Development of In-Line Printing Press Calendering Station

Paul D. Fleming, Margaret K. Joyce, Marian Rebros, Dania Al-Said, Erika Hrehorova, and Matthew Stoops; Center for the Advancement of Printed Electronics, Western Michigan University; Kalamazoo, Michigan/USA

Abstract

Metallic conductive inks used for printed electronics have a tendency to create low conductivity layers unless they are fully dried. Yet, even after the drying, conductivity can be further improved by a post-heat-cure process. A hot soft nip calender was modified to be able to place it in line with a flexographic printing press and to study the effect of calendering on electrical performance of printed conductive layers. Two print trials were performed on the Comco Commander Flexographic Press at the Western Michigan University (WMU) Printing Pilot Plant with the calender placed in line with the moving web of the press. Three substrates were employed, a commercial label paper, a folding carton boxboard and a polyethylene-terephthalate (PET) polymer film. Two inks, one containing silver flakes and the other silver nanoparticles, were employed to print conductive layers during the print trials. Traces (lines) were printed using a three-banded anilox roll, with different cell volumes, to study the effects of ink transfer. The results were analyzed to evaluate the effects of the design variables, calender nip temperature and pressure, anilox cell volume, ink and substrate on electrical performance of printed silver inks.

The results showed that, for all inks and substrates, the electrical resistance is reduced at higher nip temperature and pressure, relative to that without calendering. Furthermore, the same resistance can be obtained with a lower anilox cell volume and calendering than with higher cell volume but without calendering. This translates to a higher ink mileage (lower ink usage) for the same electrical performance if the calendering is used. The economic benefits of this are large, because of the high cost of the silver inks.

Introduction

The advancement in functional materials, printing and electronics has led to the evolution of the new field of printed electronics. Conventional printing methods, very well known to the graphic art industry, such as screen, offset, gravure, flexography and inkjet printing are being used for manufacturing of electronic components. They provide an opportunity for high speed, low cost manufacturing and flexible electronic products[1,2]. However, there are certain limitations on functional material selection and ink formulation in terms of viscosity, particle size, drying and how material properties relate to final device performance. Each printing method has its requirements in material properties to run the printing press successfully. Functional materials are being printed onto flexible substrates to create a variety of devices that are used in electronic paper, wearable computers or sensors, conductive traces, resistors, antennae for wireless identification tags, and electrodes in printable batteries [2,3].

Typical functional materials for printed electronics include conductive, semiconductive and dielectric inks. Conductive inks can be polymeric or metallic materials. In metallic conductive inks, a metallic nano or microparticle component (e.g., silver) is dispersed into a polymer vehicle[4]. The nanoparticle ink is usually cured by heat, which melts or sinters the particles into a continuous conductive layer and decomposes the vehicle[4]. The curing temperature is in the range of 210 °F to 750 °F and the curing time varies from 5 to 60 minutes, depending on the curing temperature. Other sintering methods include laser, microwave radiation or photonic curing. In microparticle inks, the solvent is removed during drying by heat to form a conductive layer. The drying is faster than nanoparticle inks, which make them suitable for printing at speeds up to several meters per second. However, the conductivity depends on the contact between particles and is lower than that obtained with sintered nanoparticle inks[4]. In either case, the use of largely off-line curing and drying methods is a detriment to productivity,

For this study, the flexographic (flexo) printing method was employed to print conductive inks. Flexography uses relief image areas that are formed onto a polymer plate; image areas are raised above the non-image areas. The ink transfer in flexography is accomplished through anilox rolls. These rolls are engraved with small cells. The size of the cells controls the amount of ink transferred to the printing plate. The printing speed in flexography can reach up to 1500 ft/min.[2] Inks used in flexography have viscosities in the range 25 to 100 cP. Materials chosen for flexo plates must be compatible with the solvent system used for functional inks to avoid any deterioration over the length of a print run[2].

