Printability of PE-coated paperboard with water-borne flexography: Effects of corona treatment and surfactants addition

By B. Mesic, M. Lestelius, G. Engström and B. Edholm

Abstract: The influence of corona-treatment and surface roughness on print quality was studied for two polyethylene coated paperboards of different surface roughness, printed with a water-borne flexographic ink. The surface energy of the ink was varied by addition of a surfactant. The results illustrate the significance of the surface roughness on the print quality and the significance of the corona-treatment on the rub resistance. The surface energy of the ink did not affect the print quality.

With the advent of polyethylene (PE) onto the marketplace following World War II, many changes occurred in both the plastics and the converting industries. Low-density polyethylene (LDPE) is widely used in various applications due to its abundant supply, low cost, good processability, low energy demand for processing, and its resistance to chemicals and harsh environments. Currently, the packaging industry demands LDPE because of its high specific modulus and strength; it is used directly or in the form of laminates with paper, aluminium foil, etc. [4]. However, its surfaces are chemically inert and it is non-porous and hydrophobic, and it needs to be made more hydrophilic in nature to improve printability, paintability, or adhesion to adhesives or inorganic substances [4,11]. The printability of PE depends on the PE surface. Satisfactory printing is most problematic with water-borne inks, included flexographic inks on non-porous, low surface energy substrates such as polymeric films. Proper wetting and adhesion, in particular, are necessary [12]. The surface energy affects the adhesion of the ink to the substrate. It also affects the way the ink spreads on the film surface [9].

Some modification of the PE surface characteristics without altering the bulk properties of the material is required. One of the processes most commonly used in industry to modify PE surfaces is corona treatment (CT). CT is the application of a high-energy electrical discharge onto a substrate as it passes through a capacitor or treating unit [8]. Precisely what the treatment does to a surface is still the subject of much discussion and controversy [10]. A rough general description of this treatment is that, in the CT system, the voltage build-up ionizes the air in the air gap creating a corona, which increases the surface tension of the substrate passing over the ground roll [11]. Pure polyethylene with no surface oxidation has a low surface energy (slightly less than 31 mJ/m²) that will not support ink adhesion [8]. Corona treatment is one way of increasing the surface energy of the PE.

The equilibrium surface tension of water-borne inks ranges from 20 to 40 mJ/m² [6]. To reduce the surface energy of the ink, a surfactant may be used. A surfactant is generally a small molecule that, when added in low concentrations to an aqueous solution or suspension, reduces its surface energy [1,7]. In practice the surface energy of the solid phase should be 10 mJ/m² higher than the surface energy of the liquid [11].

In the present study, the influence of the surface roughness and the surface energy of PE-coated paperboards and the surface energy of the water-borne ink on the printability were studied using a laboratory-scale flexographic printing press (IGT F1). Printing trials were performed on PE-boards with different surface energies and different surface roughness. Printability was assessed with respect to print density, white areas, dot gain, mottling and rub resistance of the water-based ink. In addition to the printing trials, the spreading of the ink on the surface was studied by contact angle measurements.

Material
Ink
Printing was performed with a water-borne ink, Cyan 707-44024 SCANBRITE RASTER B 60750 supplied by Sun Chemical. The viscosity of the ink was 30 s, according to a Zhan 2 cup. The ink was used in two ways: as a standard ink (SI) and as an ink with the addition of 1% Surlynol 420 surfactant (Air Products and Chemicals, Inc., Netherlands). Surlynol was added in order to reduce the surface energy level of the ink. The surface energy of the SI was 29.8 mJ/m² and that of the ink with Surlynol addition was 27.2 mJ/m². With only 1% Surlynol, the viscosity of the ink was same as that of standard ink. The ink with Surlynol is here-after referred to as modified ink (MI).

Substrate
Two types of substrate were used: a commercial extrusion-coated (CEC) and corona-treated PE-board and a laboratory extrusion-coated (LEC) and corona-treated PE-board. PE extrusion coating on both types was done on basically the same commercially produced Coated Kraft Back (CKB), with a grammage of 205 g/m². The PE layer was 19g/m² for LEC and 20 g/m² for CEC. The surface energy of the PE boards determined directly before printing using dyne test ink was approximately 31 dyn/cm (mJ/m²) for untreated (UT)
samples and 44 dyn/cm (mJ/m²) for corona-treated (CT) samples.

