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A method to compensate fluorescence induced white point differences in proof-processes by printing liquid fluorescent brightening agents using inkjet

Daniel Bohn[1], Michael Dattner[2], Stefan Fehmer[3], Peter Urban[1]

[1] University of Wuppertal,
Faculty for Electrical Engineering,
Information Technology and Media Technology,
42119 Wuppertal, Germany

[2] BST eltromat Int. GmbH,
Heidsieker Heide 53, 33739 Bielefeld, Germany

[3] Laudert GmbH & Co. KG,
Von-Braun-Straße 8, 48691 Vreden, Germany

E-mails: Dbohn@uni-wuppertal.de
Michael.dattner@bst-international.com
S.Fehmer@laudert.de
Purban@uni-wuppertal.de

Abstract

One of the key goals in producing paper and cardboard in the print industry is to achieve a high whiteness degree. This is usually realized by fluorophores called OBAs (Optical Brightening Agents) or FBAs (Fluorescent Brightening Agents). The heavy use of FBAs in production substrates, while proof substrates contain a varying amount of FBAs, results into serious difficulties in any color management process, especially in terms of a white point correction.
In this study, an alternative procedure is presented to achieve an illumination independent colorimetric correlation and a visual match between most proof- and production substrates. This is achieved by printing defined amounts of liquid FBA using inkjet with variable area coverage.

Keywords: OBA, color management, FBA compensation, OBA compensation, carrier

1. Introduction

One of the key goals in producing paper and cardboard in the print industry is to achieve a high whiteness degree. A high whiteness level not only imparts the impression of cleanliness and worth, but also leads to a large printable contrast of the substrate and the print (Blum et al., 2002, p. 3). Therefore, paper producers make great efforts to increase the level of whiteness of papers. Already, in the production of raw materials, be that pulp, mechanical pulp or pigment, great efforts are done to obtain raw materials with a very high whiteness level. In the actual paper production, additives and auxiliary materials are avoided which could jeopardize the whiteness of the substrate (Blum et al., 2002, p. 3).

Despite all that, fiber based substrates usually show an increase of absorption in the blue region of the electromagnetic spectrum between 380 nm and 450 nm. The main reason for this spectral behavior is the absorption of typical fiber based substrate components such as lignin.

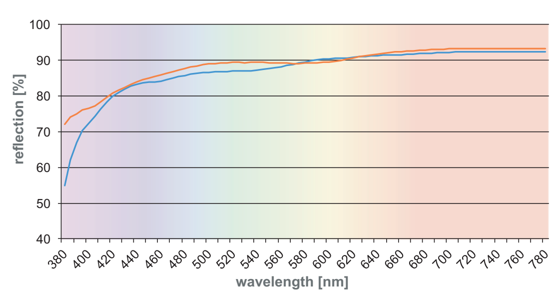

Figure 1: Reflectance of bleached and semi-bleached pulp

Figure 1 shows the spectral response of a bleached and a semi-bleached pulp. The spectral response of pulp is lower in the short wavelength range than in the long wavelength region. Therefore, paper looks yellowish. By bleaching the pulp, the yellow tint is reduced to some extent but not fully eliminated (Bieber et al., 1989, p. 18). To overcome this yellowish tint, several additives (fillers) are available and used by the paper industry to improve the whiteness level, such as titan dioxide (TiO_2) or calcium carbonate ($CaCO_3$) (Nelson, 2007, p. xix). Bieber demonstrates that papers loaded with expensive fillers like TiO_2 show an overall decrease of absorption for each wavelength but they still cannot fully compensate for the yellowish tint (Bieber et al., 1989, p. 19).

1.1 Fluorescent Brightening Agents in general

A very cheap, and therefore, commonly used additives to overcome the yellowish tint are so called Fluorescent Brightening Agents (FBAs) – fluorophore substances that show fluorescence effects (Ai, 2015, p. 3). They are excited in the invisible UV range as well as partially in the blue region (250 nm to 425 nm) of the electromagnetic spectrum and emit mainly "blue light" in the visible spectral range between 410 nm and 550 nm.

More generally, fluorescence is the result of a three-stage process that occurs in certain molecules called fluorophores or fluorescent dyes. A fluorescent probe is a fluorophore designed to respond to a specific stimulus. The process responsible for the fluorescence of fluorescent probes and other fluorophores is illustrated by a Jablonski diagram and can be divided into three stages (Ai, 2015, p. 3):

Stage 1:
A photon is supplied by an external source such as an incandescent lamp and is absorbed by a fluorophore, creating an excited electronic singlet state (S_1').
Stage 2:
The excited state exists for a finite time (typically 1–10 nanoseconds). During this time, the fluorophore undergoes conformational changes and is also subject to a multitude of possible interactions with its molecular environment. These processes have two important consequences. First, the energy of S_1' is partially dissipated, yielding a relaxed singlet excited state (S_1) from which fluorescence emission originates. Second, not all the molecules initially excited by absorption (Stage 1) return to the ground state (S_0) by fluorescence emission. The fluorescence quantum yield is the ratio of the number of fluorescence photons emitted (Stage 3) to the number of photons absorbed (Stage 1).
Stage 3:
A photon of energy is emitted, returning the fluorophore to its ground state S_0. Due to energy dissipation during the excited state lifetime, the energy of this photon is lower, and therefore of longer wavelength, than the excitation photon.

1.1.1 Fluorescent Brightening Agent types

Widely used FBAs are 1,3,5-triazinyl-derivates of 4,4'-diaminostilbene with the ability to carry additional sulfonic acid groups (Blum, Linhart and Frenzel, 2002, p. 2). Next to such "classical" FBAs, newer types such as derivates of 4,4'-distyrylbiphenyl are known (Blum et al., 2002, p. 3). Figure 2 shows a general chemical formation of a diaminostilbene molecule.

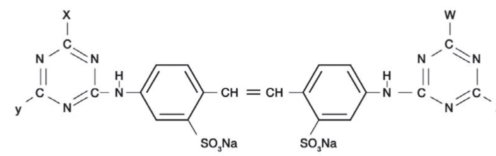

Figure 2: A typical chemical structure of diaminostilbene (Brandt, 2008, p. 7)

Mainly three types of optical brighteners or FBAs are used by the paper industry, all based on the mentioned diaminostilbene molecule shown in Figure 2. The main difference originates from the number of solubilizing sulfonic acid groups (Blum, Linhart and Frenzel, 2002, p. 3; Holik, 2006):

- The diaminostilbene-disulfonated FBA has two sulfonic groups; the two other substituents could be hydrophilic groups. This FBA has a very good affinity to cellulose, but limited solubility and is mostly used in the wet-end.
- The most commonly used FBA is the tetrasulfonated type. The tetrasulfonated FBA is a versatile substance because of its medium affinity and good solubility. It can be used in most stages of papermaking: wet-end, size-press and coating.
- The hexasulfonated FBAs are especially used in coatings where high brightness is required.

1.1.2 Fluorescent Brightening Agent concentration

The intensity of the fluorescent emission of FBAs highly depends on the FBAs concentration in the substrate coating and/or the pulp (Bieber et al., 1989, pp. 22–23). Generally, a higher FBA concentration results in a more intense emission. Figure 3 shows four laboratory substrates that only differ in their FBA concentration.

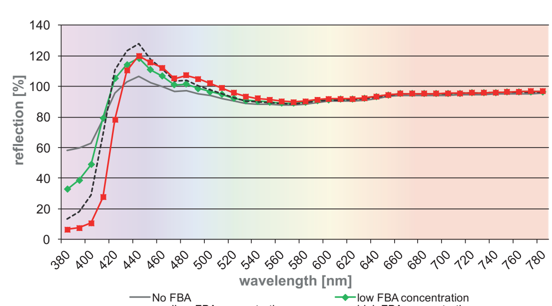

Figure 3: Influence of the FBA concentration on the emission, under M1 measurement condition

However, the FBA concentration cannot be increased infinitely. At a certain point, a so called "greening effect" can be observed (Bieber et al., 1989, p. 40). This effect results because FBAs need sufficiently enough OH binding groups to develop a stable blue emission (Blum, Linhart and Frenzel, 2002, p. 2). As a result, the emission between 460 nm and 550 nm is still increasing, while the main emission (peak emission) at 450 nm stagnates or even drops ('medium FBA concentration' vs. 'high FBA concentration' in Figure 3). Hence, the original blue emission shows a greenish tint. Please note that the substrates offer a limited amount of OH binding groups (cf. chapter 1.1.4).