This paper examines the effect of in-line calendering on the electrical resistance of conductive traces printed onto various substrates. In the calendering process, a substrate is pressed against a polished metal cylinder with controlled pressure and temperature. In hard nip calendering, the pressure is concentrated on the high points of the web, which results in a more uniform thickness of the web. Calendering can be done off-line, where the calender is a detached standalone unit, or in-line with the printing press, which is more productive, economic and cost efficient[5]. Most calendering is performed in-line with a paper machine and coater to improve the surface properties of paper for improving print quality[5]. Although calendering is commonly practiced by papermakers, the effect of in-line calendering on the properties of printed layers or printed conductive inks has not been studied or examined before. Recently[6,7], the effects of off-line calendering on previously printed conducting traces were reported. These results suggested that the conductivity of traces could be improved with in-line calendering, thus eliminating the need for further post-treatment. This work grew from the observation that printed conducting traces often require a "post-cure"[4,8,9], before reaching the best electrical performance (Figure 1) and that an increase in contact between metallic ink particles through the pressure of calendering could further improve conductivity. Off-line post-curing is a serious impediment to productivity; so identifying an in-line drying/curing method is of great importance for the success of printed electronics. The previous work and work reported here is towards specifying such a method.

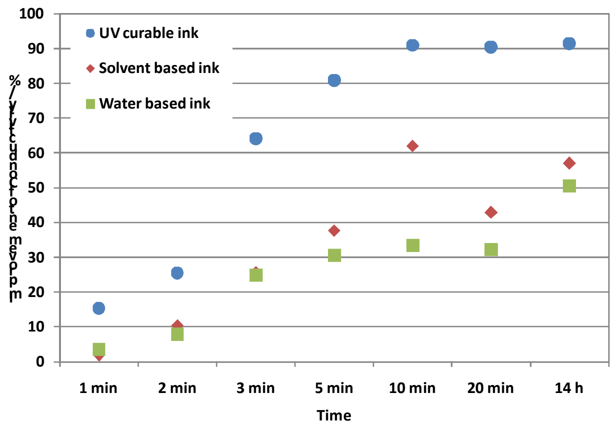

Figure 1. Conductivity improvement of silver based inks with post-curing.

Controlling ink drying/curing is crucial to achieving the main objective of manufacturing conductive traces printed onto flexible substrates with improved functional properties and surface smoothness[5]. Smoothness is an essential property, especially when several layers of functional materials are printed over each other. With calendering, the printed substrate is subjected to pressure in the z-direction and to heat from the calender roll. Due to this compression, calendered materials go through plastic and viscoelastic deformations that cause particle alignment in the ink layer and reduction in thickness, density, porosity, and roughness. In addition, raising the temperature of the substrate can further dry the ink layer and soften the ink resin to aid in particle alignment for increased particle contact.

Experimental

For this study the flexographic printing process was used to print conductive traces. Figure 2 shows a part of the design used for the evaluation of printed and calendered samples. Solid lines (1 x 50 mm) were used to measure the effect of calendering on electrical performance (resistance). A solid patch (30 x 100 mm) was used to evaluate the effect of calendering on smoothness.

The calendering unit was built and positioned in-line with the flexo printing press (Figure 3). In the following section, print/calender trials are described in more detail.

A Comco Commander narrow-web 3 unit-flexographic press, located at the WMU Printing Pilot Plant, was used to print conductive traces. To study the effect of in-line calendering, a soft nip calender with the option to control pressure and temperature levels was used. Traces were printed onto three commercially available substrates: label paper, polyethylene terephthalate film (PET), and paperboard (CNB). Two ink systems were selected to compare their printability and performance at different calendering conditions: water base (WB) silver flake ink and solvent based nanosilver (Nano) ink.

The design had traces printed in both machine direction (parallel with web path) and cross-machine direction. A banded anilox roll was used with three screen rulings, 120 lpi (Band A), 180 lpi (Band B) and 220 lpi (Band C). The cell volume/unit area for band A is 18.6 µm or 12 BCM (Billion Cubic Microns per square inch)[9]. The cell volumes per unit area for bands B and C are 15.5 µm (10 BCM) and 12.4 µm (8 BCM), respectively. Different cell volumes transfer different amounts of the ink from the anilox roll onto the printing plate and consequently onto the printed substrate. As a result, different ink layer thicknesses can be achieved. Multiple anilox cell volumes were chosen to determine if the calendering process could result in the need for less ink use. To verify this effect, the electrical resistances of the traces printed with the different anilox bands before and after calendering were compared.

