptation on the other hand.

The operational acceptation of technology derives from the ergonomics where the usability, ergonomic criteria and model of interaction are at the heart of technological acceptance. However,

beyond operational acceptance which is important from a design standpoint, Orlikowski and Baroudi (1991, p. 7) pointed out that researchers need to understand social processes that underlie the introduction, creation, use, mis-use and abandon of ICT. For this reason, several theoretical attempts were made in order to conceptualise technology use. One of the most prominent models is the TAM -Technology Acceptance Model, proposed by Davis (1986, 1989) and refined in subsequent publications. This model explains intention to use and actual use by attitude toward technology, perceived usefulness and perceived ease of use. This model is very popular and has been tested in many settings. Critiques and refinements have also emerged with time (Legris, Ingham and Collerette, 2003; Turner, Kitchenham, Brereton, Chareters and Budgen, 2010, Brangier et alii, op. cit.). Alternative conceptualisations have also been proposed. Among the most fruitful, we can cite the Expectation-Confirmation Theory - ECT (Bhattacherjee, 2001; Bhattacherjee and Premhumar, 2004) derived from the Satisfaction Approach (Oliver, 1980), the Task-Techno Fit Model - TTF (Goodhue and Thompson, 1995), the Structurationist approach (DeSanctis and Poole, 1994; Orlikowski, 1992), and the Coping Model of User Adaptation (Beaudry and Pinsonneau, 2005, 2010). These approaches address different stages and aspects of ICT acceptance and use. The ECT model suggests that expectations toward the technology at one point in time and the satisfaction derived from its first use conditions future use. Continued ICT use is then related to expectations and their confirmation via satisfaction. The TTF model holds that utilization depends on the task to be accomplished and the technological characteristics. When task requirements and technology characteristics fit, utilization and satisfaction are more liable. Fit assessment depends on the use context. In the long run, the individuals' performance in ICT use is also more likely. The structurationist approach points out that IT use, mainly in organizations, challenges theory and organizations as it induces changes. A dynamic and interactionist view is proposed and emphasizes social interactions emerging from IT adoption and use. The Coping Model of User Adaptation suggests that emotions constitute a significant part of ICT acceptation, first use and continued use. Different emotions have been studied: enjoyment, pleasure, anxiety and playfulness.

All in all, these different approaches hold a common view that technology acceptation and use is socially constructed, influenced by utilization context, and by individuals' emotions and attitudes. In line with this, IT use or non-use does not merely appear as a matter of meeting ergonomic or technical requirements. The social stance at accepting and using technology is also prevalent in the non-usage literature.

Non-usage approaches include many digital divide studies such as Rice and Katz (2003) and represent a dynamic stream of research (Brandtzæg, Heim and Karahasanovic, 2011; Cruz-Jesus, Oliveira and Bacao, 2012). The most classical predictors of digital inequalities are income or socio-economic status as well as gender, age, education, and family structure. Beyond these traditional variables, Verdegem and Verhoest (2009) suggest that integrating usage-centred literature, and particularly the utility concept, opens alternative interpretations of non-use. They develop the ASA model, comprising Access, Skills and Attitude, in order to explain technology appropriation and thus e-inclusion or exclusion. Their investigation distinguishes five different profiles of non-users.

This echoes the work by Selwyn (2003, 2006) who already underlined that the patterns of nonengagement in technology and media vary between technologies and that there are different types of non-users. Typically, Selwyn distinguishes three reasons for non-usage: non-access (lack of economic, cultural or cognitive resources); technophobia and ideological refusal. Other refinements exist, for instance, Jauréguiberry (2012) focuses more precisely on voluntary non-usage where non-use is not necessarily absolute, but is rather partial (certain usages are simply paused) and segmented (limited to certain periods of the day for instance).

All this research suggests that understanding the representational space of consumers, especially those that remain prudent or even avoid new media adoption, is of foremost importace for both researchers and professionals.

2. Materials and Methods

We first describe the general method (2.1) and then, the investigation instrument (2.2.).

2.1 A brief presentation of Q-method

The Q-method (see q-method.org and Brown, 1993), was developed by the psychologist Stephenson (1935; 1953). Contrary to other classical approaches in psychology, where the aim is to objectivise the inner state of an individual, he considered the subjective view most important. He conceived a method aimed at the study of subjectivity. Subjectivity is conceptualised as what "emanates from a particular vantage point" (Brown, online). Fundamentally, Q-method belongs to qualitative approaches.

The Q-method rests on two important pillars: one is theoretical (concourse theory) and the other is methodological (q-sorting procedure and q-factorial analysis). Concourse theory suggests that meaning is dependent upon context and therefore not given in abstracto. Concourse can be defined as the volume of available statements on a topic. In other words: "concourse is the common coinage of societies large and small, and is designed to cover everything from community gossip and public opinion to the esoteric discussions of scientists and philosophers" (Brown, op. cit.). Even for one individual, a single word can have several meanings, depending on circumstances and the individual's mind-set. What is important to note is that these meanings partly overlap with other people's meanings, and this is what makes interpersonal communication possible.

Based on these considerations, Stephenson suggests that initial qualitative interviews or literature reviews should be conducted to generate as many meanings as possible concerning one topic. These meanings can be formulated as assertions. Altogether, assertions constitute the q-sample. Then, individuals are asked to rank-order assertions according to the degree to which they represent their subjective view of one topic (say, augmented reality). The respondents are designated as the p-sample. This ranking procedure, called q-sorting, requires that only a small portion of assertions will be selected as highly representative of one's personal vantage point, and only a small portion will be selected as badly representative; the majority will be neither representative nor non-representative. The result of the q-sorting process is a q-sort.

Factor analysis is then used to analyze the data. This is called q-factor analysis because instead of analyzing individuals, assertions or statements are analyzed. A map of the representations that people have is produced, which helps to identify the different visions that people share. Factor analysis is thus used to identify underlying q-factors which correspond to shared visions. It should be emphasized that the q-factors should be in no way assimilated to groups of people as in typological approaches. The factors are not a statistical representation of groups in the general population. Q factors are shared views, shared interpretation of one object.

2.2 Investigation instrument

The q-sort instrument is based on 29 statements (so q-sample=29) that were selected to cover different viewpoints. Interestingly, the emerging statements cover the different theoretical dimensions identified in both research traditions (Information Systems Research and Digital Divide/Non Usage tradition).

Nine conditions of instruction were given, resulting in 9 q-sorts per respondent (p sample=3). The nine conditions entail the participant's a-priori view of digital media – as opposed to print - and his a-posteriori view once the different technologies and services have been presented and judged. The different technologies and services have been chosen considering several aspects:

- the current state of media technologies (m-ticket, QR code)
- popularity: "in-progress" media technologies that benefited from buzz and that most people are aware of (like google glass, augmented reality)

- the functionality of media technologies (m-ticket, m-payment)
- the fact that technologies are clearly documented online so that the participant can understand how it works.

In order to gather more social dimensions we also include the view of what future generations or parents might think. Finally, 9 conditions of instructions are retained. The conditions of instructions are as follows:

- 1) What is your general viewpoint about available mobile media technologies?
- 2) About m-ticket for public transportation?
- 3) About QR code advertising?
- 4) About augmented-reality product pre-visualization?
- 5) About Google glass?
- 6) About m-payments?
- 7) Now that we have seen several possibilities, please reassess your view of mobile services and technologies
- 8) In your opinion, what would be the view of your parents?
- 9) In your opinion, what would be the view of future generations?

For each q-sort instrument, an example of the media technology was first given either through pictures or online video demonstrations in order to make sure that the participants understood well the media technology features.

2.3 Case study selection

Three individual case studies were selected based on their mobile technology use. A reasoned sampling is adopted with users we firstly interviewed (on a slight basis) in order to assess where they were using or not mobile technology. Then if they accepted to participate to the study a more detailed questionnaire was included in order to precisely asses (and confirm) their profile. The questionnaire entails question such as: how do you possess an cell phone, have you ever bought product or services with your well phone, do you use apps? Do you have a mobile internet access?

	Mobile internet access	Attitude toward tech	Mob apps use	Mobile purchase
Marnie	No	Unfavorable (2/5)	No	No
Juliette	No	Unfavorable (2/5)	No	No
Louise	Yes	Neutral (3/5)	Yes, emails and FB	No

Table 1.	Individual	cases	brief	description	

From this we can see that two cases can be easily considered as non users and one as an occasional user.

3. Results and Discussion

We will first review the results of two non-users and then analyse the results of an occasional user.

3.1 Non-users

3.1.1 Non-user case 1, Marnie

The first prudent consumer is a French 36-year-old female called Marnie. She is a well-educated person (master degree in management science) and she can be qualified as a media technology-aware person as she's been using a computer and internet for years for both professional and private purposes. In terms of mobility, she has long possessed a cell phone and has had a smart phone for 4 months. She does not use mobile media (except SMS) because she did not subscribe to any internet access.

Centroid analysis is used and judgemental rotation applied to selected factors as recommended in Qmethod. In the present case, thanks to the two factors, we can identify actually four visions including one specific case (q-sort#6) because factor 2 is bipolar. Factor 1 (31% of variance) entails four q-sorts: general opinion ex-ante and ex-post, QR-codes and Google glass. Factor 2 (33%) opposes (a) the vision for future generations, together with the vision of m-tickets and AR-pre-visualization versus (b) the vision for parents. The fourth vision corresponds to q-sort #6 which pertains to m-payment.

Vision#1 corresponds to Marnie's general vision of mobile media technologies. This general vision is stable over time as the ex-post evaluation is in line with this. QR Codes applications seem to be very typical of her general vision which is one of defiance toward these technologies: "I am not fond of these technologies", "data protection is important. Marnie does not "feel curious" at all about mobile technologies. This defiance and potential rejection is nurtured by critical considerations: "we are dependent upon our mobiles", "I don't really need it". This vision of mobile media technologies is driven by rational and critical thinking and leads to distanciation.

