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## Study of the effects of the ink layer on selected properties of multilayer packaging films

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### Abstract

Application of multilayer films in packaging allows the reduction of the average weight of packaging and a selection of its optimal barrier properties. This kind of material is particularly important for the development of packaging. The aim of this research was to determine the influence of the ink layer thickness on the mechanical properties of interlayer printed laminates. Studies were performed for a double-layer film (poly(ethylene terephthalate)/glue/biaxially oriented polypropylene) prepared by an adhesive method using a two-component, solvent-free adhesive. The bond strength of the laminates and the thickness of each layer were measured. It was found that the thickness of the fixed ink, interlayer printed on the reverse side of the PET film, has a significant influence on the mechanical properties of the laminates. Regardless of the ink film thickness, the values of the bond strengths of the printed laminate are significantly reduced.

**Keywords:** laminate, double-layer film, interlayer printing, bond strength

### 1. Introduction

Despite the fact that plastic packaging on a European scale represents only 17 % of the weight of all packages, they are used for more than 50 % of consumer products on the market (PlasticsEurope, 2014). Furthermore, flexible plastic packaging sales grew at an annual rate of 4 % during 1999–2003. In recent years, the fastest growth rates (14 %) were observed on the Eastern European markets. The Russian market has been the fastest growing one, although the USA is the world's largest market. The next largest markets in 2008 were: Japan, UK, Italy and Germany (World Packaging Organisation, 2008). In Europe, packaging applications is the largest application sector for the plastics industry. In 2012, it represented 39.4 % of the total plastics demand (PlasticsEurope, 2013). The most important plastics types are: polyethylene, including low density (PE-LD), linear low density (PE-LLD) and high density (PE-HD), polypropylene (PP) and poly(ethylene terephthalate) (PET).

High efficiency has been achieved in the production of plastic packaging, which allows the reduction of the average weight of packaging. Over the past 10 years this has been reduced by approximately 28 % (PlasticsEurope, 2014). Among other reasons, this was possible through the use of multilayer films, so called laminates.

A proper selection of films for laminate composition allows the achievement of the desired barrier properties and characteristics, such as heat sealability, reduced weight of the packages, etc. On the market, there is no ideal material, but joining the various properties of plastic films (polyethylene, polypropylene, polyesters, polyamides, etc.) or combining them with paper or aluminium film makes it possible to approximate an optimal material, which is a guarantee for a modern packaging with all its required functions.

A literature review leads to the conclusion that research on multilayer film is particularly important for the development of packaging. The number of available publications is still limited and they cover just a part of the subject. Several patents describe novel compositions of multilayer films (Berlin, Bentmar and Flemmer, 2005; Toshiyuki et al., 1996). On a general level, Wagner (2010) describes various problems of multilayer films for use in packaging. Kao-Walter et al. (2004) and Bjerken, Kao-Walter and Stahle (2006) have studied the mechanical properties of a laminate in relation to the kind of materials and the adhesion level. Hare, Moyse and Sue (2012) have published a new methodology for the study of the resistance of laminates to destructive scratching. Taniguchi, Sasaki

and Kitamura (2003) have described the possibility of producing multilayer stretch compositions. Quite a large number of publications describe the results of research on the migration of substances from packaging into the packaged food (e.g., Aznar et al., 2011; Ellendt, Gutsche and Steiner, 2003; Lawson, Barkby and Lawson, 1996).

## 2. Methods

The aim of the study was to determine the influence of the ink layer thickness on the mechanical properties of interlayer printed laminates.

Studies were performed for a double layer film prepared by the adhesive method using a two-component, solvent-free adhesive. Liofol 2K (Henkel) polyurethane adhesive was used. Two kinds of films were selected: polyester film (poly(ethylene terephthalate)) and polyolefin (biaxially oriented polypropylene, OPP) (see Table 1). PET/glue/OPP laminates were prepared for the tests in the laboratory.

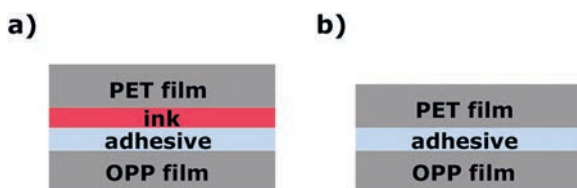


Figure 1: The structure of the laminates used for the tests, a) printed, b) unprinted

The preparation of the laminates included printing on PET film and lamination of the reverse side printed PET film with clear OPP film (Figure 1). Two inks were used for printing: magenta and black (see Table 2). These were special printing inks dedicated to interlayer printing of laminate. Printing was performed using a handheld printer (K Control Coater) with rod applicators. Five rods with different ink transfer to the substrate were used (see Table 3). Then, the printed sheets were laminated with a second film. Constant thickness of the adhesive was achieved by using an appropriately selected rod applicator (number 3).