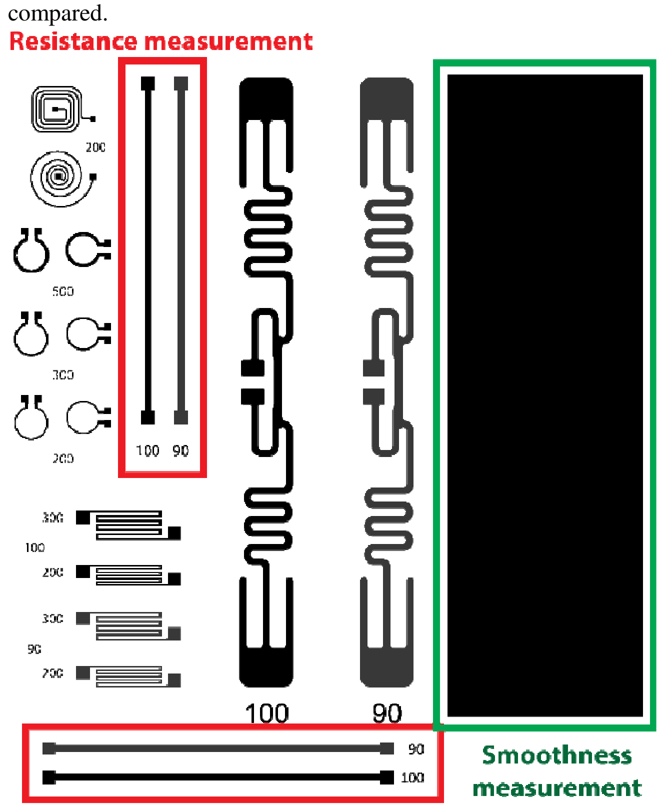

Figure 2. Section of a printed design.

Figure 3. In-line set-up of calendering unit built and placed at the end of a flexographic printing press.

Pilot Trials Description

Overall, two print/calender pilot trials were run and evaluated. Table 1 summarizes the run conditions for the first trial. To maintain good print quality and sufficient drying of the inks, a press speed of 100 ft/min for the WB ink and 50 ft/min for the Nano ink was maintained during the trials. In addition, during the pilot trials, the temperature of the substrate was monitored with an infrared non-contact thermometer. The temperature of the web was recorded before and after entering and exiting the calendering unit.

Table 1. Run conditions for first pilot trial.

		Calendering conditions	
Ink	Substrate	Temperature [°F]	Nip Pressure [pli]
WB	CNB	75 (ambient)	50, 500, 950
	Label	75 (ambient)	50, 500, 950,1500
	PET	75 (ambient)	50, 500, 950,1500
Nano	PET	75 (ambient)	50, 500, 950
	Label	75 (ambient)	50, 500, 950
	CNB	75 (ambient)	50, 500, 950
WB	PET	104	500, 950,1500
	Label	104	500, 950,1500
	CNB	104	500, 950
Nano	CNB	104	500, 950
	Label	104	500, 950,1500
	PET	104	500, 950
WB	Label	140	500, 950,1500
	CNB	140	500, 950
Nano	CNB	140	500, 950
	Label	140	500, 950,1500
	PET	140	500, 950,1500
WB	CNB	158	500, 950
Nano	CNB	158	500, 950
Nano	Label	176	500, 950,1500
WB	Label	176	500, 950,1500
	CNB	176	500, 950
	PET	176	500, 950,1500

The conditions for the first trial were set based on the conditions used in the preliminary studies performed on a sheet-fed calender, the results from which merited this work. After the first calendering study, it was recognized that the higher speed of the in-line calendering unit prevented the duplication of the sheet temperatures obtained on the sheet-fed calender. Sheet temperatures were lower because at the higher press speed, the sheet was in contact with the heat roll for a shorter period of time. So, the goal of the second trial (Table 2) was to increase the sheet temperature. The temperatures were raised stepwise to determine the maximum calendering temperature that both paper substrates, Label and CNB could withstand without any deterioration in their structure. The calendering pressure was fixed for each substrate. An additional drying unit was added before the calender to prevent sticking of the ink and coating to the calender as a result of the large temperature gradient. In commercial practice, the temperature gradient is avoided by placing the calender directly after the last dryer section, but due to the layout of the printing press that was used for this study, this could not be accomplished. Hence, the additional dryer better simulated what would be commercially practiced.