1.1.3 Available ultraviolet radiation

Dattner, Bohn and Urban (2011) showed that the relative spectral power distribution (RSPD) in the UV range indicates the intensity of the FBA emission in the blue-range of the electromagnetic spectrum. That means that a high intensity of UV radiation within the RSPD leads to an intensive emission and vice versa.

Figure 4 shows the spectral response of a brightened substrate for several UV radiation levels.

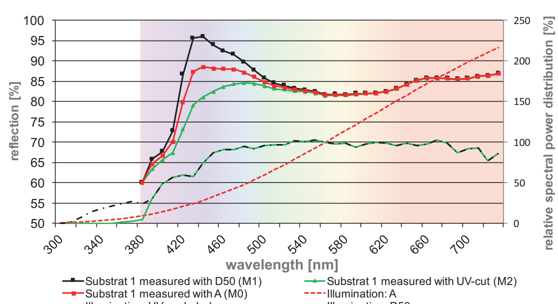

Figure 4: Influence of available UV radiation on the FBA emission concerning measurement condition M1, M2 and M0

The differences and the individual influences of the illuminant 'UV excluded' (M2; no radiation in the UV range), the illumination 'A' (M0; low radiation in the UV range) and the illumination 'D50' (M1; high radiation in the UV range) are shown in Figure 4 (Please note: The measurement conditions M0, M1 & M2 are explained in chapter 1.2). It is shown that a higher amount of UV radiation leads to a stronger emission in the visible blue spectrum. Especially this FBA property leads to serious issues in proofing process and will be discussed in detail.

1.1.4 Carrier and substrate composition

In order to enable fluorescence of FBAs, so called carrier substances are needed. Carrier enables a monomolecular distribution of FBAs in one layer (Blum, Linhart and Frenzel, 2002, p. 2). Typically, carriers are added to the coating and/or the paper pulp. Possible carriers are water soluble polymers (e.g. polyvinyl alcohol $[(C_2H_4O)_x]$), carboxymethyl cellulose (a cellulose derivative with carboxymethyl groups $[CH_2CO_2H]$), anionic or non-anionic degraded starch, and casein (Blum, Linhart and Frenzel, 2002, p. 3). Several other substances as carriers are possible (Blechschmidt, 2013, p. 447).

Many substrates, especially uncoated substrates, show a high concentration of cellulose fibers which also act as carriers (Blechschmidt, 2013, p. 447). Other substrates (especially proof substrates) contain only a small number of carrier molecules or none at all, as shown also in the results of this study.

1.1.5 Competitive absorbing substances

While the intensity of the emission highly depends on the amount of UV radiation available in the extinction range of any FBA, the FBA absorption can be reduced by other competitive absorbing substances in the paper composition that also show a strong absorption in this range. Figure 5 shows the spectral response of several slightly brightened (Substrates 1 & 2) and unbrightened substrates (Substrates 3 & 4) including the UV range.

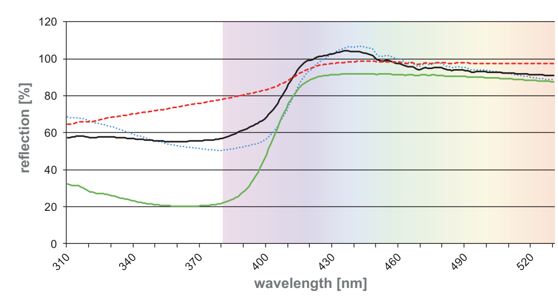

Figure 5: Spectral response of four substrates in the UV and blue range, under M1 measurement condition

When comparing the four substrates in Figure 5, only 'substrate 4' shows a decent absorption in the UV range. It can be assumed that loading this substrate with FBAs would lead to a high FBA emission efficiency. Compared to this, the assumed FBA emission efficiency would be lower for substrates 1, 2 & 3 because of their higher absorption in the UV range due to other competitive absorbing substances in the substrates.

1.1.6 Spectral response of Fluorescent Brightening Agents

Fluorophores are available with various excitation and emission characteristics. All types of FBAs used in the paper industry (cf. chapter 1.1.1) show a very comparable emission characteristic (shape of the spectral response curve) (cf. Figure 6). Please note that differences in the emission intensity occur because of different amounts of FBAs in each substrate.

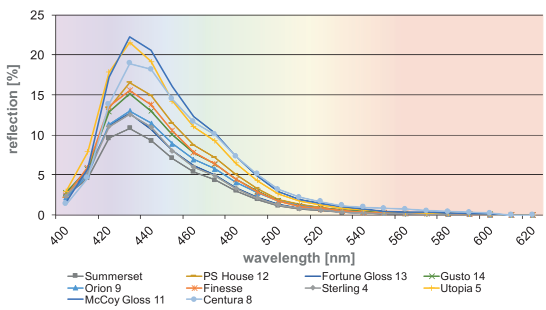

Figure 6: FBA emission of several proof- and production substrates, measured under M1 measurement conditions

Figure 6 shows the FBA emission of a representative collection of production substrates. Note: Only the FBA emission is shown. This is done by measuring each substrate once with the M2 condition and once with the M1 condition and calculating the difference of both measurements for each wavelength (cf. Equation 1).

$$\widehat{\beta_{emission}}(\lambda) = \beta_{M1}(\lambda) - \beta_{M2}(\lambda) \qquad [1]$$

where $\widehat{\beta_{emission}}(\lambda)$ is the wavelength depending emission of a brightened substrate, $\beta_{M1}(\lambda)$ is the measurement of a substrate using M1 condition, and $\beta_{M2}(\lambda)$ is the measurement of a substrate using M2 condition.

1.2 Influence of Fluorescent Brightening Agents on the proofing process

ISO 3664:2009 (International Organization for Standardization, 2009a) in combination with ISO 13655:2009 (International Organization for Standardization, 2009b) defines the measurement/illumination conditions M0, M1, M2, and M3. The measurement condition M1 is based upon the known D50 relative spectral power distribution defined by ISO 13655:2009, but it is extended to the spectrum of the illuminant's UV range in its definition. This means a "real" illumination must have a comparable amount of UV radiation as defined in ISO 13655:2009 within defined tolerances (International Organization for Standardization, 2009a). The M1 condition is also obligatory for new light boxes. Practically, using ISO 3664:2009 conforming measurement equipment and light boxes induces an increase of excitation of the FBAs. Hence, FBA induced effects are increased.

The standardized measurement condition M0 is based on standard illuminant A (low UV radiation), M2 is based on any standard illuminant without UV component and M3 is based on any illuminant with polarization filters.

The consequence of the heavy use of FBAs in production substrates is the now obligatory consideration of the UV amount. Therefore, it becomes possible that a brightened substrate (e.g. an offset production paper) is measured with a spectral photometer as it is perceived by the operator in a light booth if both are relying on the M1 condition (and therefore a comparable high UV intensity).

Today, most proof substrates are not or only slightly brightened, while many production substrates show a very high FBA concentration. This difference leads to serious issues in proofing processes, especially with regard to ISO 3664:2009. Figure 7 shows two measurement situations (M2 & M1) that result in different ΔE_{00} values for unequally brightened substrates. The expected visual color difference $\Delta E_{percepted}$, in the case of a light booth using the M1 condition, is also presented to show the correlation to measurements with the M1 condition and the difference to the M2 condition (ΔE_{00} and $\Delta E_{percepted}$ are theoretically equal if both devices, the measurement device as well as the light booth, offer the same UV amount). Please note that a conventional white point correction is already performed for the following proof substrates. Therefore, the identified ΔE_{00} values are only caused by the fluorescence effect.

Table 1 shows that a measurement of the production substrate 'Heaven42' from the company Scheufelen GmbH + Co. KG, and the proof substrate 'Epson SPP 205' from the company EPSON Deutschland GmbH result in a ΔE_{00} = 1.99, if measurement condition M2 is used. In contrast, measurement condition M1 results in a high ΔE_{00} = 8.02 because of large differences in the FBA concentration of both substrates. Consequentially a $\Delta E_{percepted}$ = 8.02 can be expected.