METHODS
Grammage and surface roughness
Grammage was determined according to SCAN-P 6:75. For the determination of surface roughness a profilometer (Perthometer C5D from Perthen, Germany) equipped with a narrow diamond needle, FRW-750 (radius 10 μm), was used. The needle was in contact with the surface and the z-directional position of the surface was determined mechanically with an iron core differential transformer Perthometer PRK, (Perthometer PRK from Perthen, Germany). Profilometer analysis was performed in a square of the test sample. A 58 x 60 mm area of each surface was analyzed and the test results were presented as the average roughness, Ra.

Laboratory printing
All printing was performed using an IGT FI equipped for flexographic printing (IGT Testing Systems, Amsterdam, NL). Three different printing plates (DuPoint ACE, produced by Flexopartner AB, Sunne, Sweden using straight light illumination) with 100%, 70% and 30% tone values were used. For the evaluation of the ink distribution on the surfaces, only a printing plate with 100% tone value was used. The screen ruling of the printing plate was 24 lines/cm, the thickness was 1.70nm and its hardness was 71° shore A. The anilox roller had a screen ruling of 180 lines/cm, with an ink volume of 2.7 \( \frac{mL}{cm²} \) and a nip line load of 150 N and the printing speed was 0.3 m/s. For each type of substrate, 6 samples were printed and evaluated, and results were presented as the average values of the 6 evaluated samples. The errors are expressed as standard deviations.

Measurement of print density and dot gain
An optical densitometer (Gretag Macbeth D19/17B/P, Regnondorf, Switzerland) was used, connected to a computer for data acquisition. The dot gain was calculated using the Murray-Davies equation, which gives the percentage dot area in density-based measurements as follows:

\[
FD(\%) = \frac{1 - 10^{-DR}}{1 - 10^{-DF}} \times 100\% \quad (1)
\]

where,

\[
D_R: \text{Print density of the halftone print.}
\]

\[
D_F: \text{Print density of the solid area (100% tint).}
\]

\[
F_P: \text{The percentage dot area.}
\]

The dot gain \( Z \) was calculated as the difference between \( F_D \) and the nominal percentage dot area on the printing plate \( (F_P) [3] \).

\[
Z(\%) = F_D(\%) - F_P(\%) \quad (2)
\]

Measurement of non-printed areas (white dots)
The white dots on solid areas were evaluated with the image analysis software MOFFE (developed by Stora Enso Research, Falun, Sweden) developed for cyan ink. Images were captured by a digital camera. The software analyses and compiles the size of the non-printed areas, defined as white dots. Defects in the range from 0.04 to 0.9 mm\(^2\) were taken into account by the software.

Measurement of print mottle
Print mottle was evaluated by image analysis with software developed to quantify the degree of mottling, STFI Mottling v 2.4 software (developed by STFI, Stockholm, Sweden). This analysis was carried out on all the printed surfaces (one sample per surface with a size of 3 by 5 cm). For imaging, a flat bed scanner (AGFA Digital T2500) was used and images were captured with a resolution of 300 dpi. Print mottle analysis was based on an FFT (Fast Fourier Transform) technique and was related to the grey tone value calibrated by reflectance. Information in different spatial wavelengths, 1-8 mm, was calculated from the FFT-spectrum. Print mottle was expressed as the coefficient of variation of the grey tone in the non-filtered image.

Rub test (abrasion resistance)
Rub resistance was assessed according to ASTM D 5264 - 98 method. All the printed samples were tested 45 days after printing. Setting parameters were as follows: number of strokes (a stroke is one back-and-forth cycle) was 7, block weight was 1860 g and motor speed was 2, (speed 2 = 0.7 cycle/s). To quantify and compare rub resistance, the mottling software was used. The result was expressed as the difference in the coefficient of variation (COV) of mottling before and after performing the rub test. An example of a sample before and after the rub test is shown in Fig 1.