Table 2. Run conditions for second pilot trial.

		Calendering conditions	
Ink	Substrate	Temperature [°F]	Pressure* [pli]
WB	CNB	75 (ambient)	50, 950
	Label	75 (ambient)	50, 1500
	Label	176	1500
	CNB	176	950
	CNB	194	950
	Label	194	1500

Sample Evaluation

The printed traces were characterized by measuring the electrical properties, roughness and print quality characteristics. The electrical properties of the printed traces were measured before and after calendering in terms of resistance (R), using a 4338B Milliohmmeter. An ImageXpert image analysis system was employed to measure print quality in terms of line width and raggedness. The line width for 100% tone printed traces was measured at 10 different places and the average was reported to investigate the temperature and pressure effect on line width gain. An Emveco 210R stylus profilometer was used to measure the roughness of the printed ink films, before and after calendering according to TAPPI T575 standard.

The effect of calendering temperature and pressure on resistance was studied and statistically analyzed by analysis of variance (ANOVA) and regression analysis, using Minitab 15. Table 3 shows a summary of all factors and their levels.

Table 3. Analyzed factors and their levels.

Factors	Levels
Calendering Temperature (°F)	75, 104, 140, 158, 176
Calendering Pressure (pli)	50, 500, 950, 1500
Bands (µm)	12.4, 15.5, 18.6
Ink	WB, Nano
Substrate	Label, CNB, PET

The analysis was conducted for all factors in both trials. The response in each case was electrical resistance in both machine direction, MD (R1) and in cross-machine direction, CD (R2). ANOVA General Linear Model was applied, since several factors were studied. The p-value is an indication of a factor's significant effect on a response. In this situation, a p-value less or equal to 0.05 indicates a significant effect of the factor (with 95% confidence or better).

Results - First trial

The following is a discussion of the ANOVA results and plots obtained for the effect of calendering temperature, calendering pressure, anilox band volume, ink and substrate on resistance measured in MD. The analysis also examines the improvement in conductivity with calendering.

The results of ANOVA in Appendix 1 show the significant effect of all the variables except ink. Figure 4 shows a great reduction in resistances when samples are calendered at high pressures and temperatures when compared to those uncalendered. According to the p-value obtained from the effect of ink (0.224) and Figure 4, similar resistances were obtained with both WB and Nano inks. However, better drying was achieved with WB inks especially on PET, which can explain the high resistances on PET, when compared to both CNB and Label. This is clear in Figure 4, where traces printed on Label paper had the best conductivity of all the substrates.

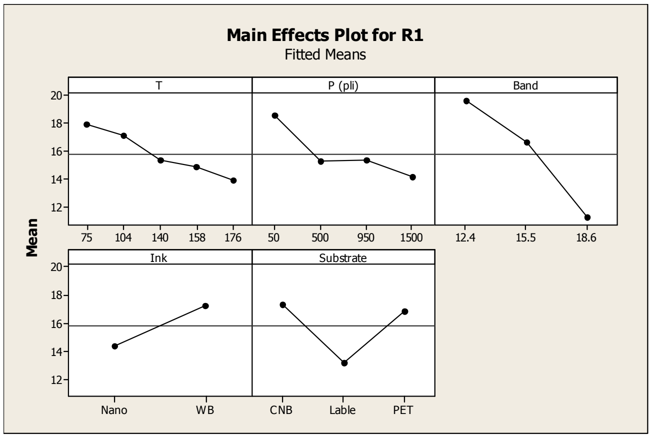

Figure 4. Main Effects Plot for R1 from first trial.