Vision#2 (a & b) is a more complex one. Marnie considers that her parents and future generations would hold opposite attitudes toward mobile media technologies. Both hedonic and functional dimensions of mobile services appear in relation with future generations. A hedonic instance is given with mobile Augmented Reality Apps helping to interactively visualize the effect of wearing glasses on people's face. A functional instance is given through mobile ticketing for public transportation. Marnie considers that mobile services may "make life easier" and can even be "exciting!", "pleasant" and "playful". A certain level of "curiosity to try these technologies" arises. Some issues appear without relevancy such as "data protection". On the contrary, older generations, such as her parents, would consider mobile services and technologies to be potentially "dangerousand would consider it to make us "more and more dependant upon technology". We also observe that "distrust concerning security" also arises.

The last vision (corresponding to q-sort#6) pertains to mobile payment. Marnie appears highly concerned with this and considers this technology to involve a "danger" and generates distrust which probably derives from perceived financial risks. She also feels more comfortable with "conventional" payment methods. While recognizing that this technology will be more and more present "in the future", she does not consider it as "exciting" or "playful and fun" at all. This view echoes Vision#1 where Marnie's core vision is expressed. This q-sort exemplifies however a sharper standpoint than vision #1.

3.1.1 Non-User Case 2, Juliette

The second non-user of mobile technologies is a French 32 year-old woman called Juliette. She has had a smartphone for over three years because it didn't cost her much, but she doesn't have access to the Internet on it. She wouldn't be willing to buy with her cell phone at the moment and if she would she would choose preferably already known merchants.

The analysis of the q factor analysis reveals that her vision of technology is dual. The first vision she expresses encompasses all questionnaires but the one about future generations and is a very distanced one. It explains 64 % of the variance. The second vision she expresses includes only the q-sort on future generations and explains 10 % of the variance.

Vision #1 is negative. Juliette reckons she's not « fond of this technology », doesn't need it and doesn't want to try. Yet, she makes it clear that it is « modern » and that it's « probably the future ». Mobile technologies don't trigger any strong emotional response in her, she doesn't feel excited about them, doesn't find it pleasant, but she is not upset by the technology either. She doesn't think these technologies are dangerous. As far as ease-of-use is concerned, Juliette first vision considers mobile technologies are complicated. Finally, she says she doesn't need mobile technologies, yet they could be practical. The irrelevance of mobile technologies and complicatedness appear as her biggest barrier to using it.

Vision #2 is diametrically opposed to this. In this view, mobile technologies are seen as « fun and playful », « pleasant » and « exciting ». Juliette's vision of the future is one where mobile technologies are easy to use, to access, and fun to use. The lifestyle Juliette imagines for future generations, different from her reality, could explain the discrepancy between the two visions. Another explanation could be in the distance Juliette puts between herself and mobile technologies as seen in the first vision, which may not apply when she thinks of others.

3.2. Occasional Users

The third case is the one of a 26 year-old French woman named Louise. Louise has had a smartphone for 2 years and uses mobile media occasionally. She says that her mobile phone is a functional thing. Mobile internet is used only in special circumstances when she's in a waiting room for example or when travelling in order to access her email and social networks. She prefers to use her laptop to access internet services.

The analysis of Louise's q-sorts shows a dual attitude towards mobile technologies: factor 1, explaining 36 % of the variance, encompasses all questionnaires except the ones about previsualization, google glasses and future generations. Factor 2, explaining 19 % of the variance, includes only q-sorts on google glasses and future generations. These two factors reveal a dramatic change of attitude towards mobile technologies depending on the temporal dimensions Louise projects herself in. She is skeptical towards mobile technologies that are already available and that she could be using today, but she shows enthusiasm for technologies less spread technologies like Google Glasses and imagining the future.

Vision #1 expresses skepticism towards present mobile technologies. First, lack of trust explains this attitude. Statements like «I feel it raises distrust about payment security » and « Data protection is essential » are determinant in shaping Louise's point of view. Then, Louise has a distant relationship to mobile technologies: she doesn't see a role for them in her life, « It's not vital, I don't need it », and they don't trigger a positive emotional response. Indeed, Louise agrees that she is « not fond of this technology » and disagrees with the statements « I feel curious about trying it », « These technologies are exciting! », and « I find it pleasant to be able to use these technologies ». Therefore, she's not keen on trying. From an ideological perspective, she seems to fight the idea of technologies replacing going to the store, not because she needs the haptic experience but likely because it takes away human contact from the experience. Despite this, Louise admits that these technologies could be convenient. She might resolve to use them in specific situations.

In the second, enthusiastic and future-oriented view Louise expresses, statements about enjoyment such as « I feel curious about trying it » or « I find it pleasant to be able to use these technologies » are positively rated . Her concerns about payment security and data protection vanish. When thinking

about the future, she believes that « It's modern and coherent with our more and more mobile way of life », and rates negatively the idea that these technologies are not « vital ». It seems that mobile technologies make more sense to her in a future context because she imagines an alternative lifestyle, so her attitude to technology may be highly depending on context.

3.3 Discussion

The analysis of these case studies shows that determinants of use and non-use are varied. It appeared that ideological aspects, the lack of positive emotional relationship to technology, the functional aspects of technologies and their relevance to one's lifestyle, play a major role in structuring respondents' attitudes and behaviour toward mobile media.

Indeed, non-users and the occasional user have a neutral relationship to mobile media, and both of them didn't identify situations when mobile technologies would be handy. Both respondents reckoned some issues linked to privacy or security, which vanished when they were thinking of next generations. These prudent (and even reluctant) users seem to wait for an evolution of mobile technologies, which would make it safer to use them.

In the case of the occasional user, we see that there is no positive emotional relationship to mobile technologies. Besides, ideological considerations are playing a role and might be a barrier to using mobile media more. Convenience and relevance of mobile technologies explain that the respondent resolves to use them in particular situations. But again, the respondent seems convinced that in the future, technologies will be more exciting and relevant.

4. Conclusions

Our investigation of three prudent consumers documents how new media developments are subjectively appraised and indicates ways of improvement in terms of theory and practice. The study suggests that even if a negative attitude toward new media development has been formed, the benefits may be recognized and the media may be partially and occasionally adopted. A sense of "bricolage", including paradox, arises. Thanks to Q method, the different shades of media acceptation, adoption and appropriation can be documented and brought into light. "Non-use" can be pictured as a changeable geometry between different aspects and should encourage further development not only to understanding satisfaction criteria but also to unveil motivational factors of acceptation in relation with special use contexts.

References

Beaudry, A., & Pinsonneault, A. 2005. Understanding User Responses to Information Technology: A Coping Model of User Adaptation. *MIS Quarterly*, 29(3), pp. 493-524.

Beaudry, A., & Pinsonneault, A. 2010. The Other Side of Acceptance: Studying the Direct and Indirect Effects of Emotions on Information Technology Use. *MIS Quarterly*, 34(4), pp. 689-710.

Bhattacherjee, A. 2001. Understanding Information Systems Continuance: An Expectation-Confirmation Model. *MIS Quarterly*, 25(3), pp. 351.

Bhattacherjee, A., & Premkumar, G. 2004. Understanding changes in belief and attitude toward information technology usage: A theoretical model and longitudinal test. *MIS Quarterly*, pp. 229–254.

Brandtzæg, P. B., Heim, J., & Karahasanović, A. 2011. Understanding the new digital divide—A typology of Internet users in Europe. *International Journal of Human-Computer Studies*, 69(3), pp. 123–138.

Brangier, E., Hammes-Adelé, S., & Bastien, J. 2010. Analyse critique des approches de l'acceptation des technologies: de l'utilisabilité à la symbiose humain-technologie-organisation. *Revue Européenne de Psychologie Appliquée/European Review of Applied Psychology*, 60(2), pp. 129–146.

Brown, S.P. 1993. A Q methodological tutorial. *Operant Subjectivity*, 16, 91-138. Retrieved October 3, 2005, from <u>www.qmethod.org</u>.

Cruz-Jesus, F., Oliveira, T., & Bacao, F. 2012. Digital divide across the European Union. Information & Management, 49(6), pp. 278-291.

Davis Jr, F. D. 1986. A technology acceptance model for empirically testing new end-user information systems: *Theory and results*. Massachusetts Institute of Technology.

Davis, F. D., Bagozzi, R. P., & Warshaw, P. R. 1989. User acceptance of computer technology: a comparison of two theoretical models. *Management science*, 35(8), pp. 982–1003.

DeSanctis, G., & Poole, M. S. 1994. Capturing the complexity in advanced technology use: Adaptive structuration theory. *Organization science*, 5(2), pp. 121–147.

Goodhue, D. L., & Thompson, R. L. 1995. Task-technology fit and individual performance. *MIS quarterly*, pp. 213–236.

Jauréguiberry, F. 2012. Retour sur les théories du non-usage des technologies de communication. *Connexions: communication numérique et lieu social* (éds. S. Proulx et A. Klein), Namur, Presses universitaires de Namurn, 335–350.

Legris, P., Ingham, J., & Collerette, P. 2003. Why do people use information technology? A critical review of the technology acceptance model. *Information & management*, 40(3), pp. 191–204.

Oliver, R. L. 1980. A cognitive model of the antecedents and consequences of satisfaction decisions. *Journal of marketing research*, pp. 460–469.

Orlikowski, W. J. 1992. The duality of technology: Rethinking the concept of technology in organizations. *Organization science*, 3(3), pp. 398–427.

Orlikowski, W. J., & Baroudi, J. J. 1991. Studying information technology in organizations: Research approaches and assumptions. *Information systems research*, 2(1), pp. 1–28.

Rice, R. E., & Katz, J. E. (2003. Comparing internet and mobile phone usage: digital divides of usage, adoption, and dropouts. *Telecommunications Policy*, 27(8), pp. 597–623.

Selwyn, N. 2003. Apart from technology: understanding people's non-use of information and communication technologies in everyday life. *Technology in society*, 25(1), pp. 99–116.

Selwyn, N. 2006. Digital division or digital decision? A study of non-users and low-users of computers. *Poetics*, 34(4), pp. 273–292.

Stephenson, W. 1935. Correlating Persons instead of Tests. Character and Personality, 4, pp. 17-24.

Stephenson, W. 1953. The study of behavior: Q-technique and its methodology. Chicago, University of Chicago Press.

Turner, M., Kitchenham, B., Brereton, P., Charters, S., & Budgen, D. 2010. Does the technology acceptance model predict actual use? A systematic literature review. *Information and Software Technology*, 52(5), pp. 463–479.