Table 1: Resulting color differences ΔE_{00} caused by highly deviating FBA concentrations

Substrate	**Measurement condition M2**	**Measurement condition M1**	**Light booth**
Proof substrate: Epson SPP 205 (unbrightened)	L^* = 94.6 a^* = −0.1 b^* = −1.9	L^* = 94.6 a^* = −0.4 b^* = −2.9	
Production substrate: Heaven 42 (highly brightened)	L^* = 93.1 a^* = 0.0 b^* = −3.9	L^* = 93.7 a^* = 2.3 b^* = −12.9	
Color difference	ΔE_{00} = 1.99	ΔE_{00} = 8.02	$\Delta E_{percepted}$ = 8.02

Table 2: Resulting color differences ΔE_{00} caused by medium high deviating FBA concentrations

Substrate	Measurement condition M2	Measurement condition M1	Light booth
Proof substrate: Trust Premium Glossy (slightly brightened)	L^* = 95.1 a^* = 0.2 b^* = −5.3	L^* = 95.6 a^* = 1.2 b^* = −5.3	
Production substrate: Heaven 42 (highly brightened)	L^* = 93.1 a^* = 0.0 b^* = −3.9	L^* = 93.7 a^* = 2.3 b^* = −12.9	
Color difference	ΔE_{00} = 1.69	ΔE_{00} = 5.59	$\Delta E_{percepted}$ = 5.59

Even if a proof substrate is brightened, it is not assured to gain a sufficient match, especially for highly brightened production substrates (cf. Table 2).

Table 2 shows that measurements of the production substrate 'Heaven42' and the proof substrate 'Trust Premium Glossy' from the company Schoeller Technocell GmbH & Co. KG lead to a ΔE_{00} = 1.69, if measurement condition M2 is used. In contrast, measurement condition M1 results in a medium high ΔE_{00} = 5.59 because of still large differences in the FBA concentration of both substrates. Consequentially a $\Delta E_{percepted}$ = 5.59 can be expected.

Table 1 and 2 show that it is quite possible to identify color differences between a brightened production paper and a corresponding unbrightened proof substrate using the M2 condition. These differences do not correspond to the differences that are perceived in a light booth using the M1 condition. Furthermore, if both substrates are observed under a non ISO 3665:2009 conforming illumination, FBA induced differences on the perception become hardly predictable.

So far, only the direct effect of the substrate is shown. Usually, printing substrates are printed with various colors and various ink systems. Each color, color combination and area coverage combination leads to an individual influence on the FBAs absorption and emission (Dattner, Bohn and Urban, 2011).

Figure 7 gives an overview about color shift induced by FBAs at different locations in the CIELAB color space for the production substrate 'Heaven42'. Please note: The L^* value is only very slightly affected by FBAs and is therefore not plotted in this graph.

It is shown that almost all color coordinates are more or less affected by the substrate's FBA. This results in a gamut grow in the blue region of the CIELAB color space while a slight gamut decrease in the green and yellow region can be identified. It can be concluded that not only the substrate is affected and critical in the proofing process, but also colors printed on it.

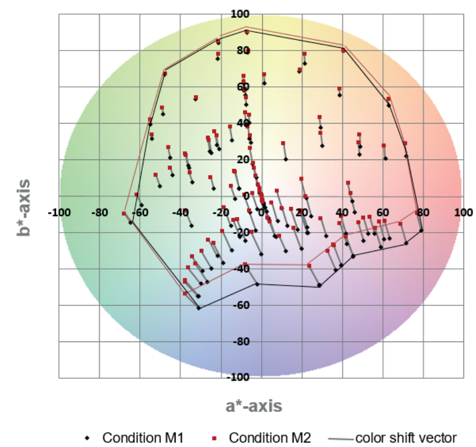

Figure 7: Influence of FBAs on color coordinates of colors and the resulting color gamut

1.3 State of the scientific knowledge

It is documented and explored that the effect of different FBAs in various printing substrates leads to individual, illumination dependent effects with unprinted and fully printed surfaces (Erhard, Alber and Götze, 2003; Löffler and Green, 2008; Zwinkels, 2008). The optical behavior of halftone samples on brightened substrates is also empirically determined (Kraushaar and Geßner, 2006; Fiebrandt, 2007; Pertler and Bertholdt, 2007), but not physically founded. In the past few years, several physically founded models have been developed and described (Dattner, Bohn and Urban, 2011).

While these models show a good prediction performance, they cannot solve the initial issue, that a brightened paper will often show a higher reflection in the blue region of the spectrum for the measurement condition M1 than a corresponding unbrightened proof substrate.

The ISO standard 3664:2000, which specifies viewing and measurement conditions for images on reflective media, has been revised in 2009 and is therefore replaced by ISO 3664:2009. While ISO 3664:2000 defines a UV metamerism index (MI_{UV}) to be smaller or equal than 4, ISO 3664:2009 now requires MI_{UV} < 1.5 and recommends MI_{UV} < 1.0. That relates to a tolerance of the UV amount, compared to D50 being 100 %, from 60 % to 130 %. This results in a much closer correlation between the actual used illumination and the CIE daylight illuminant D50. In case of a perfect match of the UV content in the spectra, MI_{UV} becomes 0 (Kraushaar, 2012). Therefore, if the measurement device as well as the used illumination of a light booth both comply to ISO 3664:2009, the influence of FBAs are taken into account within the defined tolerances.

The Graphic Technology Research Association (fogra, 2014) recently released fogra51/fogra52 in the context of ISO 3664:2009. This is the "codename" or the label for the characterization data reflecting ISO 12647-2:2013 based printing on matt and glossy coated offset papers (International Organization for Standardization, 2013). Using FBA rich proofing substrates, in particular to simulate FBA rich production substrates, was subject in a fogra analysis reflecting the upcoming permanence criteria of ISO 12647-7 by achieving a proof to print match (Kraushaar, 2014). Fogra51 (PSO Coated V3) defines an M1 target of L^* = 95.0, a^* = 1.5 and b^* = −6.0 to take FBA induced effects into account. In contrast, fogra52 (PSO Uncoated V3) defines an M1 target of L^* = 93.5, a^* = 2.5 and b^* = −10 (Kraushaar, 2013). In this context, new proof substrates have been developed and certified, that include some amount of FBA. The Felix Schöller Group (2016) offers fogra51 and fogra52 certified proof substrates having white points as shown in Table 3. The shown CIELAB values are provided by the Felix Schöller Group (2016).

Table 3: Properties of typical brightened proof substrates conforming to fogra51 & fogra52 under M1 condition

Substrate	Certification	Appearance	Grammage (g/m²)	Color coordinate (M1)		
				L^*	a^*	b^*
TRUST premium	Fogra 51	Glossy	250	95.6	1.2	−5.3
TRUST premium	Fogra 51	Semi-glossy	250	96.8	0.9	−5.1
TRUST economy	Fogra 52	Matte	150	96.9	2.3	−10.2
TRUST commercial	Fogra 52	Semi-glossy	195	96.0	1.5	−10.3

2. Methods

The final goal of this study is to simulate illumination independent any brightened production substrate with any unbrightened or slightly brightened proof substrate by printing an FBA ink layer with a variable area coverage using inkjet.

2.1 Equipment

2.1.1 Inkjet printers

In this study, two commercially available printers are used. For the actual print of the FBAink compositions, a Canon ix6550 (DIN A3+) is used, while additional color test patches are printed in a second step using an Epson Stylus Pro 4900 proof printer.

2.1.2 Measurement devices

For the analysis of substrates and printed FBA layers a TEC5 UV-VIS laboratory spectral photometer in combination with a 45°/0° geometry and a xenon permanent light source is used. The spectral resolution is 1 nm and the spectral range is 310 nm to 990 nm. By using filters, several illumination types can be simulated, including M0, M1 and M2 in accordance to ISO 13655:2009 (International Organization for Standardization, 2009b). The realized UV amount of the M1 measurement condition is 115 % compared to nominal value, i.e. within tolerances according to ISO 3665:2009 (International Organization for Standardization, 2009a). For the analysis of printed color test charts on the FBA layers, an EFI ES-2000 is used. This device is capable to perform 45°/0° measurements with the measurement conditions M0, M1 & M2. The spectral range is limited to the visible range of the electromagnetic spectrum (380 nm to 730 nm) and offers a spectral resolution of 10 nm.