Contact angle
Contact angles for ink were measured on the TS of the PE-extruded paper during contact times from 0 to 2 s, with a contact angle measurement apparatus (FTA 200, First Ten Angstroms). The contact angle goniometer is equipped with a horizontally aligned microscope and digital high speed camera to capture the evolution of the drop-surface interaction. The initial period after triggering was 0.033 s with a post-trigger period multiplier of 1.166. The drops were dispensed by touching (i.e. the substrate was raised to touch the bottom of the hanging drop) with an automatic stepper-motor-driven syringe pump.
The syringe volume was 5mL and needles with 22 gauge diameters were used. The measurement apparatus also allows the drop base diameter to be determined.

RESULTS AND DISCUSSION

Measurement of surface roughness
The surface roughness of the LEC sheets increased after CT. The average surface roughness (Ra) for the LEC CT sheets was 0.64 µm, and for the LEC UT sheets 0.54 µm. For the CEC sheets, no statistically significant differences were observed between CT and UT sheets. Ra for both types of CEC sheets was 0.43 µm.

Ink wetting
Figures 2 and 3 show the contact angle and the drop base diameter for a drop of ink on LEC and CEC samples. Despite the difference in surface roughness, no statistically significant difference was observed between the LEC and CEC samples. This may be due to the greater size of the drops used in contact angle measurement (compared to individual half-tone dots). This is also true considering the different surface energy of the ink. In the view of the surface energy level for paperboard samples, differences were observed between corona-treated samples (i.e. relatively high surface energy level) and samples without corona treatment (i.e. relatively low surface energy level). Corona-treated samples showed a faster initial reduction in the contact angle than samples without corona treatment, Fig. 2. These differences were apparent already within 0.3 s. It can be seen, in Fig. 3, that the base diameter of a drop of ink on the surface of corona-treated samples was larger than the base diameter on untreated samples. However, the base diameter did not increase as much or as fast on samples without corona treatment as on the corona-treated samples.

Influence of ink distribution on print density
The ink distribution was evaluated by a four times repeated printing on the same sample. Corona treated samples with Ra = 0.43 µm (CEC) and Ra = 0.64 µm (LEC) were printed with MI ink. The print density ($D_p$) and ink amount ($y$) g/m² were measured immediately after each printing run. Results are presented in Fig. 4 as a plot of $D_p$ versus $y$. It is evident that during the first printing run more ink was transferred to LEC samples than to CEC samples, but that the amount of ink transferred during the second, third and fourth printing runs was roughly the same. Nevertheless, CEC samples gave a higher $D_p$ than the LEC samples. That is, for a given amount of transferred ink the CEC samples gave higher $D_p$ than the LEC samples. The CEC samples also gave a higher print density maximum ($D_{p1}$) than the LEC samples.

These results suggest that the higher surface roughness can improve ink transfer to the printing surface [14], and also that a high amount of ink (ink amount g/m²) on the surface does not always lead to a high print density. A high degree of surface roughness can probably cause an uneven ink distribution and uncovered areas (white areas) in the printed surface, resulting in high print mottle and uneven print density, which bring out a lower mean print density.

The dependence of the print density, $D_p$, on the ink distribution (ink amount g/m²) on the substrate may be described by a schematic model, a model that we call the “square wave ink distribution model”, Fig. 5a, and by the Tollenaar-Ernst equation [5, 15]:

$$D_p = D_{p1}(1–e^{–m})$$  

where $D_p$ is print density, $D_{p1}$ is the print density maximum or saturation print density, $y$ is ink amount on paper (ink amount g/m²) and m is a parameter for the steepness of the print density curve.

We assume in Fig. 5a that the ink is distributed on the P Ebord surface according to: $y_1$ for the smoother surfaces; $y_2$ for the rougher surfaces. $y_1$ exhibits small variations in the ink amount, whereas $y_2$ shows larger variations in the ink amount. Irrespective of differences in ink layer variation, both surfaces can have the same average ink layer ($\bar{y}$) as indicated in Fig. 5a and Table I.

If using the Tollenaar-Ernst equation, Fig. 5b, we calculate the $D_p$ at points A and B, Fig. 5a, the values presented in Table I are obtained.