Verdegem, P., & Verhoest, P. 2009. Profiling the non-user: Rethinking policy initiatives stimulating ICT acceptance. *Telecommunications Policy*, 33(10), pp. 642–652.

Wiredu, G. O. 2007. User appropriation of mobile technologies: Motives, conditions and design properties. *Information and Organization*, 17(2), pp. 110–129.

Experimental Comparison of the User Experiences of Different Digital and Printed Newspaper Versions

Janne S. Laine

VTT Technical Research Centre of Finland Ltd

E-mail: janne.laine@vtt.fi

Short Abstract

A laboratory experiment comparing the user experiences elicited by five different newspaper publication versions (4 digital versions and the printed tabloid format newspaper) was carried out. The main goal was to learn of the different styles of news reading and reader preferences regarding the design and layout of the content, as related to these publication versions, and also to test user experience measurement and analysis methods in the context of news reading and varying layouts. The results show considerable differences between the user experiences evoked by all five publications versions. The participants could be roughly divided into two main clusters based on their preferences of different publication versions. The most significant difference between the two groups seems to be the attitude towards reading news articles in the form of continuous streams as often found on different web sites. One group preferred the printed newspaper and digital versions that can be understood to stem from the tradition of the printed newspaper. The other group preferred the web style of news presentation and saw no need for digital versions mimicking the printed paper.

Keywords: user experience, digital newspaper, experience mapping, gaze tracking

1. Introduction and background

The ongoing digital transformation is strongly affecting the news reading habits of consumers. Media companies are increasingly offering different types of digital newspapers and digital news services. While the long tradition of printed newspaper making has resulted in a solid practical understanding of what kinds of designs work for printed newspapers, these design rules do not necessarily apply as such in the digital world. Thus there is a need for understanding the user experiences of digital newspapers, what kinds of experiences are preferred and how different design choices affect them. It is also useful to understand how the user experiences of various kinds of digital editions compare to the conventional printed newspaper.

We carried out a laboratory study comparing the user experience of the current 5 Helsingin Sanomat newspaper publication versions (4 digital versions and the printed tabloid format newspaper, as they were offered in the summer of 2013). (Helsingin Sanomat is the largest newspaper in Finland.) The main goal was to learn of the different styles of news reading and reader preferences regarding the design and layout of the content, as related to these publication versions, and also to test user experience measurement and analysis methods in the context of news reading and varying layouts. While necessarily restricted to publication versions from a single publisher, the results of these experiments were expected to provide more generally useful understanding of user experiences and preferences concerning news reading. A specific question we sought to answer was whether any single digital version would reasonably well satisfy different reading styles, suggesting that it would be appropriate to cut down the number of considerably different digital layout versions currently offered in favour of aiming for a more consistent publication design and reading experience across digital platforms.

2. Materials and Methods

2.1 Participants

40 persons participated in the laboratory experiments. The participants were recruited by various means: by contacting people who expressed their interest when answering a web survey aimed at the library patrons using a new e-book loaning system, and by e-mailing and directly contacting people working at the VTT premises in Espoo in various positions, as well as their family members and other acquaintances, and through contacts within the Next Media research program.

The sample of participants was convenient for this study, and, while not a random sample, was comprised of persons with relatively varying backgrounds. 19 of the participants were women and 21 were men. The ages of the participants ranged from 19 to 64 years old. The average age among the participants was 40 (with standard deviation of 9 years). Due to the requirements of the eye-tracking device used in the experiments, only participants who were able to read without wearing eyeglasses were recruited (contact lenses were allowed).

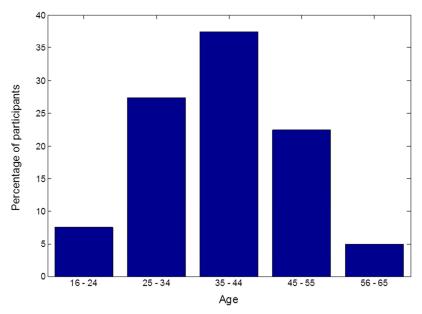


Figure 1: Age distribution of the participants

2.2 Newspaper versions

The five publication versions of Helsingin Sanomat newspaper shown in Figure 2, with current daily content, were used in the experiments. The different publication versions essentially share the same content, but there are considerable differences between the versions in the layout and the overall design. The four digital publication versions were all used with a black-framed Apple iPad 4 tablet.

A: Printed newspaper



B: iPad app

C: Digital edition (print replica)



D: Browser newspaper

E: News web site



Figure 2: Publication versions used in the experiments. Printed newspaper not to scale.

2.3 Experimental setup

After general introductions to the proceeding of the experimental session, the participant filled a digital survey form concerning the participant's background information (basic demographic information, and questions on news reading habits and the use of media technology, as well as questions measuring the personality traits of the respondent).

The core part of the experiment consisted of the participant browsing and reading each of the five publication versions (presented in randomized order) for five minutes. The participant was instructed

to imagine herself in a situation in which she had around five minutes to spare (e.g. waiting for an acquaintance to arrive in a cafeteria) and that she had decided to spend that time to take an overall look at the contents of today's newspaper (perhaps to read particularly interesting articles in more depth later), browsing and reading the publication in any way she wanted. The participants were instructed on how to use each version just before they were given that version for browsing. While reading the publication the participants wore SensoMotoric Instruments eye-tracking glasses, recalibrated before reading each publication version and connected to a recording unit which recorded the eye movements of the participants while browsing the publication as well as a video of the scene they were seeing in front of them (i.e. the publication being browsed).

Immediately after finishing reading the publication version, the participant answered a digital questionnaire containing statements related to the publication and its layout, as well as how it felt to use the publication. The questionnaire contained, in random order, the relevant statements from the Next Media MX Questionnaire (Helle et al., 2011), as well as additional statements from The User Engagement Scale (O'Brien, 2010). In total, the questionnaire was comprised of 92 items. The Likert-type items were in the form of statements, and the participants responded to the items using a 9-step slider from 1 ("completely disagree") to 9 ("completely agree").

After finishing the questionnaire, the respondent was shown a video recording of their five minutes browsing the publication, with a cursor showing their gaze path augmented on the video. They were asked to retrospectively comment, while viewing the video recording, on their browsing session: How they used a publication? What they thought and felt about the publication and its design and layout – from their point of view, how well did the publication present its content to the reader? When needed, the instructor probed the participant with additional (unstructured) questions during the retrospective commenting. Figure 3 shows a participant retrospectively commenting his experience with the publication version B.

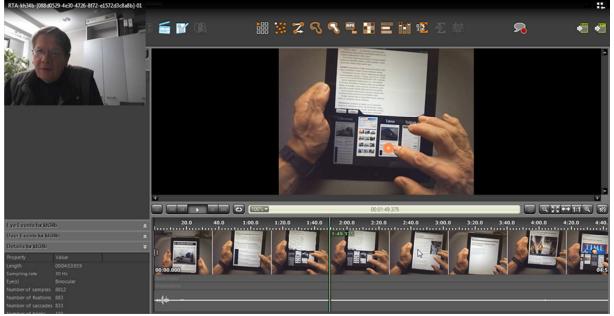


Figure 3: A frame from a video recording of the retrospective commenting session, using the SMI BeGaze analysis software. Participant (top left corner) comments on the iPad app version (in this case) and his experience with it while viewing the freshly recorded video showing his view during the session. The orange gaze cursor indicates the point of his visual focus.

After browsing all five publication versions, and providing the answers to the questionnaire as well as the retrospective comments for each version, the participant was asked to rank the publication versions in order of preference: Which publication version would they use if they could only choose one of them to use from now on in their daily? Once the most preferred version was chosen, that version was removed and the participant was asked to choose the preferred version to use from the remaining ones. This was repeated until all five versions had been chosen. The participant was asked to comment on his preferences while making the choices. What were the pros and cons of each version for him and what made it suitable (or not suitable) for his style of news reading?

Multivariate analysis was applied to the questionnaire data, including the Experience Mapping approach to further describe and compare the user experiences elicited by the different publication versions. The Experience Mapping, described for example by Mensonen et al. (2012), is based principal component analysis of multivariate observations (here: questionnaire items related to different aspects of user experience) of multiple samples (here: different publication versions), and is intended for visualizing and describing the most significant experiential differences within a given set of products, services, or concepts. The gaze paths were analysed using the SMI BeGaze software and other tools. The retrospective comments of the participants concerning their experiences with different versions were qualitatively examined, in order to further interpret the other results.

3. Results and Discussion

3.1 Preferences

Considerable variation in preferences and reading styles was evident among the respondents. The histograms in Figure 4 sum up the overall variability in the preference rankings of the five different publication versions. The first group of bars on the left side of the graph indicates the percentage of respondents that ranked the given publication version in the first place in their order of preference. The highest bar corresponds to version A, indicating that 42.5% of respondents ranked the printed newspaper first in their order of preference. While this was by far the favourite choice compared to the four other publication versions individually, it should be noted that the majority of participants would still prefer to use some digital publication version, rather than the printed newspaper, if they had to choose only one version for their daily news reading.

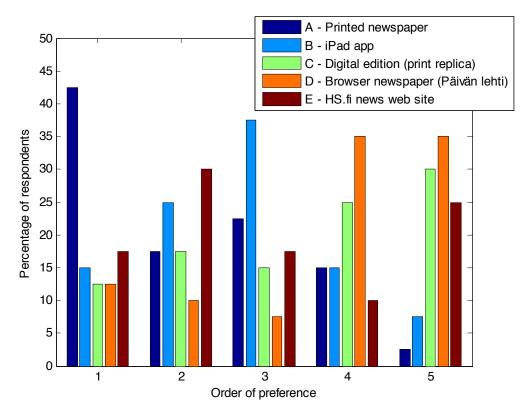


Figure 4: Histograms showing the overall distribution of preference rankings of the publication versions among the participants of the experiment.

Looking further at the graph of Figure 3, the second group of bars indicates the percentage of participants that ranked each publication version second in their order of preference – i.e. the percentage of participants that would choose a given publication version if their most preferred version (the one they ranked first in their order of preference) was not available to use in their daily news reading. Similarly, the following three groups of bars indicate the percentage of respondents that ranked specific publication versions on the 3rd, 4th, or 5th, respectively, in their order of preference.