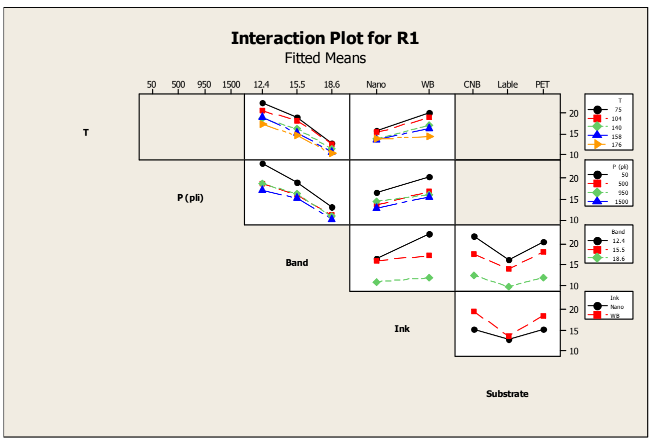

Figure 5. Interaction effects plot for R1 from first trial.

To further elucidate the dependences, a regression analysis was conducted for the above data to obtain the relation between resistance and the random variables: temperature, pressure, and band volume. To obtain a useful analytical tool, the discrete variables ink and substrate were mapped to numerical ones. In particular, for the inks, 'Nano' was mapped to 0 and 'WB' was mapped to 1. Likewise, for the substrates, 'Label' was mapped to 0, 'PET' was mapped to 1 and 'CNB' was mapped to 2. This allowed us to regress on 5 main effects and up to 10 interactions. It turned out that, for the regression, the pressure main effect is not significant, but all the interactions are significant at better than a 98.8% confidence limit. The following regression equation was obtained:

$$R_1 = 34.3 - 0.0532T - 1.35b + 18.8i + 6.24s - 0.000051TP + 0.000510Pb - 0.00161iP - 0.000699Ps + 1.96is + 0.00386Tb - 0.852bi - 0.267bs - 0.00551Ts - 0.0279Ti \qquad (1)$$

where R_1 is the resistance measured in MD in Ohms

T is the calendering temperature in °F

P is the calendering nip pressure in pli

b is the cell volume per unit area in µm

s is the substrate number as describe above

i is the ink number

The R^2_{adj} for this regression is 83.0%.

According to Equation 1, a lower resistance can be obtained with calendering at high temperatures and pressures and with lower band volumes (less ink), when compared to the resistivity of uncalendered samples at higher band volumes. Therefore, we can conclude that calendering provides higher ink mileage and hence a pronounced cost reduction of ink. This can be quantified by solving this equation for the calendered cell volume necessary to obtain the same resistance as an uncalendered print at a given cell volume, for any given ink. Likewise, other equivalent conditions can be solved for in order to optimize the process.

Results - Second Trial

The goal of this trial was to find the maximum calendering temperature that both paper substrates, Label and CNB board, can handle without any deterioration in their structure or sticking to the calendering roll. An additional drying unit was added before the calendering to simulate the sheet temperatures that would be experienced in commercial practice. Resistances at temperature 75 and 176 °F were compared to that of the first trial. It was found that addition of the drying unit lowered sheet resistivity. However, the reduction in resistance was within a few Ohms when compared to the results obtained in the first trail, which may not compensate for energy cost to run additional drying units.

Results of two-way ANOVA (Appendix 1) show a significant effect of temperature on resistance for Label and CNB in both MD and CD. Figures 6 to 9 show the effect of calendering temperature of the second trial on MD and CD resistances at different band volumes for Label and CNB. As the temperature increased from ambient conditions (around 75 °F), the resistance decreased by a great deal. Above 176 °F, the effect of temperature was reduced and the difference in resistances between 176 and 194 °F was within a few Ohms. Therefore, calendering at temperatures as high as 194 °F for both Label and CNB may not be economically beneficial, since more energy must be supplied to achieve these higher temperatures.

