Evaluation of Flexographically Printed Conductive Traces on Paper Substrates

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Abstract

Results of print trials of different silver-based conducting inks on different paper and paperboard substrates are presented. Printability (based on optical appearance) and electrical properties are analyzed for their dependence on substrate properties. In particular, it was observed that traces printed with solvent-based inks have better print quality compared to water-based inks at higher anilox cell volume. However, sheet resistivity of traces printed with water-based inks were less than those achieved with solvent based inks for the same printing conditions, probably due to higher solids content and smaller particle size. Bulk resistivity was observed to be less for traces printed with 100% tint due to better ink coverage and higher ink film thickness. The results indicate that printing of conductive components on paper substrates is a viable option for low cost electronics.

Antenna performance was seen to be slightly better for water-based inks than for solvent-based inks. However, acceptable antenna performance was obtained for both inks on all substrates.

Introduction

Printed electronics have gained significant importance, due to the convergence of printing, electronics and materials science (Bartzsch, 2006). Techniques used today for electronics manufacture are slow and have limited production volumes (Sangoi, 2005), due to their use of batch processing. High volume printing processes (Fleming, 2007,) are currently being investigated for their potential use in printing of electronic devices, such as RFID antennae.

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Electronic features printed on plastic substrates have led to low cost devices with the elimination of complex manufacturing procedures (Sangoi, 2004). Printing techniques such as screen-printing (Bao, 1997) and inkjet printing (Bharathan, 1998, Hebner, 1998a, b) have been widely explored for printed electronics. Due to the limitation of speed, volume of production, and limitations in obtaining the resolution required for printed electronic features and layer-tolayer deposition accuracy for the above techniques, high volume printing techniques such as flexography (Kattumenu, 2008a, b, Mäkelä, 2005, Yu, 2005) and gravure (Hrehorova, 2007, 2008, Rebros, 2008, Pudas, 2005) currently have attracted much interest for their potential use in printing of RFID tags.

The focus of our recent research has largely been on printing RFID tags on paper-based substrates (Fleming, 2007, Hrehorova, 2007, 2008, Kattumenu, 2008a, b, Rebros, 2008). Why paper? Paper and paperboard are widely used for packaging (Chas-Amil, 2000), most of which are being printed with steadily increasingly complex designs. Therefore, printing the RFID tag directly to the packaging materials can improve efficiency and reduce the cost of the tag to single items levels (Sangoi, 2005).

In this work, flexographic printing has been evaluated as a suitable method for printing conductive traces on paper (4 papers denoted by P1-P4) and paperboard (3 paper boards B1-B3 along with the backside of B3, B3 bs) substrates. The effect of substrate properties in characterizing print attributes and electrical resistance has been studied. A summary of some of the properties of these paper based substrates is given in Table 1. The roughness of the substrates was measured with an Emveco Stylus Profilometer 210A. The Permeability coefficient was calculated from the PPS "Porosity" (Parker, 1965) using the methods developed by Pal (2006). The surface energies of the substrates were estimated using the Owens-Wendt (1969) method.

Water and solvent based flexographic conductive inks, with silver flakes, have been used to print conductive traces and were evaluated for print quality. Two print trials were conducted on the Comco Commander Flexographic printing press in the Western Michigan University Printing Pilot Plant. For the first trial, run at 0.62 m/s, the anilox roll had a cell volume of 12.4 μ m (8 BCM) and 79 lines/cm (200 lpi) screen ruling. For the second trial, a banded anilox roll with three different cell volumes (17.05, 18.6 and 23.3 m, which corresponds to 11, 12 and 15 BCM, respectively) was used. A consistent set of units for cell volume per unit area (l_{CV}) is used, because it gives a measure of the actual dimensions of a cell. The conventional unit, BCM, is a mixed unit and is not obviously related to cell dimensions. The two are related by (Kattumenu 2008b):

 l_{CV} (m) = 1.55l_{CV} (BCM), (1)

where l_{CV} (m) denotes cell volume per unit area in m and l_{CV} (BCM) denotes cell volume per unit area in BCM.