3.2 Comparison of the experiences evoked by the publication versions

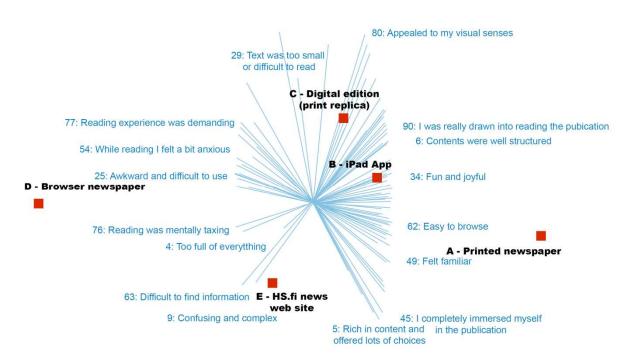


Figure 5: An Experience Map visualizing the experienced differences between the publications, for results averaged over all participants.

The Experience Map in Figure 5 visualizes the main differences in the user experiences evoked by the different publications. The blue vectors (only some of which are labelled for clarity) pointing in the general direction indicate attributes that were most strongly associated with the given publication version. Based on the experiment map and other material from the experiments, one can make the interpretations of the preference judgments and other observations on the user experiences of the five publication versions:

3.2.1 Version A: Printed newspaper

The printed tabloid format newspaper, which many participants considered easier to handle in many reading situations than the previous broadsheet version, was generally well regarded. The larger size of a newspaper spread, and the well-used possibilities it offered for laying out the content, was appreciated by many participants. Enjoyable reading experience overall, as well as familiarity, supported the choice of this version for many participants. However, a considerable number of participants, while acknowledging the enjoyable reading experience of a printed newspaper, preferred to read their daily news articles from a digital medium – reading daily news from a printed newspaper had no place in their current daily routines. Considering the gaze paths, and the allocation of attention to different elements of the publication, many participants found the way the advertisements were incorporated in the layout of the printed newspaper to be natural and pleasant for them: they could easily pay closer attention to advertisements if they spotted something interesting (or decide not to do

so) but they did not feel that this took away from the flow of reading the publication, as opposed to advertising in digital publications, which many participants commented to be distracting to their reading experience.

3.2.2 Version B: iPad app

The iPad app was generally considered to be visually rather impressive, and the navigation between sections via the bar available from the bottom corner to be intuitive to use. This native app also felt more responsive to most participants than the versions D and E, which were used in a web browser. Due to the relatively intuitive navigation, freedom from severe usability problems, and the pleasing visual appearance, the iPad app was generally rather well liked (77.5% of the participants ranked it in their top 3 when choosing the preferred version).

3.2.3 Version C: Digital edition (print replica)

Some participants thought that pleasant layout of the print version transferred rather nicely to the digital device in this digital edition that replicated the pages of the printed newspaper. Other advantages mentioned included the immediate familiarity to those used to reading the printed newspaper. Also, the navigation through the multi-page view of the miniaturized pages was considered intuitive in its simplicity by some participants, and provided a clear overall view of all content available within the publication (something that was often perceived to be lacking from the other digital versions). Some participants used this miniaturized view extensively to browse the publication, only tapping to go to individual pages if they spotted something that appeared interesting. On the other hand, some participants did not see the point of reading a replica of the printed pages on a digital device (especially on a relatively small screen of a tablet). Reading the text was particularly problematic on this version for those participants that were not able to comfortably read the text at the default size at which the pages were presented: they had to constantly combine two-fingered zooming (in order to read paragraphs) and dragging (to scroll the page), which was cumbersome.

3.2.4 Version D: Browser newspaper

Preferred by some for the ability to use it across different digital platforms, the browser newspaper (used on the Safari browse in the iPad) did not generally provide a very good user experience. The navigation was considered to be confusing. While some positive comments were given for the ability scroll the article headers separately on the right side of the front page, most participants found the fact that the dragging on the front page only affected a part of the page to be confusing. The fact that links to all sections were not initially visible in the navigation bar was considered a disadvantage by many participants. Many participants reported feeling lost when moving from article to another within the publication; for example, it was not intuitively obvious how to get back to an article or a part of the publication where they had previously been in. The user experience also suffered from occasional delays in responses to user actions (problems in network connections may have caused some of the delays).

3.2.5 Version E: News web page (HS.fi)

Unlike the other versions, which were daily publications, version E was a continuously updated news web site, and in this aspect essentially different from the other versions. It also divided the opinions the most among the participants. It is likely that the different styles of news reading were most strongly reflected in the opinions regarding this version. Many participants felt that the layout was too full of everything (with many screens worth of material to scroll through on the home page, and additional links on the right side) and preferred what they considered to be a cleaner structure of conventional news publications. However, for the participants used to reading their daily news on the web, this version offered what they wanted in a familiar format, a serving of news for quick reading, with possibilities to quickly jump to the latest or most popular articles. Other flavours of the web, like

seeing comments from other readers, also contributed to making this the preferred version for some readers.

3.3 Clustering of participants based on preferences

Looking further at the differences in the preferences among the participants, multivariate data analysis revealed two distinct clusters of participants. The preferences among the participants were similar within each cluster but differed considerably from the preferences of the other cluster. 19 of the 40 participants were classified as belonging to the first cluster, and 12 participants formed the second cluster. The remaining 9 participants were left outside these two clusters due to their different preferences, and did not form a third cluster of like-minded persons either.

The mean preference rankings for each publication version in the two participant clusters are shown in Figure 5. The main difference between the two clusters appears to be the preference for web style of news reading in cluster 1 (news web site, version E, highly preferred) and the preference for more conventional style of newspaper-like news reading in cluster 2 (version E least preferred, printed newspaper, version A, most preferred). While the printed newspaper was rather highly ranked in both clusters (higher in cluster 2), in cluster 1 the participants saw no need for a digital version replicating the printed newspaper (version C less preferred). In contrast, in cluster 2 this rather straightforward transformation of the conventional newspaper into a digital format was appreciated (version C more preferred).

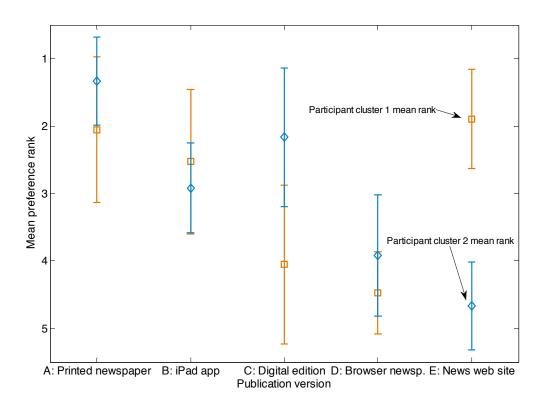


Figure 5: Mean preference rankings of the five publications versions for the participant clusters 1 (orange squares) and 2 (blue diamonds). The error bars indicate the standard deviation of the rankings around the mean for each publication version in each cluster. Notice that the ranking value decreases (preference increases) towards the top of the graph: for example, in participant cluster 1 version E (news web site) was the most preferred choice while in cluster 2 it was the least preferred publication version.

4. Discussion and conclusions

A laboratory experiment investigating and comparing the user experiences of five different versions of a newspaper was carried out. These versions, a printed tabloid size newspaper and four digital versions with different types of layouts and design choices, essentially provide five different ways of presenting the same content to the reader. The craft of designing layouts for printed newspapers has evolved over decades and centuries, and accumulated practical understanding of how to provide the contents of the day's newspaper to the readers in a format that is pleasant to read and use in different ways. In the experiment several different general motivations and styles for reading a newspaper were evident, such as:

- 1. Finding something interesting or entertaining to read, quickly and easily. The motivation is not to read news of any specific category or the find specific information, but rather to read news content as an enjoyable pastime.
- 2. Keeping up with what is going on in the world generally. Readers with this motivation typically want to browse through the whole publication, stopping to focus more on articles that they consider to be important.
- 3. Keeping informed of what is going on in areas that are personally interesting. Readers with this motivation value quick access to sections that are of interest to them, without having to pay attention to anything else.

The above three general categories covered the principal motivations among all the participants of the experiment, with many additional personal idiosyncrasies in the reading styles, as was to be expected. The printed newspaper lends itself well to most of these different styles of reading, assuming it is at hand in the given context. The accumulated learnings from the design of printed newspapers are not directly applicable to the world of digital publications, and digital newspapers are still finding their form. It is not clear what kind of design choices are most suitable in digital news publications, or if any single type of layout would satisfy different reading styles generally. A given digital version may be suitable for a certain style of reading but not for another one, depending on its layout and how its navigation is implemented. The results from the experiment indicated that this was indeed the case here. The vast majority of participants liked the reading experience of a printed newspaper, even if they reported that printed newspapers did not anymore have a place in their daily routines. However, there was considerable variation among the participants in which of the digital versions was preferred for the reading experience it provided, not explained by a person's attitude to printed newspaper.

The results indicated that none of the digital versions included in the experiment, while satisfying some readers, would do very well in catering to all styles of reading. Specific aspects in which all digital versions struggled to various degrees in comparison to the printed newspaper was in giving the reader an intuitive feel for all the content that is available in the publication and allowing the reader perceive her current position in the publication. Printed newspaper naturally enjoys all the benefits of a tangible physical object in this regard, while in the case of digital versions the layout choices, including the implementation of the navigation system, had a strong influence on how this is perceived. For some readers it was very important to have a good understanding of where a newspaper starts and where it ends, and to have a good idea of how to go back to a specific place in the publication. Others were fine with and preferred a web style continuous stream of news over an easierto-perceive set of news that they could digest. Indeed, these were the two main preference-based clusters among the participants. In both groups the reading experience that the printed newspaper provided was valued rather highly, the difference being in their attitudes and preferences regarding the digital newspaper version. One group preferred to have their digital news to be presented in a publication that followed the tradition of printed newspapers in packaging content, while the other group saw no need to carry this kind of layout over into the digital domain, preferring a news web site style of dynamic and continuous news stream.