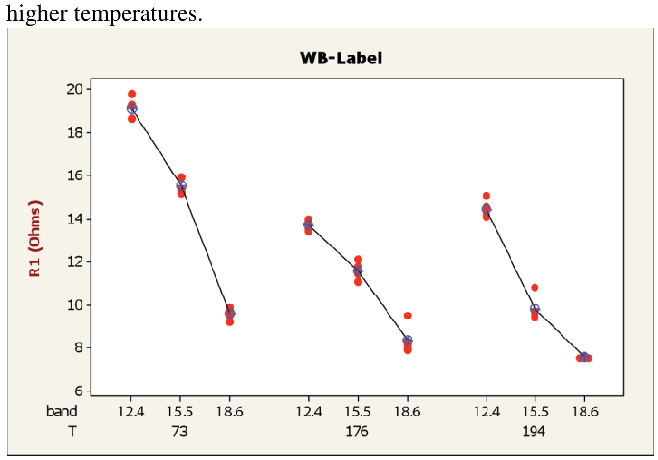

Figure 6. Effect of Calendering Temperature on MD Resistance for Label.

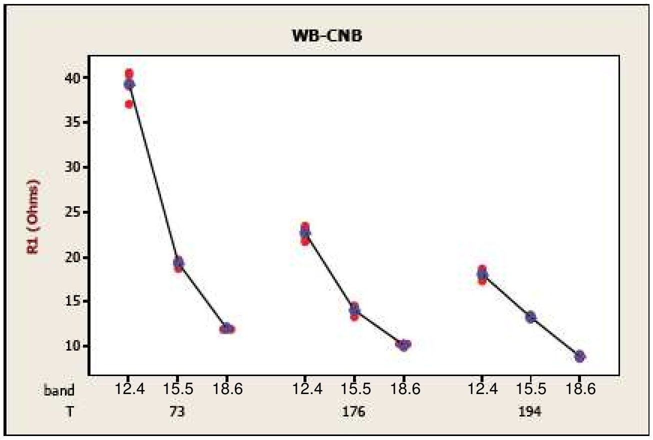

Figure 7. Effect of Calendering Temperature on MD Resistance for CNB.

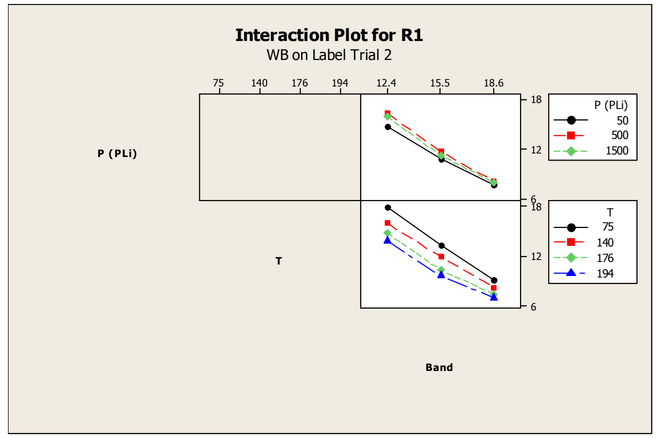

Figure 9. Interaction Plot for R1: WB on Label.

ANOVA and Regression for the Second Trial

WB traces were printed on Label at temperatures varying from 75 °F to 194 °F and pressures from 50 pli (uncalendered) to 1500 pli. ANOVA analysis is summarized in Appendix 1. The analysis and main effect plot, Figure 8, show again the significant effect of calendering temperature on resistance of the printed traces. From the interaction plot between temperature and band and pressure and band, Figure 9, we can see that high temperature calendering reduces the resistance a great deal when compared to uncalendering for the same amount of ink. The statistical software would not compute the pressure-temperature interaction effect because of deficiencies of the data. As will be seen in regression analysis below, the effects of pressure-temperature interaction are significant

As above, we can regress on three main effects and up to 3 interactions to quantify the implications of the results of this trial. From the regression, it was found that the dependence on the pressure-band interaction was not significant. All of the other main effects and the pressure-temperature and temperature-band interactions are significant at better than a 99.9% confidence limit.