2.1.3 Measurement conditions

All measurements are done with a black backing in accordance to ISO 13655:2009, using a 45°/0° measurement geometry (International Organization for Standardization, 2009b). Colorimetric values are calculated for a 2° observer and the illuminant type D50.

2.2 Substrates

As pointed out, the goal of this study is to use any proof substrate to simulate any production substrate. Because interactions especially between different FBAs and the substrate are expected, four different proof substrates are examined.

Table 4: Overview of properties of used proof and production substrates

Substrate type	Substrate name	Condition M1			Grammage (g/m²)	Appearance	FBA amount
		*L**	*a**	*b**			
Proof	Epson SPP 205	94.55	−0.39	−2.88	205	Semi-matte	Non
Proof	EFI BEST Xpress Paper	96.47	0.30	−2.79	220	Semi-matte	Low
Proof	EFI Proof Paper 9120XF	97.15	0.84	−4.82	120	Matte	Medium
Proof	EFI Budget Proof 3170	96.60	2.64	−6.64	165	Semi-matte	Medium
Production	Heaven 42	94.55	−0.39	−12.88	170	Semi-matte	Very high

For the actual simulation, the highly brightened substrate 'Heaven42' is chosen out of 100 examined production substrates as a worst case scenario. Table 4 shows the properties of the used proof substrates, as well as the production substrate 'Heaven42' with corresponding CIELAB values. Please note that all CIELAB values are calculated from measurements with the UV-VIS spectral photometer and the M1 condition. The proof substrates are used as base materials to investigate the different spectral and colorimetric behavior of FBA ink compositions (cf. chapter 2.3). Figure 8 shows the spectral response of all four proof substrates used.

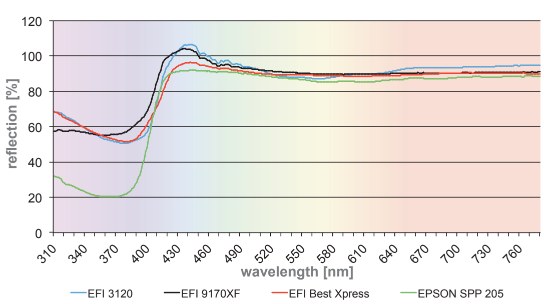

Figure 8: Spectral response of the proof substrates using measurement condition M1

2.3 Ink compositions

To achieve the goal of this study, different ink compositions with individual properties are developed. To achieve a good agreement between the emission shape found for all production substrates (cf. chapter 1.1.6), only fluorophores with a comparable emission and excitation shape are suitable for this study. Furthermore, fluorophores which fulfill this requirement need also to be water soluble and colorless in the visible region of the electromagnetic spectrum. Finally, these fluorophores need to be compatible with ink-typical ingredients, i.e. glycerin ($C_3H_8O_3$).

Typical fluorescent brightening agents commonly used in the paper production fulfill these requirements. Mainly three types of optical brighteners are used in the paper industry, all based on the mentioned diaminostilbene molecule shown in Figure 2. The main difference is found in the number of solubilizing sulfonic acid groups (cf. chapter 1.1.1).

Preliminary investigations to this study show that hexasulpho-based FBAs show the most intense emission compared to disulpho- and tetrasulpho-based FBAs. Therefore, we focus in our study on a hexasulpho-based FBA (Leucophor SAC, Archroma).

Finally, FBA ink compositions need to contain at least one moistening agent to protect the nozzels of the inkjet printer from drying out and therefore from nozzle clogging. The chosen moisture agent we used in this study is glycerin. In a second step, polyethylene glycol is added as a carrier to Ink 3 (Ink 3 → Ink 3_b) to avoid greening and increase the FBA emission intensity.

Table 5: Investigated ink compositions

Ingredient	Ink 1	Ink 2	Ink 3	Ink 4	Ink 5	Ink 6
	(%)					
Water	89.5	87.5	85.0	80.0	70.0	50.0
Glycerin	10.0	10.0	10.0	10.0	10.0	10.0
FBA (hexasulpho-based)	0.5	2.5	5.0	10.0	20.0	40.0
Sum (∑)	100.0	100.0	100.0	100.0	100.0	100.0

2.4 Test chart

The test chart, developed for this study, has a wedge with test patches with area coverages ranging from 0 % up to 100 %. Because the black channel of the printer is used for the FBAink, the test chart is build up out of different area coverages of black. Therefore, a 100 % black area coverage equals a 100 % FBA ink layer area coverage (cf. Figure 9).

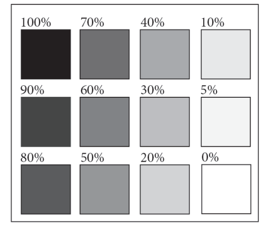

Figure 9: Test chart for FBA-ink area coverage variable investigations

3. Results

3.1 Area coverage controlled Fluorescent Brightening Agent emission

The intensity of the fluorescence is directly controlled by the area coverage of the printed FBA upon a unbrightened or slightly brightened proof substrate. To illustrate the principle, Figure 10 shows the influence of a printed FBA layer upon a proof substrate (Epson SPP 205) with area coverages starting with 0 % up to 100 % (Figure 10 top: Illumination condition M0; Figure 10 middle: Illumination condition M1; Figure 10 bottom: UV black light).

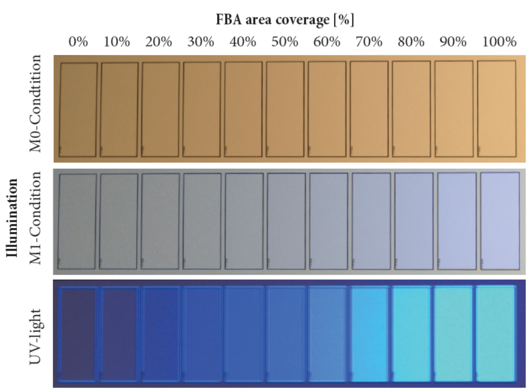

Figure 10: Illustration of the FBA induced emission effect for several area coverages and illuminations in a light booth

It can be demonstrated that an increasing FBA area coverage leads to a more intense emission in the blue region of the electromagnetic spectrum. This effect is slightly visible under M0 (low UV amount), but clearly visible if viewed using M1 (high UV amount). To outline the effect, an additional image with pure UV light is presented (cf. Figure 10).

Figure 11 shows the spectral response of these test patches, if measured using the M1 condtion.

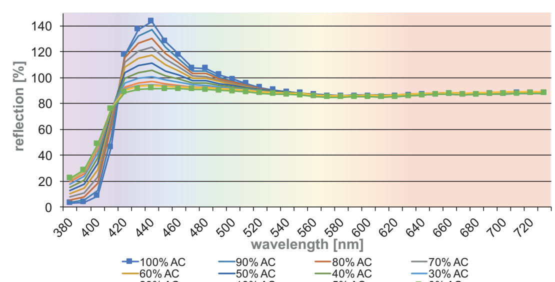

Figure 11: Spectral response of a variable area coverage (AC) of FBA ink for the proof substrate 'Epson SPP 205' with different FBA ink layer coverages, measured under M1 measurement condition

As expected, an increase of the FBA area coverage leads to a more intense emission between 420 nm and 550 nm, while the spectral response between 550 nm

Table 6: Color coordinates of investigated proof substrates with and without FBA ink (0 % or 100 % area coverage, respectively)

Substrate	FBA concentration	FBA area coverage (%)	Condition M1			Δb*
			L*	a*	b*	
Epson SPP 205	no FBA ink	0	94.57	−0.39	−2.84	13.18
	5 %	100	95.44	2.95	−16.03	
EFI 3170	no FBA ink	0	95.92	2.64	−6.72	6.61
	5 %	100	96.60	4.37	−13.33	
EFI Best Xpress	no FBA ink	0	96.21	0.24	−2.69	11.87
	5 %	100	96.71	3.42	−14.56	
EFI 9120XF	no FBA ink	0	96.54	0.84	−4.72	8.33
	5 %	100	97.34	2.13	−13.06	

and 780 nm remains unaffected. It can also be shown, that a higher FBA area coverage leads to a stronger absorption between 380 nm and 420 nm. Thereby, an 100 % FBA area coverage shows the strongest absorption while a 0 % FBA area coverage (pure, unbrightened substrate) shows an absorption minimum.