Further findings were related to the flow of attention in different versions. A pleasant flow of attention is known to be central aspect of a good reading experience. Again the printed newspaper excelled in

this regard, with more variability among the digital versions. A common example that came up in the experiments was the placement of advertisements and how it affected the flow of reading. Many participants commented on how a newspaper spread allowed one to smoothly direct ones attention to interesting parts, quickly noticing different elements such as advertisements but not paying much attention unless they appeared interesting. In some digital versions participants were in many cases clearly distracted and irritated by advertisements when they suddenly interrupted their flow of attention: the publications did not include pop-up advertisements as such, but the effect was as distracting if the person suddenly found himself looking at an advertisement, partly forced on him by the layout, and had to make an effort to continue past the advertisement. Again, while the results did not provide clear-cut design rules for directing attention in digital news publications, it did show how different designs had considerable effects on the flow of attention and consequently on the overall experience provided by the publication.

Beyond the observations on the reading experiences and styles, the experiments served as test of combining and integrating different approaches to evaluating the user experience of media products and services. We found user testing with eye-tracking, followed by a retrospective commenting session to be a very useful approach for extracting valuable information for interpreting the other results. During the retrospective commenting we showed the participant a video recording of their reading session, augmented by the gaze path from the eye tracking glasses, and asked them to explain their own experience: what they were doing, what they thought and felt. Seeing the video recording with the visualization of their own gaze path appeared to be a good motivator for most persons to retrospectively and introspectively consider and discuss their own reading experience, and also to relate it to their reading style more generally.

The gaze path data revealed the focuses and transitions paths of attentions, the retrospective commenting provided qualitative data, the both of which served to explain results of the multivariate analysis the media experience questionnaire data. The final preference judgements between different publication versions provided and the comments to justify those judgments indicated what the aspects of the experience the participants found valuable to themselves, and together with other data, helped to paint a more complete picture of the user experiences of the tested publication versions.

The analysis of the quantitative data from the application of the rather extensive Next Media MX Questionnaire (Helle et al., 2011) showed that the questionnaire did in fact indicate meaningful differences between the tested publication versions on the different dimensions of media experience that it was designed to measure, and that the measures were consistent with the data acquired from other approaches. The questionnaire has not been so far been extensively tested, and the work done in this study does not yet constitute a proper validation of the questionnaire, but these results do suggest that the questionnaire can be a useful tool in measuring media experience. The results provide useful information for future considerations and validation efforts for media experience measures.

References

Helle, M., Ravaja, N., Heikkilä, H., Kallenbach, J., Kankainen, A., Kätsyri, J., Laine, J., Marghescu, D. (2011). *A Theoretical Model of Media Experience and Research Methods for Studying It*, Deliverable D0.4.1/0.4.2, Next Media research program.

Mensonen, A., Aikala, M., Laine, J., Seisto, A. (2012). Consumer perception of printed point-of-purchase displays, Journal of Print and Media Technology Research, 1(2012)4, pp. 205 – 213

O'Brien, H. L., (2010). *The Influence of Hedonic and Utilitarian Motivations on User Engagement: The Case of Online Shopping Experiences*, Interacting with Computers 22, pp. 344-352.

Paper or Digital? A Cross-Cultural Exploration of Students' use of Paper and Pen versus Digital Media in University Environments

Jane Vincent¹, Leopoldina Fortunati²

¹Digital World Research Centre, Faculty of Arts and Human Sciences, University of Surrey, Stag Hill, Guildford, Surrey GU2 7XH

²University of Udine, Via Prasecco 3°, 33170 Pordenone, Italy, Tel. +39-0434-239424, Fax +39-0434-239419

E-mail: j.vincent@surrey.ac.uk, fortunati.deluca@tin.it

Corresponding author: j.vincent@surrey.ac.uk

Short Abstract

This paper investigates the impact of digital media on writing and reading within universities from different countries and it addresses the particular aspect of the student's experiences of paper and pen versus digital media. This cross cultural study is based on the results of individual research conducted in ten countries in two continents by members of the COST Action FP1104, Work Group 1 – Customers and Users. The methodological approach used is qualitative content analysis of surveys originally conducted in the national language of each country with the results translated into English; the same research questions were used in each survey. The principal results show that there are many similarities between the countries studied but that some use pen and paper less whereas others are more prepared to use hand writing, this may link to the availability and use of digital technologies as well as to personal preferences. Reading and writing competencies are changing with the use of digital technologies but students still see benefits of reading and writing with paper which they continue to use, especially to convey private emotions and intimate feelings. This qualitative study provides new learning about the contrasting use of paper and digital media within an educational rather than business setting and on the uses of pen and paper for reading and writing. It provides the basis for further study and a quantitative survey in the same universities has commenced enabling opportunities for new scientific knowledge in this field of research.

Keywords: Writing and Reading; Pen and Paper; Keyboard and Screen; Educational Setting, Digitalization

1. Introduction and background

In this paper we report a cross cultural, transdisciplinary qualitative study comparing and contrasting the use of pen and paper and the use of keyboard and screen in 10 university educational settings. The study was conducted by members of COST Action FP1104 Work Group 1 (WG1) Customers and Users who are exploring the effect of the changing media use habits on traditional media within the context of new possibilities for print and media (see acknowledgements for details of countries and members of the survey team).

Personal and business relationships, communications, and management of everyday life are increasingly mediated via digital technologies and the use of paper books and writing on paper appears to be at risk of declining. Smartphones, tablets, laptops and personal computers are at the heart of contemporary society enabling people to work and communicate in almost any location. We learned from the work of Sellen and Harper in 2002 that, contrary to expectations, the arrival of computers did not result in a paperless office. Their study, based on business users, was conducted over ten years ago (Sellen and Harper, 2002). Our present study not only provides more contemporary research but it asks, for the first time, students in a university environment about their preferences for pen or digital. Do they still have a use for pen and paper or has digital literacy superseded writing by

hand and reading on paper? Studies that research the comparative and contrasting use of paper and pen with digital technology are scant and tend to focus on e-reading versus paper books and on electronic substitutes for paper and pen, rather than also considering handwritten notes and letters (Dillon, 1992: Chen et al, 2013). Furthermore, university students in educational settings have not previously been researched about their use of paper, pen, screen and keyboard providing opportunities for new learning in this area of study.

This present paper thus reports the outcome of an unique study of 562 university students in 10 countries from Europe and Asia in which they were asked about their use of writing and reading with pen and paper and keyboard and screen. Lead by a sociological approach the study includes academic contributions from students in media and communications, chemical and metallurgy, linguistics, psychology, HCI, design, computer sciences and paper science disciplines.

The foundational work for this study involving 24 graduate students was carried out by Leopoldina Fortunati at the University of Udine in Italy (Fortunati and Vincent 2013) and explored the research questions: how do students perceive the affordances of electronic reading/writing when compared to writing/reading with paper? And, have electronic writing and reading become richer experiences than paper writing and reading? These questions draw on theories and concepts regarding electronic writing (Ong, 1986) and media richness theory (Daft & Lengel, 1984). Contrary to expectations that paper and pen may have fallen out of use this cohort of Italian graduate students did, indeed, use this medium, valuing handwritten messages and letters for intimate communications as well as finding it to be integral to their research and learning processes. They were also voracious users of digital media.

2. Methods

The seminal paper by Fortunati and Vincent (2013) and their research questions and methodology formed the basis for a series of qualitative surveys in each of the countries represented in the WG1 survey team. The data from 10 of these surveys is used for the cross-country analysis in this paper. The same research questions were used in each survey conducted in 2013 or 2014 but the data was gathered using some variations in method (essay, questionnaire, online survey). This paper draws on both the published articles by survey team members (Fortunati and Vincent 2013; Taipale, 2014; 2015; Kaputa and Palus, 2014; Lasheva et al, 2013; 2015; Vershinskaya, 2014; Farinosi, Lim & Roll, forthcoming) and the presentation material from the WG1 Meetings 2013 to 2015.

In the surveys university students were asked to freely write about their reflections on four themes articulated in four questions (Vincent 2014). They were not given instructions on how to write about each theme because we were interested in their spontaneous thoughts.

1) Describe the differences you find when using a pen and using the computer. Furthermore describe what you like and dislike about both these modalities.

2) Describe which differences you find in reading paper and reading on a screen. Furthermore, describe what you like and dislike of both these modalities.

3) Think now of the gestures and postures you assume in reading and writing using paper and on a screen. Reflect and describe them.

4) Think now to your use of the computer/internet. This tool allows multimodal communication (images/videos, texts, sounds music and so on) How do you use it? Reflect on your personal experience and then describe it.

Most of the participating countries conducted this survey online (in the original Italian study the questions were answered in handwritten essays). The surveys were conducted in the national language of the respective country and the results translated into English by the country representative. Table 1 shows a breakdown of the respondents by University location (some of the data is missing as this material is not yet published) for the qualitative research. The final column indicates whether a follow up quantitative survey is being conducted.

Country, Lead Researcher	Number Surveyed	Period of Survey	Undergraduate Graduate Students	Gender	Range or average Age	Quantitative Survey Status
Bulgaria Veska Laysheva Chemical Technology & Metallurgy University Sofia	38	April – June 2014	25 UG 13 G	F&M	∞ 19 – 26	Completed January 2015 220 respondent
China Yao Nie Peking University	40	2013	21UG 18G 1Visiting Scholar	F&M	23	
Finland Sakari Taipale Jyvaskyla University	26	Jan – Feb 2013	13 UG 13 G	23 F 3 M	27	Commenced May 2015
Germany Joachim Höflich University of Erfurt	54	June – July 2013	46 UG 8 G	41 F 13M	21.9	To be conducted
Hong Kong China Chung Tai Cheng Hong Kong Polytechnic University	28	June – July 2013	28 UG	14 F 14 M		
Italy Manuela Farinosi University of Udine	129	April 2011 to October 2012	129 UG	52F 77 M	21	To be Conducted 2015
Portugal Pedro Isaias Lisbon	99	2013		F&M		
Russia Olga Vershinskaya Russian Academy of Sciences Moscow	25	Sept – November 2013	25UG	F & M	19-21	To be Conducted 2015
Slovakia Vladislav Kaputa Technical University Zvolen	100	2013	100 UG/G	c75F c25M	21.5	Completed 2015

Table 1: Summary of Qualitative Survey Data Sources from Participating Countries Source: FP1104 WG1 Survey Team (Hungary and Ireland also to conduct Quantitative survey)

UK Chris Lim University of Dundee	23	May 2013 – October 2014	13 UG 10 G	12F 11M	26.9	Commenced May 2015
---	----	-------------------------------	---------------	------------	------	-----------------------

These qualitative studies highlighted some challenges for implementing surveys in multiple countries simultaneously. Although the four core questions are the same the survey was variously set as hand written course work or typed online question and answer survey. The student demographic was consistent but the gender split and courses studied varied. It appears that male students were less willing to contribute to open ended questionnaires online (e.g. Slovakia, Finland), whereas the gender balance in Italy reflected the student cohort completing the survey as part of their coursework. Additionally there are nuances not explored: for example, differences between reading printed and hand written text; between reading a paper or e-book and the size of the screen. Another limitation was this study was not funded and thus had to be incorporated into the everyday work of survey team members. Nevertheless a set of rich qualitative material was obtained and has formed the basis for further discussion and finalising of the quantitative questionnaire that is currently being used to obtain more statistically valid data in at least 6 countries (Bulgaria (Lasheva et al, 2015) and Slovakia have already completed this survey).