Four different packaging papers and three different folding carton paperboards were printed with two different silver-based conductive inks. The printing plates used were DuPont DPL photopolymer material using a CDI digital plate setter with 47 lines/cm (120 lpi) screen ruling. The plates were designed to print traces of length 50 mm and width 1 mm. Different percent tints were used to study their effect on electrical performance.

An enclosed doctor blade system was used to limit the evaporation of solvent and prevent the drying-in of the inks in the recessed cells of the anilox roll. Three air-dryers were used with drying temperatures of 116°C, 93°C and 93°C, in order from the printing unit for the first trial and 106°C after each unit in the second trial. Electrical performance was interpreted in terms of different properties of the substrates.

The print patterns were repeated across the bands of the banded anilox. The patterns for trials 1 and 2 are given in Figure 1.

Figure 1. Print pattern for trial 1 (left) and trial 2 (right).

AC and DC Impedance Test Results

To improve the conductivity of the prints, a post drying (heat treatment) was required (Kattumenu, 2008a, b). To determine a suitable time interval for the post drying, a few substrates were selected for testing and the improvement in conductivity measured as a function of post drying time (Figure 2). The

conductivity of printed traces was determined using a Keithly 2400 multimeter. It was observed that a post cure of 10 minutes at 105 °C was sufficient, after which the conductivity did not improve or improved only slightly. Figure 2 shows results for samples from trial 2, but similar behavior was seen for samples from trial 1.

Figure 2. Improvement of conductivity in dependence on heat treatment time for flexographic print trial #2

Print Quality Evaluation

The print quality of the images was quantified using an ImageXpert (KDY Inc.) image analyzer. This camera based system is a calibrated high precision optical measurement system that provides a full suite of image analysis algorithms to characterize the quality of printed output (Kipman, 1998, 2001).

Figures 3 and 4 show the average line widths and lengths for 80, 90 and 100 % tone printed lines on various substrates for trial 1. Figures 5 and 6 show the average line widths and lengths for lines printed of 90% tone steps and all cell volumes for trial 2. Measured lines were designed as a line 1 mm wide and 50 mm long (50 squares). All measured widths and most of the lengths were found to have higher values than designed. The line width gain in flexography printing is most likely due to the deformation of the plates and displacement of the ink during printing. The difference between designed and measured line dimensions is referred to as width or length gain.

Trial 1.

Figure 4. Average line length for SB (a) and WB (b) inks from Trial 1.

Figure 5. Average line width for SB (a) and WB (b) inks (all 90 % tone step) from Trial 2.

Figure 6 Average line length for SB (a) and WB (b) inks (all 90 % tone step) from Trial 2.

Edge raggedness values are shown in Figure 7 for trial 1 and Figure 8 for trial 2. Although certain substrates performed better with a given ink system, overall, the P3 and B1 were the substrates most compatible with the ink systems tested. There is no significant effect of cell volume on line raggedness. The WB had lower raggedness then the SB ink over all substrates. Only for a few samples did the value of line raggedness exceed 0.02 mm. Among the tested substrates, B2 and B3 had the lowest line edge raggedness, but simultaneously the highest gain

in line width. However, there is no clear correlation between line raggedness and line width.

Figure 7. Edge Raggedness for SB (a) and WB (b) inks from Trial 1.

Figure 8. Edge Raggedness for SB (a) and WB (b) inks (all 90% tone step) from Trial 2.

Sheet Resistivity Calculation

The DC resistance values from the Keithly 2400 and measured trace dimensions obtained from the evaluation of the samples using image analysis were combined to calculate the sheet resistivity, R_{SH}, of the printed lines according to (Gilleo, 1996):

$$
R_{SH} = R \frac{w}{l} \tag{2}
$$

where: R_{SH} is sheet resistivity in Ω sq⁻¹, *R* is the measure line resistance in Ω , *w* is the measured line width in mm, *l* is measured line length in mm.

Results for the 90% tone step printed samples from trial 2 are presented in Figure 9. The lowest sheet resistivity was obtained for the SB ink among the board substrates, but for the WB ink the lowest values of sheet resistivity were obtained for the paper samples.

Figure 9. Sheet Resistivity of lines for SB (a) and WB (b) inks (all 90% tone step) from Trial 2.