These results show that the unevenness of the ink layer on the surface may result in a variation in the $D_p$ for a given ink amount. The same average ink amount $\bar{y} = \bar{y}_2 = 2.0$ g/m² on the two different surfaces, results in different print densities. $D_{p1} = 3.06$ and $D_{p2} = 2.39$ respectively. It may be due to high surface

**TABLE I. Ink amount $y$ (g/m²) and print density, $D_p$.**

<table>
<thead>
<tr>
<th>Point of measuring</th>
<th>Ink amount, $y$ (g/m²)</th>
<th>Print density, $D_p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>2.35</td>
<td>3.17</td>
</tr>
<tr>
<td>B1</td>
<td>1.65</td>
<td>2.83</td>
</tr>
<tr>
<td>A2</td>
<td>3.50</td>
<td>3.39</td>
</tr>
<tr>
<td>B2</td>
<td>0.50</td>
<td>1.38</td>
</tr>
<tr>
<td>Average (A1+B1)/2</td>
<td>2.00</td>
<td>3.00</td>
</tr>
<tr>
<td>Average (A2+B2)/2</td>
<td>2.00</td>
<td>2.39</td>
</tr>
</tbody>
</table>
roughness (as results show in our case). Probably, it is difficult to achieve an even ink distribution on rougher surfaces, and fill the pits.

Print evaluation
Both full-tone areas and half-tone areas (70% and 30% tone value) were measured, Fig. 6. The CEC gave a higher print density than the LEC in the full-tone areas, Fig. 6, which may be due to better ink coverage, as already discussed, cf. Fig 5. In the 70% and 30% tone values, no great difference was seen between CEC and LEC samples. With respect to the different surface energy levels produced by CT, the full-tone areas showed no great difference between CT and UT samples, Fig. 6. In the half-tone areas, with the exception of the CEC sheets in the 70% tone value, the corona-treated sheets printed with Surfynol modified ink (CT/MI) gave a significantly higher print density than the untreated sheets printed with untreated standard ink (UT/SI). However, in Fig. 6, (except for LEC sheets in full-tone areas) a weak trend of decreasing print density with decreasing difference in surface energy between ink and substrate was observed. Corona treating improves ink adhesion and ink spreading on the surfaces, and this is a probable explanation of the higher print density, as was mentioned above. No significant difference in print density was observed between MI and SI on CT and UT sheets. It is not clear whether the addition of Surfynol to the ink to reduce the surface energy improves the print density.

Dot gain was calculated from the print density according to equations 1 and 2 [3] and the results are presented in Fig. 7. High values of dot gain were obtained in all cases. No great difference was seen between CEC and LEC sheets, except for the decreasing on the LEC sheets. One reason for the greater differences in dot gain on the LEC sheets could be the higher surface roughness, in addition to the difference in the spreading of the ink. Untreated sheets with lower surface energy levels, printed with standard ink (UT/SI), gave the lowest dot gain. This was expected due to the high energy level of the ink and the low energy level of the substrate. A higher surface energy level increases the spreading of the ink on the surfaces, resulting in a higher dot gain.

Figure 8 shows the percentage white areas (white dots) in the full-tone areas. A visual examination also clearly indicated that the number of unprinted regions was higher on the LEC than on the CEC samples, which is also in accordance with the results of the ink distribution analyses. This may be due to bad ink coverage caused by the higher roughness of the LEC sheets, cf. Fig 5. Regarding the different surface energy level of the LEC samples, the UT/SI sheets gave the lowest percentage of white areas. On the printed CEC sheets, no difference was observed. However, both LEC and CEC sheets gave sufficiently low percentages of white areas to be accepted as good ink coverage (according to commercial practice applied by the packaging industry). Considering the combination of ink and substrate surface energy, the UT/SI on the LEC samples gave slightly, although significantly, lower percentage of white areas. For the CEC samples, no difference was observed between the modified (MI) and standard ink (SI).

Mottling in the 1 to 8 mm wavelength region in full-tone print areas is shown in Fig. 9. The print mottle was higher on the LEC samples than on the CEC samples, which is in accordance with the results of the print density evaluation, i.e. a less rough surface generates a more homogeneous ink film. The combination of corona treatment and surfactant-modified ink on CEC, the CT/MI, gave significantly lower mottling than that on the CT/SI, UT/MI and UT/SI sheets. Mottling measurements in this size range (1-8 mm) give results quite similar to the “white areas” measurements, since the size scale of the actual area on which the measurement is performed is roughly the same, and both methods measure grey scale intensity variations. It appears as though the surface energy of the ink has the greatest influence (gave lower mottling), not the corona treatment. This tendency was not observed on the LEC samples. None of the differences between the LEC samples

FIG. 5. Schematic description of print density dependence on the ink distribution of the PE-board:
  a) “Square wave ink distribution model”
  b) Print density as a function of ink amount according to the Tollenaar-Ernst equation with $D = 3.5$ and $m = 1$.