3. Results and Discussion

There is already considerable debate on digital literacy and digital natives (Helsper and Eynon 2010) and regardless of their competences university students are required to study within a digital learning environment, and their institution is expected to provide and respond to their changing digital needs. Nevertheless, student's use of paper books, pen and paper, and handwritten note-taking prevails. In this section we set out the results from the qualitative surveys that illustrate the differences and similarities, and the correlations between the student's experiences of writing and reading with pen, paper, keyboard and screen; we use quotes from students to explicate the findings (Vincent 2015).

There was consensus of views and responses across all the countries covering a variety of topics relating to the functionality and status of digital technologies in society and old and new ways of doing things, for example the aesthetic of the chosen medium is a deciding factor for many, and pen, paper and books are treated with affection and nostalgia.

Handwriting is slower and impractical, but at the same time more personal and enjoyable. Writing with a pen also relates to the joy of chirography. My chirography is unique and it often catches other people's attention. I feel that it says something about my persona and perhaps because of this I want to cherish handwriting. (Finland)

The taste of browsing something material is priceless! I like very much to enjoy the scent of the book through the fragrance of the paper. (Italy)

Writing by hand is much more personal, more subjective. This allows us to bring up all of our emotions: happiness, sadness, nervousness. Based on the features of the texts we can understand how the writers felt at the time of writing. (Italy)

I use paper to write on when I have to write important messages, with passion, because in my opinion, your own handwriting makes the message very personal. (Italy)

I am so used to hypertexts that I miss this application when reading texts on paper. (Germany)

The text is easier to edit on a computer afterwards, so it is more likely that all ideas will come out, while when writing on paper you sort of have to manage with what comes to your mind at that particular moment, and thus the outcome is easily unclear and inconsistent. (Finland)

I used to write essays by hand at high school, it would annoy me how much paper and time we would waste having to re-write the same essay over and over again until there were no mistakes and it looked neat. (UK)

There was recognition that computer mediation can be a distraction as it can fragment thinking and there are problems when articulating mathematical and scientific theory and formulae on computers – paper is essential. It was noted in Hong Kong most respondents emphasized that reading and writing using paper encourages them to treat these as a complementary behaviour, whereas using a screen makes them experience reading and writing as separate units. In Portugal students underlined the difference between the formality of the computer and the flexibility and informality of the paper and in Russia age matters. The hypothesis is that the younger the student is the less accustomed to handwriting and to reading books he/she is: *Using a pen becomes more and more obsolete*. *I learned to print quicker than to write*. (Russia)

These, together with issues regarding electricity consumption for sustaining digital technologies present some challenges and opportunities for future research. It would appear that students will continue to use, adapt and shape the technology most appropriate to their needs be it paper, pen, keyboard, screen, e-book or paper book but they still have a demonstrable need for all to be available for their use in the University setting.

E-books are sometimes necessary in modern life, they became affordable. You can find a digital copy of a rare or very old book. So I use both forms. (Russia)

Paper and pencil are always available, and do not need electricity; such as when having a sudden inspiration in bed. (Germany)

Until last year I was mostly a paper reader but I slightly evolved so that now I think I'm equally competent as a paper and online reader. I used to print articles if they were longer than 10 pages but nowadays I manage to do that online. I got used to it but I also got this application to write notes on a PDF file so that I can act on online documents as much as I would do on paper. (Finland)

Further findings from our qualitative research show that it is not simply a matter of being digitally literate, and 'doing things digitally' but there are sometimes special conditions that mean digital technologies do not work. 'Chinese characters are not just an information carrier, but also a culture carrier. Aesthetically, calligraphy is more concerning handwriting itself than the concrete information the characters bear' (Yao Nie, WG1 2013).

It is easy to write lectures on the computer but to write down formulas it is much easier to use a pen. (Russia)

Graphs or complex formulas are not easy to input while writing on screen. (China, Peking)

I use white space and the positioning and grouping of notes on a page to aid understanding. This is a rapid process which cannot be achieved on screen. (UK)

If you get prepared for an exam, no information is left in your head if you use a keyboard. Writing with a pen allows you to remember what you write. (Russia)

What I like most about a pen, is that it is quick to add notes, comments, and all kind of drawings in the text and on the side. For example, for me drawing arrows is easiest by hand. (Finland)

On the other hand, a great thing in pen writing is a possibility to write on different parts of a paper sheet, in different directions and with different coloured pens, use text of different size, and insert this and that here and there. Making a mind map with a computer is, in principle, much more difficult, and requires efforts to a different extent. (Finland)

But there are other situations when only digital technologies will do. In Bulgaria all respondents reported that they extensively use computers in everyday life, learning and work, and cannot imagine life without them (Lasheva et al 2013)

Internet simplifies the communication: my husband (professional soldier) took part in missions in Afghanistan a couple of times so I cannot imagine my existence without having contact with him for some months. (Slovakia)

Whilst the respondents are extensive users of digital technologies, using pen and paper for writing and reading as well as, and combined with, digital technologies remains part of the normative practices of the university students surveyed. Motivations for using paper and pen are influenced by the haptic qualities of writing - the feel and the smell of the paper and the grasp of the pen, and extend also to the practical usefulness of note taking and writing in margins while reading. Conversely the use of hypertext and automatic error correction in online writing are making the use of keyboard and screen more compelling. Issues of multi-tasking, chirographic skills, intimacy of paper versus digital, interleaving of using digital and printed text as well as problems of eye strain and posture were identified.

4. Conclusions

There is no doubt that students have embraced the use of digital technologies in the educational setting of their university with alacrity but they have also found that the affordances of chirographic writing and the use of paper have special qualities that cannot be matched by digital media.

The new learning from this study, which is the first to examine the topic in the educational setting of universities, has benefits for the academic and pedagogic communities some of whom place strong emphasis on digital literacy and less on the quality of handwriting skills. The normative practices of students show that there is still a demand for pen and paper as well as keyboard and screen and that in some instances the use of paper is preferred.

There are many new questions raised by our study and further research is required to explore the topic in more detail and with greater statistical validity. WG1 Survey Team has designed a new questionnaire based on the findings from this present study, informed also by the work of Naomi Baron (Baron, 2000; 2015). This quantitative survey enables further examination of the normative reading and writing practices of students and early indications from these surveys appear to support the conclusions drawn from qualitative research. The statistical evidence from these surveys will be published in the final report of the COST Action FP1104 WG1 Survey Team on completion of the Action.

Acknowledgments

COST Action FP1104 Work Group 1 Survey Team Group members:

Germany: Joachim Höflich, Julia Roll; Russia: Olga Vershinskaya; Finland: Sakari Taipale; Italy: Leopoldina Fortunati, Manuela Farinosi; Slovakia: Vladislav Kaputa, Hubert Palus; Hungary: Rozalia Szentgyorgyvoilgyi; Bulgaria: Veska Lasheva, Hristina Blazheva; Hong Kong: Chung Tai Cheng; China: Yao Nie; Portugal: Pedro Isaias; Ireland: John O'Sullivan; UK: Jane Vincent, Chris Lim. Naomi Baron American University Washington D.C.

COST Action FP1104 and University of Udine for funding and hosting the Short Term Scientific Mission in March 2015 in which this study was completed.

References

Baron, N.S. 2000. Alphabet to E-mail: How Written English Evolved and Where It's Heading Routledge, London and New York.

Baron, N.S. 2015. Words Onscreen. The fate of reading in a digital world Oxford University Press

Chen, N. Guimbretière, N. and Sellen, A. 2013. Graduate Student Use of a Multi-Slate Reading System *CHI* 2013 April 27-May 2, 2013, Paris, France

Daft, R.L. Lengel, R.H. 1984. Information richness: a new approach to managerial behavior and organizational design. *Research in organizational behavior* (Homewood, IL: JAI Press) 6, pp191–233.

Dillon, A. 1992 Reading from paper versus screens: a critical review of empirical literature. *Ergonomics* 35(10), pp1297-1326.

Farinosi, M., Lim, C, and Roll, J. Book or Screen, Pen or Keyboard? A Cross-Cultural Sociological Analysis of Writing and Reading Habits Basing on Germany, Italy and the UK (in review)

Fortunati L., Vincent J. 2013. Sociological Insights into writing/reading on paper and writing/reading digitally in *Telematics and Informatics* 31(1), pp39–51 doi:10.1016/j.tele.2013.02.005

Helsper, E., Eynon, R. 2010. Digital natives: where is the evidence? *British Educational Research Journal*, 36 (3), pp. 503-520. ISSN 0141-1926

Kaputa V., Palus H. 2013. Users and attitudes towards print and digital Intercathedra 29(2) 28-34

Lasheva V., Blazheva, A., and Lasheva, H. 2013. Comparative Qualitative Analysis of the Use of Paper and Computer in *Journal of Food and Packaging Science, Technique and Technologies* 5 (Bulgaria)

Lasheva V., Blazheva, H., and Lasheva, H. 2015. *Comparative Quantitative Analysis of the Use of Paper and Computer* University of Chemical Technology and Metallurgy Sofia, Report for Cost Action FP1104 WG1

Ong, W. 1986. *Oralità e scrittura. Tecnologie della parola*. Bologna: Il Mulino (original edition Orality and Literacy. The Technologizing of the Word. Methuen, London, 1982).

Sellen, A., Harper, R., 2002. The Myth of the Paperless Office, MIT Press, Cambridge.