Five samples of interlayer printed laminates based on each ink colour and with 5 ink film thicknesses were prepared. Multilayer films with the same structure were also produced without printing. The prepared laminates

In contrast to studies presented in the literature, the studies presented in this paper concern printing on multilayer films. Multilayer printing has a significant effect on the strength of laminates as well as on its other functional characteristics. This topic is clearly very important due to the fact that more than 90 % of the packages on the market are printed.

were left until adhesive cross-linking was complete and full bond strength was achieved. After more than 14 days, their mechanical properties were tested and a microscopic analysis was performed.

The bond strength between two films was analyzed using a Zwick tensile tester FB010TN.D30 BT1. Samples for the delaminating process were prepared according to standard (International Organization for Standardization, 1995). Strips of 15 mm width were cut. Their edges were smooth and without damage. As the first step, the laminate strips were partially pre-delaminated. For this purpose, they were dipped into Chesoll for 15 minutes. Then they were mounted in the tester and subjected to the tear and peel test. The tester settings were as follows: the initial distance between the holding jaws – 50 mm, test distance – 100 mm, test speed – 100 mm/min.

At the end, the structure of the prepared laminates was examined using an Olympus optical microscope.

Table 1: Characteristics of the films

	Film	
	OPP	PET
Film name	BIFOL BG	BOPET CA
Producer	Flexpol	Flexpol
Thickness [ $\mu\text{m}$ ]	20	12
Orienting type	biaxial	biaxial
Surface activation	yes	yes
Roll width [mm]	560	560
Density [ $\text{g}/\text{cm}^3$ ]	0.9	1.4
Grammage [ $\text{g}/\text{m}^2$ ]	18.2	16.8
Haze [%]	2	4
Gloss 45°	90	–

Table 2: Characteristics of the inks

Ink	Ink name	Type	Producer	Light fastness
Black	Process Magenta Raster	F-62GSR35206-0301	Michael Huber	6
Magenta	Process Black Raster	F-69GSR66006-0301	Michael Huber	6

Table 3: Characteristics of the rods

Rod number	Wound type	Wire diameter [mm]	Wet film deposit [ $\mu\text{m}$ ]
1	close	0.05	4
2	close	0.08	6
3	close	–	10
4	close	0.15	12
5	close	0.31	24

### 3. Results

In this work the influence of interlayer printing with different kinds of ink on mechanical properties of the laminates were studied. The results of the delaminating tests for unprinted multilayer films are shown in Figure 2.

An analysis of microscopic images made it possible to measure the thicknesses of the individual layers of the composition of the film: the films, the fixed adhesive and the ink layers. This knowledge may help to better understand the impact of the thickness of the ink

layer. A sample photo of the structure of the interlayer printed laminates is presented in Figure 3.

The results represent the bond strength of 5 samples. All tested strips came apart almost immediately after the beginning of the tear and peel test (see c) in Figure 4). Although the test distance was set up at 100 mm, the samples were fractured after just a few mm of the tear and peel process (see Figure 5). This observation indicates the high strength of the laminates and their good quality.