Ink Film Thickness

Before the ink film thicknesses could be obtained, selected samples were embedded in an epoxy resin and allowed to solidify for at least 18 hours. The samples were then ground and polished, using an automated grinder/polisher from Leco Corporation, to achieve a smooth surface. The thickness of the ink films were then measured across the ink-substrate interface in the print direction using optical microscopy (1000x magnification). Sample images are shown in Figure 10.

Figure 10. Sample micrographs for WB (substrate B2) on left and SB (substrate A2) on right.

The measured film thickness values for the different inks on the various substrates from Trial 2 are given in Figure 11.

Figure.11 Ink Film Thickness (18.6 m, 90 % tone step) from Trial 2.

Bulk Resistivity Calculation

If the sheet resistivity value is multiplied by the ink film thickness, a bulk resistivity, R_{DC}, is obtained. The bulk resistivity is calculated according to (Gilleo, 1996):

$$
\rho_{DC} = R \frac{w}{l} t \tag{3}
$$

where: $_{DC}$ is bulk resistivity in mil, R is line resistance in w is line width in mm, l is line length in mm, t is line thickness in mil $(10^{-3}$ in).

The bulk resistivity for the inks on the different substrates is given in Figure 12.

Figure 12. Bulk resistivity of lines for 18.6 m cell volume and 90% tone step for trial 2.

Among the paper substrates, the lowest bulk resistivity was observed for the WB ink printed on P3 and P1. For the SB ink, P4 and again P1 had the lowest bulk resistivity. For all boards, the SB ink (Figure) had a lower bulk resistivity than the WB ink and the substrate with the lowest resistivity was B2 for both ink systems. The calculated bulk resistivity was found to be lowest for the substrates with the thinnest ink films. Generally, when samples are compared in terms of conductivity performance considering all three ink systems, P1 and P4 are the substrates providing the most conductive printed lines.

These conclusions are based on the comparison of results for samples printed at the 90% tone step with the 18.6 m anilox cell volume, because as mentioned earlier, the 18.6 m performed the best among the tested cell volumes for trail 2 and the 90% tone step was used for printed RFID antennae measurements. High standard deviations for bulk resistivity originated from the ink film thickness measurement.

Antennae and Lines

Figure 13 shows scanned samples printed on P1 substrate for both ink types (18.6 m anilox band and 90% tone). It can be seen that there is an undesirable peripheral outline of the printed antenna edge for sample printed with SB ink. This phenomenon, known as the "halo effect", is inherent for flexographic printing and it is because the ink pressed between the plate and the substrate has a tendency to be squeezed to the edges and it is imprinted as the outline (Podhajny, 1998). This effect might also influence antenna performance. Considering printing of fine lines and gaps, it is evident that SB ink was not able to print clean gaps between lines, possibly due to the occurrence of the halo effect, which can be seen even around the numbers printed on the top. Of the two, the sharpest lines and gaps were printed with WB ink. It is also evident that the very fine lines were not printed continuously, which is unacceptable for printing of conductive traces.

Antenna testing was conducted at multiple levels of radio frequency (RF) continuous wave (CW), reflected impedance, and RFID dynamic time waveform capture, using an Agilent 4396B Network/Spectrum Analyzer. The testing provided insight into the ability and usefulness of various combinations of substrates and inks for printed antennae.

The initial RF testing was performed using a laboratory antenna measurement set up. This testing is intended to provide both antenna beam pattern and comparative antenna performance measures for the various printed antennae provided. The antenna range consists of a fixed reference antenna at a defined distance (3.6m) to a test antenna placed on a movable pedestal. The pedestal is programmed to rotate 360 degrees in the horizontal plane shown above and it can tilt the plane by $+/- 45$ degrees. This allows a three-dimensional pattern of the antenna performance to be collected for viewing and analysis. The measurement set up is shown in Figure 14.

Figure 14. Laboratory RF Antenna Measurement set up.

An Alien Technology UHF RFID tag "squiggle" (2nd generation) was used as the pattern for printed antennae. As for the line measurements, a heat treatment was performed on all samples measured. Conditions for post heat treatment were the same as for samples used for resistance measurements. The magnitude limit for acceptable antenna was set at -46.4 dB (yellow dot-and-dashed line inside charts). All antennae with value above this value were deemed to pass the performance test.