3.2 Fluorescence emission of ink compositions without carrier

3.2.1 Influence of Fluorescent Brightening Agent area coverage

Figure 12 shows the color coordinates in an a^*b^* diagram of all four proof substrates (cf. chapter 2.2) with FBA area coverages ranging from 0 % up to 100 % for the measurement condition M1 and an ink with a 5 % FBA concentration (Ink 3, cf. chapter 2.3). The area coverage of 0 % for each substrate is marked with a red dot. A 100 % area coverage is marked with a green dot. Additionally, Table 6 gives detailed information about the color coordinates of each substrate/ink combination (0 % & 100 % area coverage are shown). Please note that the L^* value is not considered in the following because it is only slightly affected by different intensity of FBA emissions (cf. Table 6). In the case of the substrates 'Epson SPP 205', 'EFI 3170' and 'EFI Best Xpress', a comparable behavior can be observed: With an increase of the FBA area coverage, a strongly increasing negative b^* value can be found, while the a^* value only increases slightly. Thereby, the a^*b^* shift is almost linear in all four cases (cf. Figure 12). Differences can be found for the maximum realizable FBA emission, represented by the Δb^* value (cf. Table 6). While the 'Epson SPP 205' shows with $\Delta b^* = 13.18$ the strongest FBA induced shift, the 'EFI Best Xpress' shows with $\Delta b^* = 11.87$ only a slightly lower realizable emission. Interestingly, the 'EFI 3170' substrate shows with $\Delta b^* = 6.61$ a much lower maximum emission. Only the substrate 'EFI 9120XF' shows, regarding the a^*b^* shift, a significant divergent behavior. While the b^* value is developing as expected, the a^* value only increases slightly. Hence, the FBA induced emission becomes slightly greenich (cf. chapter 1.1.2).

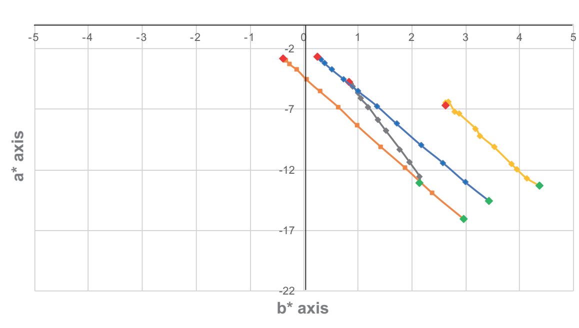

Figure 12: Influence of a variable area coverage of FBA ink (5 % FBA) for four proof substrates, under M1 measurement condition

Figure 13 shows exemplarily for two substrates ('EFI 3170' & 'EFI Best Xpress') the corresponding spectral responses for 0 % FBA area coverage, 50 % FBA area coverage and 100 % FBA area coverage, each. It can be observed, that only the emission intensity increases while the emission shape remains the same for both substrates. Also, the wavelength range between 550 nm and 780 nm remains unaffected.

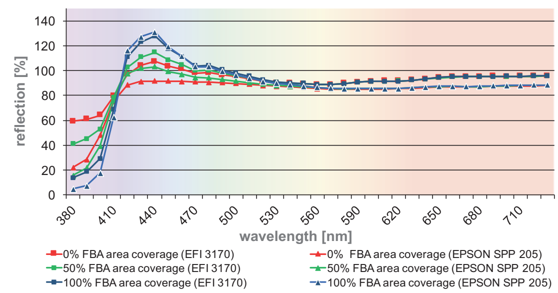

Figure 13: Spectral response of two substrates with 0 %, 50 % & 100 % FBA ink area coverage each, under M1 measurement condition

3.2.2 Influence of Fluorescent Brightening Agent area coverage in combination with concentration

Figure 14 (top-left) shows the interdependence of the FBA area coverage and the FBA concentration for the proof substrate 'Epson SPP 205'. It can be seen that an FBA concentration of 5 % results in a value of $b^* = -16.07$ ($\Delta b^* = 13.22$) for a FBA area coverage of 100 %. With an increasing FBA concentration a maximum b^* value of 17.81 can be achieved for a 20 % FBA concentration. Up to this concentration, the already known linearity between a^* and b^* holds true (cf. chapter 3.1). For a 40 % concentration, a different behavior can be identified. Already for lower FBA area coverages it can be observed that the b^* value is still increasing while there is (compared to lower concentrations) a significant drift in the a^* value. An FBA area coverage of 90 % leads to a maximum b^* value of 16.10 but the a^* value starts to drop. For an FBA area coverage of 100 % the b^* value starts to decrease while the a^* value drops even more. As a consequence, the bluish emission becomes greenish. This effect is known as "greening" or "greying" (cf. chapter 1.1.2). Fluorophores need to be fixed in a monomolecular planar layer to a substrate to develop a stable emission (cf. chapter 1.1). It can be stated that the 'Epson SPP 205' substrate does not offer sufficient OH groups for this FBA concentration.

Figure 14 (top-right) shows the interdependence of the FBA area coverage and the FBA concentration for the proof substrate 'EFI Best Xpress'. This substrate shows a comparable behavior like the 'Epson SPP 205' proof substrate. This substrate shows for 20 % FBA concentration and 100 % FBA area coverage the most intense emission ($\Delta b^* = 18.27$). This substrate also shows for

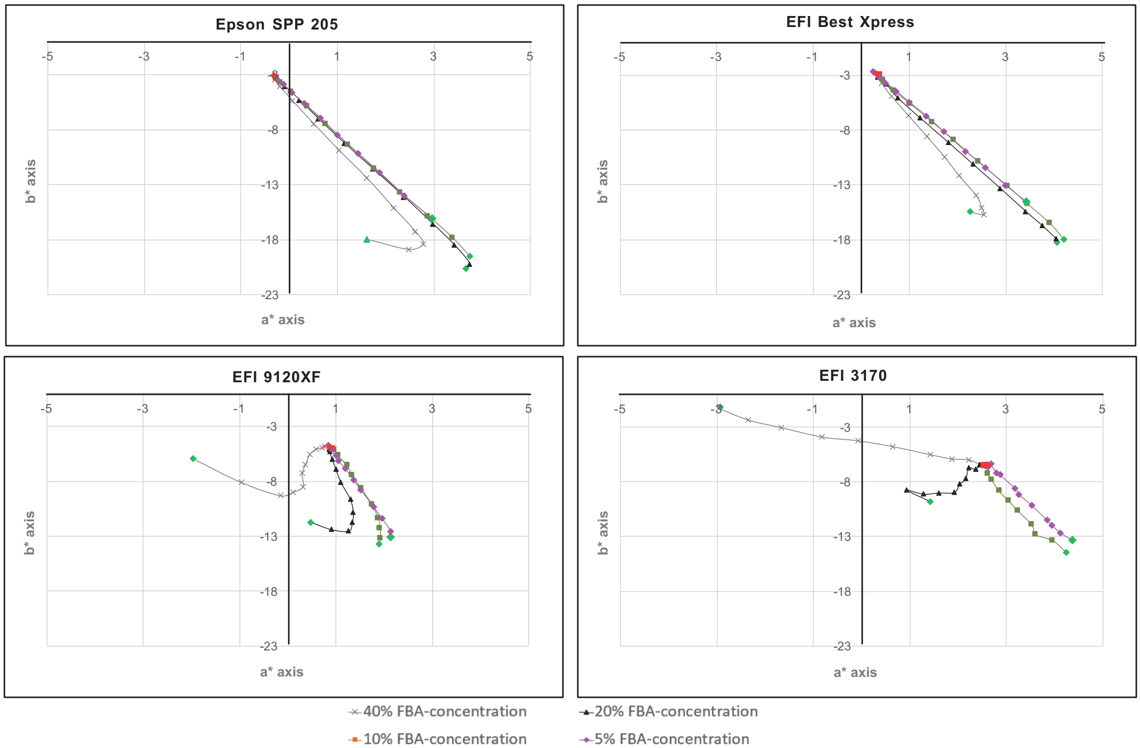

*Figure 14: The a*b* color coordinates depending on the FBA area coverage and concentration for four proof substrates (top-left 'Epson SPP 205'; top-right 'EFI Best Xpress'; bottom-left 'EFI 9120XF'; bottom-right 'EFI 3170'), under M1 measurement condition*

40 % a comparable, uncharacteristic behavior like the 'Epson SPP 205'. Again, it can be assumed that this is a greening effect because the substrate does not offer sufficient OH binding groups.