Taipale S., 2014. The affordances of reading/writing on paper and digitally in Finland in *Telematics and Informatics* 31(4), 532–542.

Taipale S. 2015 Bodily dimensions of reading and writing practices on paper and digitally in *Telematics and Informatics* 32(4), 766-775.

Vershinskaya, O. 2014. *Reading and Writing on Paper and on the Screen. Russian Case* Report from Russian Academy of Sciences for COST Action FP1104 WG1

Vincent J. 2014. Writing/Reading on Paper and Writing/Reading Digitally Presentation at COST Action FP1104 Plenary Ljubljana 4 November 2013

Vincent, J. 2015. Writing and reading using pen, paper, keyboard and screen -a cross cultural exploration Presentation at COST Action FP1104 Plenary Zagreb 27 March 2015

How Today's Audience is searching for Contact with traditional Media via Facebook

Liis Velsker¹, Ragne Kõuts²

¹Postimees, Estonia ²University of Tartu, Estonia

Email: liiswelsker@gmail.com, ragne.kouts@ut.ee

Corresponding author: ragne.kouts@ut.ee

Short Abstract

This study examines what motivates today's audience to search for contact with traditional media outlets – newspapers, radio & television channels – via social networks by looking at both individual media repertoires and the different rationales behind news consumption and liking news sites / publications on Facebook. All data related to news consumption preferences and Facebook usage habits was collected during three cycles of interviews involving individuals from different age groups in Estonia.

The study found that multiple media repertoires can be identified in every age bracket. In one group, FB is considered the only news source that addresses public and state issues and life around the world – making the role of pre-selection determined by FB-algorithms and friend networks a decisive factor. Other groups have contact with other media outlets via FB as well. The results indicated that 77% of students and 40% of adults had been looking for contact with at least one of Estonia's traditional media outlets via Facebook. The survey mapped 12 groups with different motivations for liking a media outlet on Facebook.

Keywords: traditional media, Facebook, news, user profiles

1. The Objectives of Research

Social media has become an integral part of life for the vast majority of people. Moreover, many individuals are already related to society only via social networks (see Westerman et al 2014, Fortunati et al 2014).

A number of reports by the Reuters Institute indicate that more and more often consumers access news content through social media (Newman/ Levy 2013; Newman/ Levy 2014). Clear boundaries between different mediums have become blurred – television, radio and newspapers have moved into the realm of the Internet and are increasingly consumed on digital platforms rather than by means of the technological device for which they were initially designed. The human experience of different forms of media is changing – some watch television on their iPad or smartphone more often than they do on a TV, some consume news from a print media's web edition through videos instead of plain text. Given this status quo, it becomes necessary to look at how the audience combines different media channels and how social media affects their news consumption.

The study relies on the assumption that the use of mass media is a rational and motivated behaviour that serves a certain function for or profits the individual (Katz et al 1974). At the present time, however, the uses and gratification theory is considered to be the most comprehensive, empirically grounded approach to examining media use in the context of the new media environment (Hasebrink/Domeyer 2012). Nevertheless, we will go further, following the Sullivan's belief that "for a deeper analysis of the audience one needs to start with the daily experience" (Sullivan 2013). In fact,

many additional factors influence users' choices and selections when they consume news content. Courtois et al (2015) summarised that in order to become a part of an individual's news diet, a news medium must 1) be worth the time spent, 2) be acceptable to peer networks, 3) maintain a public connection, 4) have participatory potential, 5) be affordable in terms of price, 6) have technological appeal, and 7) offer a situational fit (Courtois *et al.*, 2015: 125). As far as technology is concerned, the internet has given people the opportunity to be in contact with both various traditional media channels as well as other audience members. Connectivity is the keyword that best encompasses social networks and the internet (Chayko 2002). More and more often, news is consumed via the internet (Newman/Levy 2013; Newman/ Levy 2014). Social networks have extended the 'free culture' and this has enabled the distribution of free information to a far greater degree (Cingel *et al.* 2014; Jansson/ Lindell 2015).

Hermida (et al 2012) found that Facebook and Twitter have become sites where Canadian people find a large portion of their daily news. A significant part of social network users use their personal friend network to filter the amount of information in their news feed instead of trusting the choices made by professional journalists or editors (Hermida et al 2012). Digital media, such as blogs, homepages and social networks have given each user the classic role of a gatekeeper usually attributed to a professional journalist (Schweisberger et al 2014, Westerman et al 2014), while Burns (2008) renamed the role of the gatekeeper as the gatewatcher.

The research reported in this paper aimed to find out how actively journalistic content is consumed and shared via Facebook. First of all, it was important to map out which groups of media users have intense contact with traditional media outlets, or established journalistic "brands", via Facebook. At this stage, the study looked at clarifying what motivates users to like, share and recommend different news products published on FB, asking how the consumption of traditional media in traditional ways relates to liking the same channels on Facebook and whether consumers would rather approach the media channel or the intermediary. It is intriguing to see that if FB as a social medium fosters bilateral relationships, how actively users comment on journalistic product published by the intermediary. In addition, the flow of information in FB news feeds is shaped by each user's personal network of friends. Thus, it is worth examining how so-called friends in the online environment affect the consumption of journalistic products and whether a person prefers to watch news shared by a friend or trusts more in content shared by a media brand?

2. Research Methods

This study followed the idea of media repertoires (Hasebrink/Domeyer 2012) in combining qualitative and quantitative research methodologies, with the qualitative part of the study answering the whyquestions behind the phenomena in hand. In fact, the fields of social studies and humanities now afford much greater attention to the combination of qualitative and quantitative research methods (Creswell 2003), and it follows that the concept of simultaneity played an important role in this paper as the information gathered during the interviews contributed to both quantitative and qualitative analyses.

The study is based on the assumption that macro-level institutional structures have an impact on micro-audience practices (Peruško *et al.* 2015). On the first level, the media system available via social networks was documented in Estonia. Only 4 local newspapers, 3 magazines and one TV station from those observed did not have a Facebook account – FB is the largest social network in Estonia (Metrix.Station 2015). According to the number of followers on Facebook, the top five include: the national daily newspaper *Postimees* (103 197); the *Sky Plus* (hot AC format) (83 494) and *Power Hit Radio* (dance CHR/ TOP 40) (47 584) radio stations; the national tabloid newspaper *Õhtuleht* (43 523); and the daily news TV-magazine produced by the Estonian Public Broadcasting *ETV* (38 644) (Nov 17th 2014).

Next, data for the study was gathered through 72 semi-structured interviews with 52 adolescents and 20 adults, all of whom were Facebook users. The selection consisted of secondary school students

aged between 16 and 19 years of age and middle-aged adults whose ages averaged at 39. One additional selection was made during the following step – the internet news consumption habits in a group between 20 and 34 years of age were studied through 40 additional interviews. In the last group, the specific focus centred on the question of what respondents considered as news in the online environment. As indicated by Yadamsuren and Erdelez (2012), due of the multiplicity of news platforms and diverse internet offers, people's perception of news is quite broad, and there is even a group for whom "all of the Internet" can be considered as news (Yadamsuren and Erdelez 2012). Seen in this light, it is without doubt that changes in news consumption practices have consequences in relation to democracy and social cohesion.

As background information, the study revealed that all three groups live and act in similar technological and media environments: most of them have at least one printed newspaper delivered to their homes; can watch different TV stations via traditional devices as well as on the internet; and have at least one mobile communication technology device employed for personal use (a laptop with an internet connection, a smart phone or tablet). Their preferences in relation to the usage of all of these media channels proved to be quite different. The youngsters (16-19 y.o.a) watched more commercial TV stations and their television viewing was characterised by a large amount of time spent on Internet-based TV services. The adults preferred to watch the Estonian Public Broadcasting TV channel *ETV* and were not so keen on watching television on the Internet. The "in-between" group (20-34 y.o.a) had less time for TV services and preferred information that can be accessed more quickly and in shorter forms.

In talking about the respondents' usual "media day" and mapping their usage of internet sites and Facebook, their news consumption repertoires and reasons for accessing different internet websites were identified.

3. Findings

This study indicated that the number of students who sought contact with at least one of Estonia's traditional media outlets was significantly high – with 40 respondents (77%). Amongst adults, the proportion of followers was somewhat more modest – with only 8 respondents (40%).

Amongst the internet users studied, the middle age bracket revealed a group with a highly selective media repertoire oriented only to the transnational content and foreign information – this group was very critical of the content of Estonian news providers and preferred news from the *BBC*, *CNN*, or from the biggest English-language newspapers like *The New York Times* or *The Guardian* (Sukk 2015). These are the so-called 'global citizens' who's life-worlds are not limited to national borders or the concepts of a single nation state. In their Facebook news feed the Estonian providers appear as well, but do not appear to be highly valued.

If we compare users "liking" of the different media providers on Facebook, we can summarise that all age groups were mostly looking for a contact with print media – national daily newspapers had the highest average number of followers and they posted to Facebook most frequently. Young people liked traditional media outlets on Facebook more actively and their choice of media outlets was more diverse.

In 13 incidences, a student had looked for a contact with the same newspaper or magazine that was delivered to his home on FB, while this incidence was only three in the adult group. Thus, we can say that reading (or ordering) print media does not relate to liking the same media outlet on FB. On the other hand, it seemed that students mostly only liked the same radio station that they listened to on a regular basis – *Sky Plus* (18 students liked this radio station) on Facebook.

Hasebrink and Domeyer (2012) emphasized that it is important to examine whether users choose a particular brand or social media context, and in this research, as is the case in all different traditional media channels (except magazines), as well as online-media, one popular media outlet/brand stood out

by far – either *Postimees* as a newspaper, *Sky Plus* as a radio station, or *Delfi* as a web portal etc. The study showed that people might look for contact with a specific television production (shows, programs) more than with the TV channel itself, e.g., *ETV*.

However, the study indicates that routines still play a really important role in everyday media usage, including that of Facebook – the respondents do not seek and are not really open to trying out new providers' content. If they have already got some news providers on their FB news feed, they are not eager to like additional news sites.

The main reason why some of the respondents did not like any traditional media channels on Facebook was the desire/need to reduce the information overload and limit the flow of messages. In all age groups we found one specific news usage repertoire – people who get news exclusively or mostly via Facebook. They trust the choices of their friends / FB-contacts more than those of journalists or newsrooms (Härma 2015).