Performance results were observed for the SB ink (Figure 15), which showed the percentage of antennae with acceptable performance to be 87.5%. Only three samples failed to perform at an acceptable level, all printed at 17.05 m cell volume. For the rest of the cell volume values, all printed antennae showed adequate performance. Generally, for most samples, the 18.6 m was the best anilox cell volume choice to print antennae with acceptable performance. The best performing antenna for the SB ink was printed on the P3 with the 23.3 m anilox band.

Figure 15. Antenna performance measurement results for SB ink for trial 2.

Adequately performing antennae were also produced using the WB ink (Figure 16), where 91.7% of samples were found to be acceptable. Only one sample did not meet the requirement and this was for the P2 printed paper at 17.05 m cell volume. For the same sample, the 23.3 m, antenna also failed to print well. The failure to print was not due to poor paper performance but rather to ink drying issues on press, which were overlooked. Therefore, the antennae at these print conditions were not tested. Since the 18.6 m samples performed adequately, it was decided that it is not necessary to rerun the trial for this particular substrate. Once again 18.6 m is the best option among all anilox cell volume values. The best substrate among those tested for RFID antenna with WB ink was the P4 at 18.6 m.

Figure 16. Antenna performance measurement results for WB ink for trial 2.

It is also useful to observe 3D images of antenna spacial performance (Kattumenu, 2008c, Rebros, 2008). These express how the performance changes with elevation and azimuth. Figures 17 and 18 show the 3D images of performance for printed antennae on substrate P3 for the two inks with 18.6 and 23.3 m cell volumes. Areas with the color closest to the red region (top of the scale) have better performance than areas close to the blue region (bottom of the scale). It is seen that as elevation changes, performance changes too. All antennae have certain "weak" regions of performance, which is the nature of a particular antenna design and not an error caused by any given ink system or printing process.

Figure 17. 3D plots of printed RFID antenna performance for solvent-based ink on substrate P3 for 18.6 m (left) and 23.3 m (right) cell volumes.

Figure 18. 3D plots of printed RFID antenna performance for water-based ink on substrate P3 for 18.6 m (left) and 23.3 m (right) cell volumes.

Conclusions

The influence of substrate properties on ink film thickness and sheet resistivity was evaluated. Ink film thickness is affected by substrate topography and surface energy. Ink transfer was the greatest for the substrates with higher roughness and lower surface energy, resulting in thicker ink films. Sheet resistivity of the printed traces depends on surface roughness (both paper and ink) and ink film thickness. In general SB ink traces had smoother ink films but higher sheet resistivity than WB ink traces. Sheet resistivity increased with a decrease in tone step for the SB ink and was consistent for most of the substrates printed with the WB ink. The lowest resistivity was achieved with the 18.6 m (12 BCM) anilox band from trial 2 for all tone steps and ink systems.

A few antennae obtained from the first print trial failed to perform. By adjusting the properties of anilox roll and flexographic plate for the second trial, all antennae printed with SB ink and WB ink fell into the region of antennae with adequate performance.

Considering the changes for individual substrates, the best improvement was reached for B2 substrate (46.5% total improvement) and P4 substrate (46% total improvement). Based on these results, it can be concluded that the goal for the second flexographic print trial to improve antennae performance was accomplished.

Increasing the volume of the anilox roll improved the antenna performance of the 3 volumes tested 17.05, 18.6 and 23.3 m (11, 12 and 15 BCM). The 18.6 m was slightly better in most cases, but not significantly.

The flexographic print trials did not produce sufficient data to enable a correlation between resistivity and antenna performance to be statistically verified. For the SB ink an acceptable antenna performance was achieved for all substrates at 18.6 and 23.3 m cell volumes (12 and 15 BCM). The best

performance was obtained for WB inks which produced an average of -40.9 dB magnitude for all cell volumes and all substrates.

The SB ink produced an average of -42.5 dB magnitude for all cell volumes and all substrates. P3 was the best performing substrate among the paper substrates. The best performing board was the back side of B3 board. This curious finding warrants further investigation.

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