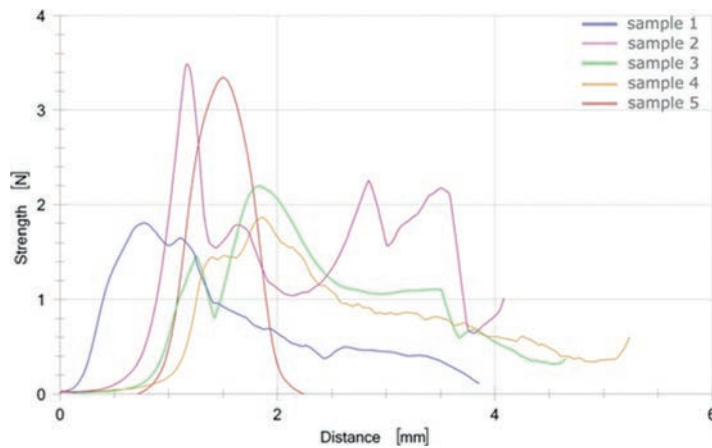


Figure 2: Bond strength of unprinted laminates

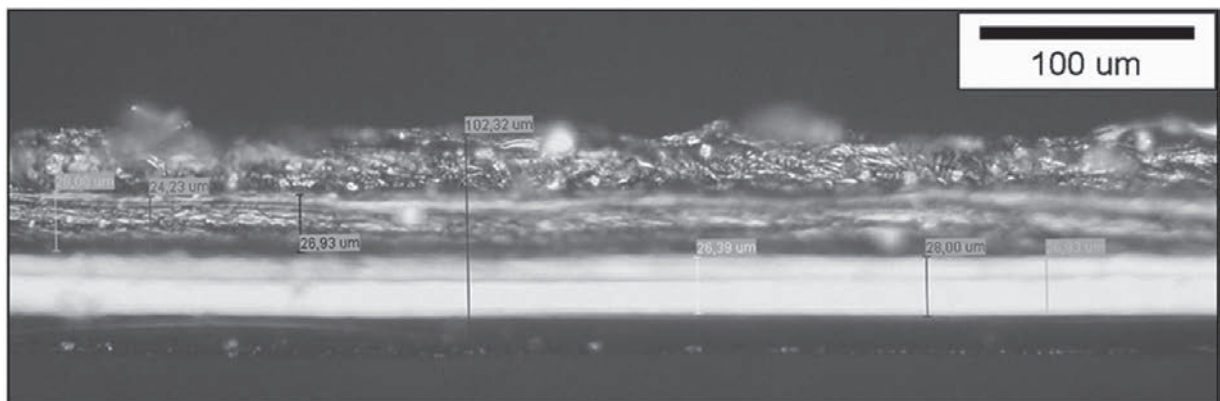


Figure 3: Sample photo of the structure of interlayer printed laminates

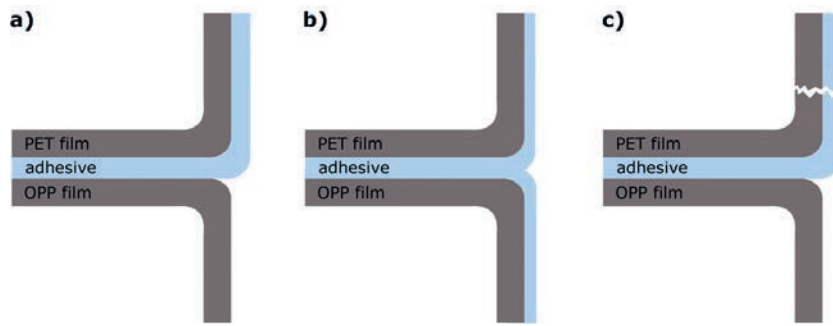


Figure 4: Types of bond failure during the tear and peel test a) adhesive bond failure, b) cohesive bond failure, c) film breakage

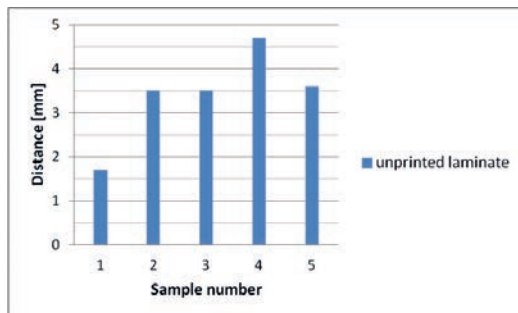


Figure 5: The distance between the tensile testing machine jaws after which the samples of unprinted laminates were broken

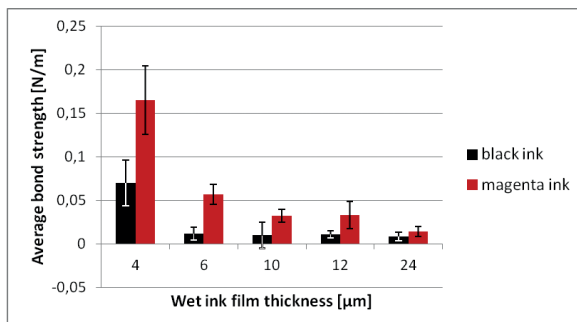


Figure 6: Influence of the ink film thickness on the average bond strength of interlayer printed laminates

The bond strength of the interlayer laminates was determined. The results received for multilayer films printed with both kinds of ink and different ink thicknesses are presented in Figure 6.