3.1 Reasons behind the phenomena

The survey mapped out 12 groups that are motivated to like certain media outlets on Facebook for different reasons. In broad terms, these motivations can be divided into two categories: those that arise from a need for information (1-8) and those that are based on an interest in the media channel (program, editorial) itself rather than its production (9-12).

The motivations for liking were categorised into 12 groups: (1) "the rapid acquisition of information"; (2) "keeping up to date"; (3) "geographic proximity"; (4) "psychological closeness"; (5) "obligation"; (6) "interest in the topic"; (7) "entertainment and gossip"; (8) "social utility"; (9) "media habits and brand trust"; (10) "being a fan"; (11) "need for a reminder"; (12) "impact of marketing and advertisements".

Media consumption via Facebook is greatly affected by friend networks. Users prefer to open media content that is shared by a friend rather that that of professional media outlets and traditional media channels. The closer the person is in a "real life" the more likely it is that the user will click onto the post and respond to it with comments or likes. The triggers that explain why Facebook users clicked on a specific post and decided to open it matched largely with traditional news value criteria.

Still, the study showed that Facebook was the primary news source for only a few members of the audience and that they preferred online news portals, TV channels and newspapers. Users tend not to visit the pages of media outlets on Facebook because they want more information showed on their news feed. The interviewees preferred not to comment on Facebook posts through a traditional media outlet, rather, they were more likely to do this on Facebook than on the website of a traditional media outlet.

However, if a user opened media content via Facebook he/she often stayed on this new web page, showing that Facebook is indeed a place where media channels can increase their audience. Some interviewees valued Facebook as a place where, thanks to their friends, they can reach media outlets that they would otherwise not consume.

4. Conclusions

Facebook, as a social network, plays an important role in Hasebrink and Domeyer's (2012) concept of media repertoire for different age groups. One can say that for a considerable number of people Facebook has not yet become a distinct media "channel", or at least, is not yet a primary news source as the Reuters Institute suggests (Newman/Levy 2013). Still, it can be said that for many people social networks provide an environment through which they can interface with society, their own community and partly with their media space.

Young people who spend more time on the Internet and social media are already searching actively for contact with various traditional media channels via Facebook. The adults' activity on FB has remained more modest, but it is clear that they are also making noticeable changes to their regular media consumption, e.g., reading news online etc. Almost all of the students interviewed visited Facebook on their mobile phone and over half of them are constantly logged in to Facebook, receiving messages and information via social network 24/7, so it is clear that the potential of media consumption through Facebook cannot be underestimated – FB can become an important intermediary for news.

The findings showed that traditional media consumption in the traditional way is not necessarily related to looking for contact with the same channel on Facebook. If we talk about fragmentation of media usage and about personal media "menus", the majority of readers are united by the main news providers that already belong to their consumption routine, while new providers have very few possibilities to take up a position in their news menu. In this case, the role of Facebook, and thus the role of friends on social networks, is decisive.

This said, one still finds individuals in all of the age groups who only access online news content via Facebook or avoid journalistic content completely. This definitely poses questions in relation to a new basis for the general public sphere and the broadening possibilities for the active function of democracy.

References

Chayko, M. (2002). *Connecting. How We Form Social Bonds and Communities in the Internet Age*. New York: State University of New York.

Cingel, D. P., Lauricella, A. R., Wartella, E., & Conway, A. (2014). Predicting Social Networking Site Use and Online Communication Practices among Adolescents: The Role of Access and Device Ownership. *Media and Communication*, 2(2), 1-30.

Courtois, C., Schrøder, K.C., Kobbernagel, C. (2015). Exploring Landscapes of News Consumption Cross-Nationally, in Zeller F, Ponte C and O'Neill B (eds) *Revitalising Audience Research: Innovations in European Audience Research*. New York, London: Routledge, 123-142.

Creswell, J. W. (2003). *Research Design: Qualitative, Quantitative, and Mixed Methods Approaches*. Thousand Oaks [etc.]: Sage.

Fortunati L, Deuze M, Luca F. (2014). The New About News: How Print, Online, Free, and Mobile Coconstruct New Audiences in Italy, France, Spain, the UK, and Germany. *Journal of Computer-Medicated Communication*, 19:(2), 121-140.

Hasebrink U, Domeyer H. (2012). Media Repertoires as Patterns of Behaviour and as Meaningful Practices. A Multimethod Approach to Media Use in Converging Media Environments. *Participations*, 9:(2), 757-779.

Hermida A, Fletcher F, Korell D, Logan D. (2012). SHARE, LIKE, RECOMMEND. *Journalism Studies*, 13:(5-6), 815-824.

Härma, K (2015) Uudiste lugemine internetis Noorte ja nooremate keskealiste hulgas [Online news consumption by young adults and early middle aged". The Master Thesis, Tartu: University of Tartu.

Jansson, A., Lindell, J. (2015). News Media Consumption in the Transmedia Age, *Journalism Studies*, 16:1, 79-96.

Katz E, Blumler J. G, Gurevich M. (1974). Utilization of Mass Communication by the Individual. Blumler, J. G. & E. Katz (toim). *The Uses of Mass Communications: Current Perspectives on Gratification Research*. Beverly Hills; London: Sage, 19-31.

Metrix.Station. Facebook Fan Pages, URL (used in April 2015) http://metrix.station.ee/?act=facebook

Newman N, Levy D. A. L. (2013). *Reuters Institute Digital News Report. Tracking the Future of News*. Oxford: University of Oxford.

Newman N, Levy D. A. L. (2014). *Reuters Institute Digital News Report. Tracking the Future of News*. Oxford: University of Oxford.

Schweisberger V, Billinson J, Chock T. M. (2014). Facebook, the Third-Person Effect, and the Differential Impact Hypothesis. *Journal of Computer-Medicated Communication*, 19:(2), 403-413.

Sukk, M. (2015) Meediarepertuaari uurimise võimalusi Q-meetodil [Q-Methodology in Researching Media Repertoires]. The Bachelor Thesis, Tartu: University of Tartu.

Sullivan, J (2013) Media audiences. Effects, Users, Institutins, and Power. London: Sage.

Westerman D, Spence P. R, Heide B. (2014). Social Media as Information Source: Recency of Updates and Credibility of Information. *Journal of Computer-Mediated Communication*, 19:(2), 171-183.

Index of authors

Alabami Hanna Laana	11	Miettinen, Päivi	275
Alakomi, Hanna-Leena	11	Miranda, Paula	33
Antonucci, Francesca	11	Moginov, Rostislav	89
Dauhanalama Anadina	17	Muck, Tadeja	33
Barbonelova, Angelina	17	intucii, radeja	55
Bauer, Wolfgang	53	Neumann, Jann	125
Beckert, Erik	99		
Bezzi, Marco	5	Nikolaus, Ulrich Novotny, Erwahat	207, 213
Bollström, Roger	107, 255	Novotny, Erzsébet	239
Bousfield, Douglas W.	175	Nurmi, Olli	231
Cl I'	105	Pabst, Oliver	99
Chen, Jian	125	Pallottino, Federico	5
Colle, Giacomo	5		
Costa, Corrado	5	Pathak, Shreyas	195
		Peiponen, Kai-Erik Pekarovicova, Alexandra	43 195
Dauer, Michael	53	r ekalovicova, mexandia	195
Desai, Saunil	223		4.47
Dörsam, Edgar	125	Radermacher, Kirsten	147
Držková, Markéta	33	Rassek, Patrick	183
		Reboun, Jan	175
Eberhardt, Ramona	99	Reif, Maximilian	99
Ermoshina, M.	157	Ridgway, Cathy	43
,	107	Rogelj, Nina	247
Fedorovskaya, Elena	223		
Figorilli, Simone	5	Sauer, Hans Martin	125
Fischer, Thomas	115	Scarascia, Giuseppe	5
Fleming, Paul D.	195	Schär, Manfred	265
Fortunati, Leopoldina	305	Schöffler, Peter	77
	53	Scrinzi, Gianfranco	5
Fuchs, Wolfgang	55	Seisto, Anu	33, 167
Come Detail A C	42 (F 107 2FF	Selbmann, Karl-Heinz	265
Gane, Patrick A.C.	43, 65, 107, 255	Silfsten, Pertti	43
Gauttier, Stéphanie	287	Sipiläinen-Malm, Thea	11, 167
Gauzente, Claire	33, 287	Smolander, Maria	,
Golob, Gorazd	265		167
Götz, Christin	207	Spieß, Janine	213
Gref, Frank	275	Sprinzing, Thomas	77
		Sundquist, Mika	275
Hakola, Liisa	11	Syrová, Lucie	175
Heinonen, Jaana	275	Syrový, Tomáš	175
Hirn, Ulrich	53	Szentgyörgyvölgyi, Rozália	33
Horváth, Csaba	239		
Hübner, Gunter	115, 183	Tåg, Carl-Mikael	43
,	,	Thorman, Sofia	139
Isaías, Pedro	33	Tiekstra, Sanne	33
Isulus, i certo	55	Toivakka, Martti	175, 275
Jung, Ulrich	147	Tünnermann, Andreas	<u></u>
Jutila, Eveliina	107, 255	,	
Jutila, Evenina	107, 233	Vehmas, Kaisa	33, 167
Karlović, Igor	33	Velsker, Liis	313
Kisters, Dirk	65	Vincent, Jane	305
		Vorozhtsov, Alexander	89
Klanjšek Gunde, Marta	247	vorožinišov, mexander	
Klemetti, Aarne	17	Wang, Yujue	223
Knauber, Henrik	77	Weichmann, Armin	77
Koivunen, Risto	107, 255		115, 183
Kõuts, Ragne	313	Willfahrt, Andreas	·
Krebs, Martin	183	Wolf, Dietmar	65
Krivec, Tjaša	265	$\mathbf{V} = C1 + 1^{\prime}$	4
Kumar, Vinay	175	Xu, Chunlin	175
Kuznetsov, Ý.	157	XZ I'	4.00
		Yang, Li	139
Laine, Janne S.	295	Yin, Jiayuan	17
Lumby, Natalia	25		
Majumdar, Himadri	167		
Marzinkowski, Joachim M.	147		
Menesatti, Paolo	5		