The numerical relation between resistance and the random variables in this part of the trial (calendering temperature, pressure and band) is obtained by the regression equation:

$$R_1 = 39.9 - 0.0524T + .00122P - 1.63b - .000010TP + .00232Tb + .000007Pb \quad (2).$$

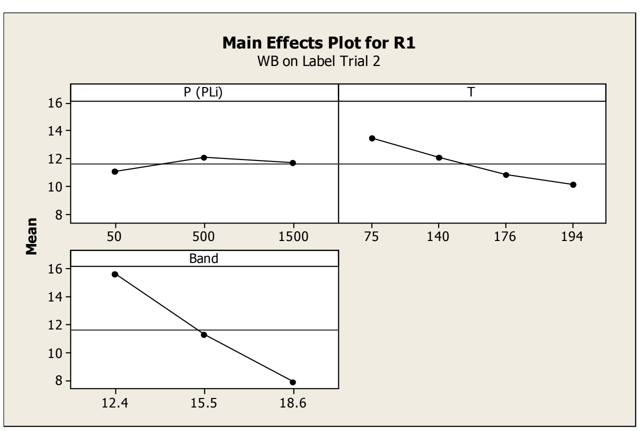

Figure 8. Main Effects Plot for R1: WB Silver Flake on Label, Trial 2.

The R^2_{adj} for this regression is 95.2%. The coefficient of the temperature is approximately the same as in Equation 1. The coefficient of the Tb is slightly more than half of that in Equation 1, but the coefficient of the b term is slightly larger. The coefficient of the TP term is approximately 20% of that in Equation 1. The coefficient of P is positive, but the coefficient of TP is negative. This means that, for low temperatures, increasing the pressure slightly increases the resistance, but at higher temperatures, increasing pressure decreases the resistance. If we factor the P and TP terms, then the coefficient of .00111P is 1 - .00899T. Thus for temperatures above 111°F, increasing the calendering nip pressure improves the conductivity. The results that the coefficient of the TBand are reduced by slightly more than half, and that the effective coefficient of P is positive only above 111°F, are likely consequences of the additional drying on press (6 passes through press dryers and 4 passes past other hot air dryers).

Conclusion

Calendering temperature had a significant effect in reducing resistance of the traces for both WB silver flake and nanosilver inks printed on all three substrates; label paper, board and polymer film. The effect of calendering pressure on resistance was not significant for all WB printed traces, although for sufficiently high temperatures increasing pressure decreases resistance for all inks. A slight decreasing trend was found for the effect of pressure on resistance for nanosilver inks on label paper. However, none of these trends appears to be significant. For CNB the effect was not significant. However, calendering pressure on CNB was not as high as that applied to label paper or PET to avoid any deterioration in the board structure. Combination of calendering pressure and temperature and their interaction was found to reduce the resistance for WB and nanosilver inks printed on all substrates. Based on the above results, it can be concluded that calendering has the ability to improve conductivity of printed traces, which offers some cost savings during manufacturing.

Increasing the volume of anilox cells reduced the resistance of the printed traces. The 18.6 µm anilox cell volume was found to be the best in terms of electrical performance. This was an expected result, since more conductive ink is deposited with higher volume of the anilox cells. A lot of scatter and variation was obtained when nanosilver ink was printed on PET. This could be due to the insufficient ink drying on the film. Calendering increased the conductivity of traces sufficiently that printing with a lower cell volume anilox can reach the conductivity of uncalendered traces printed with a higher cell volume anilox. Regression equations were obtained that can be solved to estimate the reduction in cell volume obtained with calendering.

The results of the second trail were consistent with that obtained in the first trail. However, calendering temperature should not exceed a certain value unless a non-stick coating is applied to the roll in order to avoid any sticking of the ink on the calendering roll or damaging of the substrate. In addition, ink film resistances tend to reach a stable resistance value and any additional increase in calendering temperature or pressure would be not efficient.

Acknowledgements

The authors would like to thank Mark Sorenson from ANDRITZ Küsters and Kevin Manes from Mark Andy for support and help during this study. The authors also thank Michigan Initiative for Innovation and Entrepreneurship (MIIE) program for partial financial support for this work.