Figure 14 (bottom-left) shows the interdependence of the FBA area coverage and the FBA concentration for the proof substrate 'EFI 9120XF'. In contrast to the previous discussed substrates, this substrate shows only a decent increase in its b^* value ($\Delta b^* = 8.92$) for a 100 % FBA area coverage and an FBA concentration of 10 %. If the FBA concentration is increased to 20 %, the previously observed a^*b^* drift also occurs but in this case already for 20 % FBA concentration. Even for a 10 % FBA concentration this effect can be observed. If the FBA concentration is increased to 40 %, the FBA area coverage depending color drift becomes even more complex. While a strong a^* drift can be observed already for low concentrations (compared to the corresponding FBA concentration of the 'Epson SPP 205' substrate), the b^* value increases up to an FBA coverage of 60 % for a FBA concentration of 20 %. Then it stops to increase and starts to drop again, while the a^* value drops even more (up to $\Delta a^* = 2.9$ for a 40 % FBA concentration). It can be stated that the greening effect is responsible. This already becomes relevant for a 10 % FBA concentration. Even for a 5 % FBA concentration a greening effect can be identified. As a result, it can be assumed that this substrate offers less OH groups than the proof substrates 'Epson SPP 205' and 'EFI Best Xpress'.

Finally, Figure 14 (bottom-right) shows the interdependence of the FBA area coverage and the FBA concentration for the proof substrate 'EFI 3170'. This substrate shows a stable FBA emission only for an FBA area coverage of 100 % with an FBA concentration of 5 % ($b^* = -13.31$; $\Delta b^* = 6.65$). If the FBA concentration is increased to 10 %, a greening effect can be identified. This substrate shows a much stronger a^*b^* shift for higher FBA concentrations compared to the other three substrates. If the FBA concentration is increased to 40 %, the b^* value is not increasing for any FBA area coverage. In opposite, already a 5 % FBA area coverage leads to a decrease in the b^* value. If the area coverage is increased to 100 %, the b^* value shifts from −6.56 to −1.18, while the a^* value shifts from 2.63 to −2.93 ($\Delta a^* = 5.56$). In this case, it can be assumed that this substrate does not offer any OH binding groups at all to enable a stable emission.

It can be concluded, that a 5 % FBA ink composition shows already high FBA emission intensities with only a slightly greening effect for the proof substrate 'EFI 3170'. Unfortunately, depending on the substrate, the realizable FBA emission intensity is limited. Therefore, in the next chapter this concentration is used to analyse the impact of a carrier on the emission intensity.

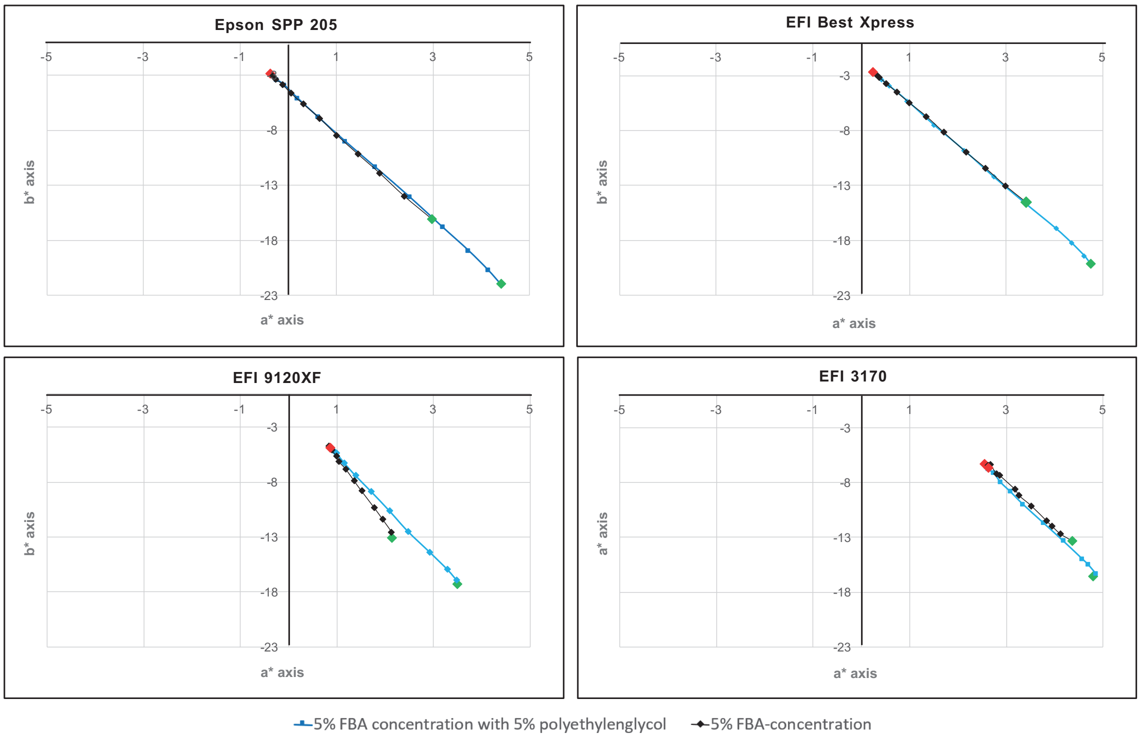

*Figure 15: The a*b* color coordinates for several FBA area coverages and a 5 % FBA concentration with and without 5 % polyethylene glycol of four proof substrates (top-left 'Epson SPP 205'; top-right 'EFI Best Xpress'; bottom-left 'EFI 9120XF'; bottom-right 'EFI 3170'), under M1 measuremnt condition*

3.3 Fluorescence emission of ink compositions with carrier

As pointed out in the chapter 1, carrier substances are known to prevent the so called "greening effect". Furthermore, these carriers are also known in principle to improve the FBA efficiency. For this study, polyethylene glycol is used exemplarily as a carrier with a concentration of 5 % in combination with Ink 3 (5 % FBA). Polyethylene glycol ($C_{2n}H_{4n+2}O_{n+1}$) is suitable because this substance fulfills the requirements: it is water soluble, colorless and compatible with the used FBA.

Figure 15 (top-left) shows the 5 % FBA ink without carrier and the 5 % FBA ink performance with 5 % polyethylene glycol as a carrier compared to each other for the proof substrate 'Epson SPP 205'. It is found that the addition of 5 % polyethylene glycol leads to a significant improvement in the FBAs emission, and therefore the efficiency compared to 5 % FBA without carrier (without carrier: $b^* = -16.07$; with carrier: $b^* = -21.98$). No greening effect is observed.

Figure 15 (top-right) shows the 5 % FBA ink without carrier and the 5 % FBA ink performance with 5 % polyethylene glycol as a carrier compared to each other for the proof substrate 'EFI Best Xpress'. Again, it is found that the addition of 5 % polyethylene glycol leads to a significant improvement in the FBAs emission compared to 5 % FBA without carrier (without carrier: $b^* = -14.55$; with carrier: $b^* = -20.12$). As well, no greening effect is observed.

Figure 15 (bottom-left) shows the 5 % FBA ink without carrier and the 5 % FBA ink performance with 5 % polyethylene glycol as a carrier compared to each other for the proof substrate 'EFI 9120XF'. In this case, the addition of 5 % polyethylene glycol leads to a strong increase of the FBAs emission compared to 5 % FBA without carrier (without carrier: $b^* = -13.07$; with carrier: $b^* = -17.29$), although still remaining lower that on 'Epson SPP 205' and 'EFI Best Xpress' substrates. Furthermore, while the use of 5 % FBA concentration without carrier leads to a slight greening effect, the addition of polyethylene glycol fully prevents this effect.