The ink layer has a negative impact on the bond strength of the laminates. The ratio ( $P$ ) of the maximum bond strength of interlayer printed laminates to the maximum bond strength values obtained for unprinted laminates is shown in Figure 7.

$$P = \frac{\max \text{Bond Strength of interlayer printed laminates}}{\max \text{Bond Strength of unprinted laminates}} \cdot 100\%$$

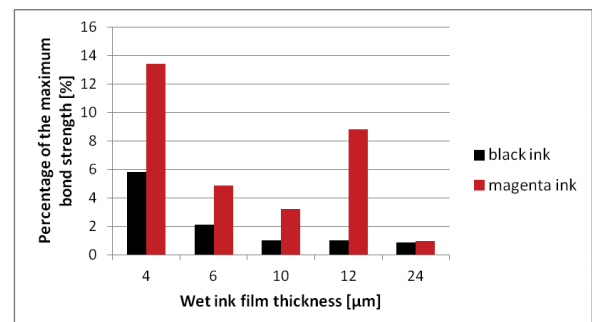


Figure 7: Changes in the maximum strength of interlayer printed laminates in comparison to unprinted ones

#### 4. Discussion

The bond strength of laminates with and without interlayer printing was analyzed. In the case of laminates without interlayer printing, all tested samples came apart, and the value of the maximum bond strength was more than 3 N/m for 2 stripes and approximately 2 N/m for the rest of samples (see Figure 2). The value of the average bond strength was consistent with data suggested in the literature. For all laminates, the PET film was broken during the tear and peel test. In addition, the distance between the tensile testing machine jaws after which the samples of unprinted laminates were broken (Figure 5) was lower than 5 mm, when the maximum distance of the test was 100 mm. This indicates that the quality of

the laboratory prepared laminates was very high and that the adhesion of the adhesive to both films was good, as well as that the polyurethane adhesive was properly chosen. Furthermore, the substrate strength was lower than both the adhesive and the cohesive bond strengths.

For both kinds of printed laminates, the bond strength was significantly lower than for the unprinted composition, regardless of the choice of magenta or black flexographic ink or the thickness of the applied layer. All samples were delaminated and two kinds of failure were found. Adhesive bond failure was observed in most cases. Cohesive bond failure occurred usually together

with the adhesive one, appearing separately only in very small number of cases (Figure 4). During the delamination process, the fixed ink layer was transferred from the PET film to the OPP film for almost all samples. This means that the adhesion of the ink film to the printed substrate (PET film) was lower than adhesion forces between ink film and adhesive. This also indicates high cohesive forces within the fixed ink layer.

The thickness of the printed ink layer has a significant effect on the bond strength of laminates. With an increase in the thickness of the ink layer, the bond strength decreases rapidly. The highest values of average bond strength for the interlayer printed multilayer film were obtained with a rod applicator that allows for the wet ink layer a theoretical thickness of 4 µm (see Figure 6). However, even in this case the bond strength values were much smaller than for unprinted laminates. The values of the maximum bond strength obtained for interlayer printed laminates were 6 and 14 % of the values determined for unprinted multilayer films, for black and magenta ink, respectively (see Figure 7).

## 5. Conclusions

The research found that the thickness of both the adhesive and the fixed ink layer has a significant influence on the properties of the packaging material. This impact is not limited only to the possible migration of compounds hazardous to health and flavour into the product, but has a negative influence on the aesthetics of the package,

The type of ink used determines the mechanical properties of the laminates. The values of the average bond strength for the laminates printed with magenta ink were higher than for those printed with black ink. The magenta ink film thickness had a significant impact on the bonding strength of the laminates. The ink film thickness is not as important in the case of black ink, but the influence can also be observed (Figures 6 and 7).

The microscopic analysis shows that the settled layers of ink and adhesive were of fairly uniform thickness for all the laminates (see Figure 3). This confirms the good quality of the laboratory preparations and shows that no uneven layers were produced in the lamination process. Furthermore, the study found that the fixed layers of ink and adhesive were of much larger thickness than expected. This result suggests that the wet layers produced by the rods were several tens of percent thicker than what the rod designation specified. This did not have an important influence on the results, because the order of ink layer thickness was maintained and the experiments covered a wide range of ink thickness values.

defined as the quality of the print and the quality and durability of the lamination. The thickness of the ink layer in interlayer printed double-layer films determines their bond strength in the delaminating tests. Regardless of the thickness of the ink layer, the values of the bond strength of printed laminates are significantly reduced.

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