FIG. 6. Print density for LEC and CEC sheets in full-tone area, 100% tone value and in half-tone area, 70% and 30% tone value. Error bars indicate standard deviations.
were statistically significant. One possible explanation of this behaviour may be the greater roughness of the LEC samples, resulting in a poorer ink coverage and higher degree of white dots. With respect to the surface energy of the ink, the standard ink on the untreated CEC sheets (UT/SI sheets) gave a higher mottling than other combinations on the same substrate. It may be explained as bad ink spreading on the surfaces, caused by high surface tension of the ink and low surface energy of the substrate. On the LEC samples no differences between sheets were observed, probably due to the roughness of the samples.

**Rub resistance**, expressed as the difference in mottling before and after the rub test, is shown in Fig. 10. The print mottle after rub test was substantially higher for UT sheets (UT/MI and UT/SI) than for CT sheets (CT/MI and CT/SI) on both the LEC and CEC samples. This means that corona-treated sheets gave a higher rub resistance than untreated sheets, which is in accordance with common practice in the industry today, i.e. PE coated paperboard must be treated to produce sufficiently good ink rub resistance. The high surface energy level of 44 mJ/m² promotes adhesion and a uniform spreading of the ink on the surface, and that is a probable explanation of the observed increase in rub resistance. No differences due to different surface energies of the ink (MI vs. SI) were seen. Ink spreading (dot gain and white dots) was apparently affected by the surfactant addition, but long-term ink adhesion was not.

Results of the ink wetting, ink distribution and printability tests, Figs. 2 - 10, show that the application of corona treatment to increase the surface energy level gives better and faster ink spreading, resulting in a better print quality, especially for full-tone surfaces [11]. At the same time, it can have a detrimental effect on the print quality, since the increased spreading results in a relatively high dot gain. This is consistent with wetting and adhesion theory [2,13] and opens the window for matching surface characteristics to achieve the desired print quality.

**CONCLUSIONS**

It is evident that corona treatment can be used to achieve improved ink adhesion and ink spreading on a PE surface. The commercial extrusion-coated samples, both corona-treated and untreated, gave a better flexographic printing result in terms of print density, white dots and mottling in full tone printing than the laboratory extrusion-coated samples. Results from evaluation of ink distribution on the PE surfaces showed that a high amount of ink on the surface does not always lead to a high print density. This substantiates the argument that substrate roughness can influence print quality. The better print quality was more evident for corona-treated samples. Whether or not the surfactant-modified ink gave better print quality is not clear in these results, i.e. no significant difference was observed between Surlynol-modified and unmodified ink.

Contact angle measurements of the ink on the PE surfaces showed that corona-treated samples gave a faster initial reduction of contact angle and a larger base diameter for a drop of ink. The faster initial reduction of contact angle and base diameter for a drop of ink brings better and faster ink spreading and gives better print quality in full-tone surfaces, but at the same time leads to higher dot gain. This seems to indicate that a mismatch of surface energetics between substrate and ink can negatively influence the print quality. The estimation of the spreading of ink on PE surfaces can be very important for
understanding the interaction with water-based printing inks on the same surface. By optimizing extruding, pre-treatment and printing parameters, a better printing result can be achieved.

ACKNOWLEDGMENTS
The authors are grateful to Mr. Jan Lewau (Trioplast Landskrona AB), Ms. Isabel Knöös and Mr. Claes-Göran Thorén (Stora Enso Research), Mr. Dan-Erik Linden (Stora Enso Packaging Boards), Mr. Thomas Petersson (Sun Chemical), Mr. Ulf Ericsson (Fors Mill Hammarby) for valuable help with the experiments. The financial support from the Institute for Research and Competence Holding AB (IRECO Holding AB) is gratefully acknowledged.

LITERATURE
4. SOO-JIN, P. Effect of Corona Discharge Treatment on the Dyeability of Low-Density Polyethylene Film.