Finally, Figure 15 (bottom-right) shows the performance of 5 % FBA ink without carrier and the 5 % FBA ink with 5 % polyethylene glycol as a carrier compared to each other for the proof substrate 'EFI 3170'. The addition of 5 % polyethylene glycol has similar effects as in the case of 'Epson SPP 205' and 'EFI Best Xpress' substrates, only in smaller scale (without carrier: $b^* = -13.31$; with carrier: $b^* = -16.53$). Again, no greening effect is observed.

Figure 16 (left) shows the influence of the 5 % FBA ink without carrier with several area coverages for all

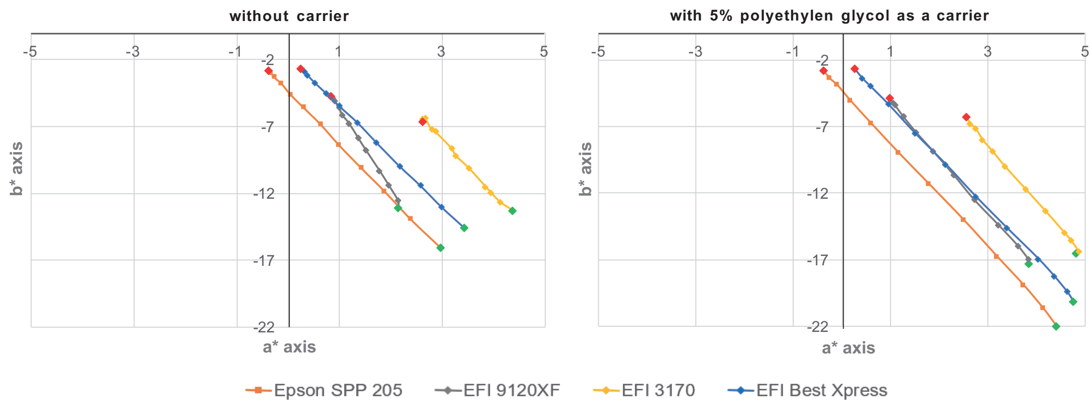

Figure 16: Visualization of the influence of polyethylene glycol as a carrier; left: without carrier; right: with 5 % polyethylene glycol as a carrier, for four proof substrates, under M1 measurement condition

examined proof substrates, as already presented in the chapter 3.1. In contrast, Figure 16 (right) shows the influence of the same ink if 5 % polyethylene glycol is added to the ink composition. Again, the previously presented negative increase of the b^* value can be seen. Additionally, Figure 16 (right) shows that the linearity observed previously for the substrates 'Epson SPP 205', 'EFI 3170' & 'EFI Best Xpress' can also be achieved for the substrate 'EFI 9120XF' by adding 5 % polyethylene glycol to the ink and prevent this way any greening.

Summarized, it is shown that the addition of 5 % polyethylene glycol as a carrier not only prohibits any greening but also leads to a significant increase of the FBAs emission, and therefore the efficiency. This is important for the final goal of this study because this way any proof substrate can be used to simulate theoretically every production substrate without the risk of greening. Furthermore, the realizable intense blue emission offers the possibility to use lower FBA area coverages to simulate even massively brightened production substrates for all proof substrates in this study.

3.4 White point simulation of production substrates

The actual white point simulation is presented and exemplarily done for the highly brightened production substrate 'Heaven42' and the proof substrate 'EFI 3170', in two separate steps.

3.4.1 Spectral fit of the wavelength range from 550 nm to 780 nm

The wavelength range of 550 nm to 780 nm is relevant to modify the overall spectral response of the proof substrates to the production substrates spectral response by printing small amounts of cyan, magenta, yellow and black. This range results from the fact that any FBAs influence is limited to the range of 380 nm to 550 nm. To achieve a good match, we propose to print a test chart with defined small amounts of CMYK upon the corresponding proof substrate before any FBA addition is performed. Each patch can be measured using either measurement condition M2, M1 or M0. The spectral difference ΔE_S between each test patch printed on the proof $\widehat{\beta(\lambda)}$ and the production $\beta(\lambda)$ substrate is calculated using the "Spectral Metric" (LSS, Least square sum) following Equation 2 (Dattner, 2010, p. 38).

$$E_S := \sum_{\lambda}\left(\beta(\lambda)-\widehat{\beta(\lambda)}\right)^2 \qquad [2]$$

Table 7 shows the actual LSS values of each CMYK combination for the proof substrate 'EFI 3170' and the production substrate 'Heaven42' in case of given CMYK combinations, calculated from the Equation 2.

Table 7: Overview of used CMYK combinations to minimize the LSS value

Cyan	Magenta	Yellow	Black	
(%)				ΔE_S (LSS)
0	0	0	0	421.25
0	0	0	1	806.62
0	0	0	2	1104.82
0	0	0	3	623.94
1	0	0	0	974.30
2	0	0	0	346.18
0	1	0	0	757.25
0	2	0	0	873.68
0	0	1	0	260.82
0	0	2	0	380.84
1	1	0	0	570.63
1	0	1	0	240.82
0	1	1	0	412.21

A yellow area coverage of 1 % in combination with a cyan area coverage of 1 % leads to a minimum. This white point modification results in a ΔE_{00} = 6.15

between both substrates for the measurement condition M1 compared to a $\Delta E_{00} = 8.25$ if no white point adjustment is performed.

Figure 17 shows the corresponding spectral responds of the production substrate 'Heaven42', the proof substrate 'EFI 3170' (without adjustment) and the proof substrate 'EFI 3170' with white point adjustment (1 % Cyan; 1 % Yellow), limited to the relevant wavelength range of 550 nm to 780 nm.

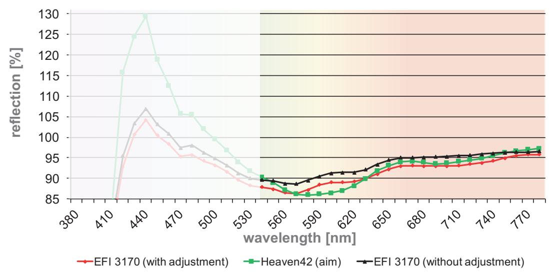

Figure 17: Spectral fit of the wavelength range 550 nm to 780 nm of the production substrate 'Heaven42' and 'EFI 3170' with & without adjustment, under M1 measurement condition

3.4.2 Spectral fit of the wavelength range from 380 nm to 550 nm

To compensate and adjust the remaining difference between both substrates ($\Delta E_{00} = 6.15$), different amount of liquid FBA is printed onto the adjusted proof substrate using inkjet printing. In a first step, the actual FBA emission of the production substrate needs to be determined. This is done spectrally by performing two measurements (M1 & M2) and applying Equation 1. Figure 18 shows the resulting spectral difference of both measurements.

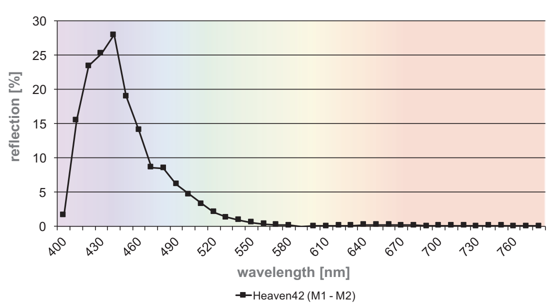

Figure 18: FBA induced spectral response of the production substrate 'Heaven42' using measurement conditions M1 & M2

By a further use of Equation 2 (limited to 380 nm to 550 nm) with $(\widehat{\beta(\lambda)})$ of the production substrate measured with the M1 condition and $\beta(\lambda)$ of the same production substrate measured with the M2 condition, the corresponding LSS value can be determined. This LSS value performs as an indicator for the FBA induced emission intensity. In this case, an LSS value of 1 364.04 is determined for the production substrate 'Heaven42'.

To determine the required FBA area coverage, a test chart with 5 % FBA ink, including 5 % polyethylene glycol as a carrier, is printed with area coverages of 0 %, 5 %, 10 %, 20 %, …, 100 % onto the adjusted proof substrate (cf. chapter 2.4). Each printed patch needs to be measured and calculated as it is done for the production substrate. Figure 18 shows the resulting LSS values for each FBA ink area coverage printed onto the 'EFI 3170' proof substrate with $(\widehat{\beta(\lambda)})$ for the already modified proof substrate and $\beta(\lambda)$ for the same but in its FBA concentration modified proof substrate.

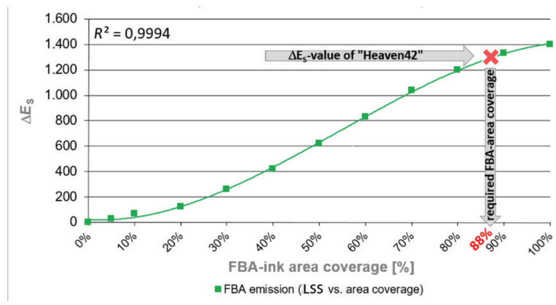

Figure 19: Determination of the required FBA ink area coverage

Figure 19 shows the correlation between the FBA area coverage and the resulting FBA emission intensity. The production papers LSS value is inserted accordingly, which leads to an interpolated FBA area coverage of 88 % to obtain a maximum correlation between the proof substrate and the production substrate. This third-degree polynomial interpolation ($R^2 = 0.9994$) is based on all measured FBA area coverages.

The final achieved color difference between the modified 'EFI 3170' proof substrate and the production substrate 'Heaven42' is $\Delta E_{00} = 0.89$. To outline this result, Figure 20 shows the spectral response of the unmodified proof substrate, the modified proof substrate and the production substrate 'Heaven42'.

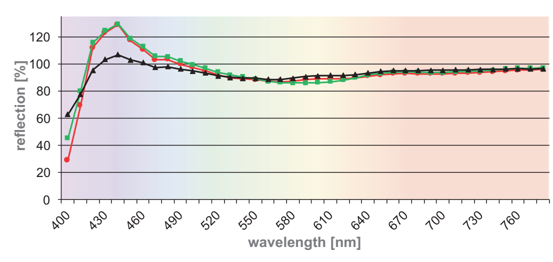

Figure 20: Spectral visualization of the performance of the enhanced white point simulation on the production substrate 'Heaven42' and proof substrate 'EFI 3170' (without and with adjustment, at 88 % FBA ink coverage), under M1 measurement condition

Figure 20 clearly illustrates the excellent correlation between the proof substrate and the production substrate, if a spectral white point and an FBA emission adjustment is performed.

4. Discussion

This study shows that liquid hexasulpho-based FBAs can be printed successfully in an area coverage variable inkjet process. While some of the investigated proof substrates show a greening depending on the FBA concentration, this effect is avoidable by adding a carrier to the FBA ink formulation, improving the overall FBA emission efficiency in addition. Furthermore, it is shown that the emission intensity cannot be extended infinitely by increasing the FBA concentration. Because all FBA adjusted proof substrates show a strong FBA emission intensity already for a 5 % FBA concentration (with 5 % polyethylene glycol), there is no need to use higher concentrations.

As a result, it is shown that all examined proof substrates can be used to simulate any brightened production substrate regardless the illumination condition. This holds true, if the overall spectral response for each wavelength of a given proof substrate is equal or higher compared to the production substrate to be simulated. This condition is fulfilled for all investigated proof substrates in this study.

Exemplarily, it is shown for the production substrate ‘Heaven42’, in combination with the proof substrate ‘EFI 3170’, that a very precise and illumination independent correlation can be achieved ($\Delta E_{00} = 0.89$ in the case of measurement condition M1). While typical fogra51/fogra52 certified proof substrates perform better compared to previously available substrates in the context of colorimetric agreement, especially extremely brightened production substrates (such as ‘Heaven42’) cannot be simulated adequately. Actually, if the production substrate ‘Heaven42’ is simulated using the fogra52 certified proof substrate ‘TRUST commercial (Semi-glossy)’, a $\Delta E_{00} = 3.61$ ($\Delta b^* = 4.21$) is measured. This match is achieved for D50 being 115 % (cf. chapter 1.3). As stated in the chapter 1.3, ISO 3664:2009 allows an UV amount of 60 % up to 130 % compared to D50 being 100 %. This still wide tolerance leads to a $\Delta E_{00} = 3.15$ ($\Delta b^* = 3.98$) if the measurement device shows an UV amount of 60 %. If a corresponding light booth shows an deviating UV amount of 130 %, a $\Delta E_{00} = 4.55$ ($\Delta b^* = 7.31$) is observed. In contrast, this study approach leads to a $\Delta E_{00} = 0.53$ ($\Delta b^* = 0.46$) if the measurement device has 60 % UV amount and a $\Delta E_{00} = 1.07$ ($\Delta b^* = 0.92$) if the light booth shows an deviating UV amount of 130 %.

This scenario points out the limitations of fogra51 & fogra52 and the benefits of the presented approach, if the proof substrate and the production substrate show a varying amount of FBA.

Still broad tolerances of ISO 3664:2009 become irrelevant if both, the proof as well as the production substrate, possess the same FBA amount. This is nearly perfectly fulfilled with this study method. Therefore, the approach of this study offers even the possibility to view a proof and a production substrate under any illumination, without the risk of obtaining FBA induced colorimetric differences.

However, only one single FBA type has been examined, as well as only one single carrier type. Other FBA types and carriers and their combinations are expected to even further extend the FBA emission intensity. Other typical ingredients, i.e. biocides, corrosion inhibitors, thickening agents or detergents, and their interaction with FBAs are included in a recently started research project. Furthermore, no results concerning the light fastness of the used FBA (and other possible FBAs) have been presented. Generally, it is stated that the light fastness of the used FBA is very limited. Anyhow, no detailed research is available in this context. All named aspects will be investigated and discussed in future studies.

Future research will also focus on the performance of the approach in combination with ink printed on it and therefore on the overall performance in proof processes. It is expected to achieve a much better match between printed proof- and production prints, again completely illumination independent.

5. Conclusion

As pointed out in chapter 1.3 State of the scientific knowledge, the effects of FBAs cannot be compensated effectively for every possible color management scenario, until now.

Therefore, the authors of this study propose a method to compensate FBA induced colorimetric differences in proof processes by printing defined amounts of liquid FBAs using inkjet printing. Thereby, the intensity of the fluorescence is directly controlled by changing the area coverage of the FBA printed on a unbrightened or slightly brightened proof substrate. As shown in chapter 3.2.2 Influence of FBA area coverage in combination with FBA concentration, an undesired greening effect can be observed, already for 5 % FBA concentrations, depending on the used substrate. Furthermore, in chapter 3.2.2 it is pointed out, that the emission intensity strongly varies depending on the used substrate. Both effects are believed to be related to varying amounts of OH groups of the substrates.

Therefore, in chapter 3.3 Fluorescence emission of ink compositions with carrier, a fixed FBA concentration of 5 % in combination with the substrate ‘EFI 3170’ has been used as a base for analyzing the impact of polyethylene glycol as a carrier on the emission intensity for several FBA area coverages. The results in this chapter clearly show that the addition of 5 % polyethylene glycol to a 5 % FBA mixture not only prevents greening effects but also helps to significantly increase the FBAs emission intensity, and therefore, efficiency.

In chapter 3.4 White point simulation of production substrates, an actual white point simulation is presented and exemplarily done for the highly brightened production substrate ‘Heaven42’ and the proof substrate ‘EFI 3170’. It is successfully shown that a two-step spectral fit can be used to achieve a very good correlation between the production substrate ‘Heaven42’ and the proof substrate ‘EFI 3170’ of $\Delta E_{00} = 0.89$.

Summed up, it is shown that applying an FBA with variable area coverage can be used to create typical FBA emissions and intensities using any desired slightly brightened or unbrightened substrate to simulate any production substrate as long as the overall reflectance of the proof substrate is equal or higher compared to the reflection of the production substrate.

Overall, it can be stated that the presented approach leads to a significantly improved white point simulation for any proof substrate, by taking the actual FBA emission of any given brightened production substrate into account, which makes proofing processes illumination independent.

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