Appendix 1

ANOVA Analysis for Resistance Measurements

First trial

ANOVA for the Data Obtained on First Trial:

General Linear Model: R1 versus T, P (pli), Band, Ink, Substrate

Factor	Type	Levels	Values
T	random	5	75, 104, 140, 158, 176
P (pli)	random	4	50, 500, 950, 1500
Band	random	3	12.4, 15.5, 18.6
Ink	fixed	2	Nano, WB
Substrate	fixed	3	CNB, Label, PET

Analysis of Variance for R1, using Adjusted SS for Tests

Source	DF	Seq SS	Adj SS	Adj MS	F	P
T	4	3926.00	2090.54	522.63	4.44	0.056 x
P (pli)	3	3699.63	2705.14	901.71	.74	0.012 x
Band	2	14226.30	9639.25	4819.63	6.91	0.074 x
Ink	1	2779.03	1446.04	1446.04	2.36	0.224 x
Substrate	2	3745.91	3490.15	1745.08	20.59	0.008 x
T*Band	8	491.40	218.66	27.33	6.15	0.000
T*Ink	4	751.96	389.57	97.39	21.91	0.000
P (pli)*Band	6	433.73	359.38	59.90	13.47	0.000
P (pli)*Ink	3	227.76	186.90	62.30	14.01	0.000
Band*Ink	2	1588.93	1456.18	728.09	163.79	0.000
Band*Substrate	4	377.48	375.90	93.97	21.14	0.000
Ink*Substrate	2	625.78	625.78	312.89	70.39	0.000
Error	1107	4920.80	4920.80	4.45		
Total	1148	37794.70				

x Not an exact F-test.

S = 2.10836 R-Sq = 86.98% R-Sq(adj) = 86.50

Second trial

ANOVA for the Second Trial:

General Linear Model: R (MD) versus T, P (pli), Band

Factor	Type	Levels	Values
P (pli)	random	3	50, 500, 1500
T	random	4	75, 140, 176, 194
Band	random	3	12.4, 15.5, 18.6

Analysis of Variance for R (MD), using Adjusted SS for Tests

Source	DF	Seq SS	Adj SS	Adj MS	F	P
P (pli)	2	11.59	12.89	6.44	9.74	0.029 x
T	3	280.01	280.01	93.34	29.47	0.001 x
Band	2	1924.26	740.68	370.34	192.24	0.000 x
P (pli)*Band	4	1.06	2.65	0.66	1.11	0.354 x
T*Band	6	19.00	19.00	3.17	5.31	0.000 x
Error	147	87.66	87.66	0.60		
Total	164	2323.57				

x Not an exact F-test.

S = 0.772206 R-Sq = 96.23% R-Sq(adj) = 95.79

Author Biography

Paul D. "Dan" Fleming III received his BSc in physics from the Ohio State University (1964) and his AM in Physics (1966) and PhD in chemical physics from Harvard University (1971). Since then he has worked in Industrial Research and Development at Phillips Petroleum and GenCorp. He is currently a Professor in The Department of Paper Engineering, Chemical Engineering and Imaging at Western Michigan University His His research interests include printed electronics, digital printing, color management and paper coatings. He is a member of IS&T, APS and TAGA

References

[1] Kirchmeyer, S. Gaiser, D., "Extremely Flat and Flexible", *Polymer Electronics, Kunststoffe International* (2007).

[2] Rigney, J., Materials and processes for high Speed Printing for Electronic Components" IS & T's NIP20: *International Conference on Digital Printing Technologies* (2004). (pp. 275-278).

[3] Rogers, J. Bao, Z., "Printed Plastic Electronics and Paperlike Displays", *Journal of Polymer Science: Part A: Polymer Chemistry*, Vol. 40, (2002).

[4] Eiroma, K. Hakola, L. Hast, J. Maaninen, A. Petaja, J., "UV Curing for Printed Electronics", *Radtech Report* (2007).

[5] Smook, G.A. (1992), *Handbook for Pulp and Paper Technologists*, Angus Wild Publications, Inc. (pp. 264).

[6] Sujay Pandkar, "Effect Of Calendering On Flexographically Printed Silver Flake Ink", MS Thesis, Western Michigan University, 2008.

[7] Margaret, K. Joyce, Paul D. Fleming III and Sujay Pandkar, "Method of Improving The Electrical Conductivity of a Conductive Ink Trace Pattern and System Therefor", US Patent Application 131347.

[8] Ramesh Kattumenu, Marian Rebros, Margaret Joyce, Paul D. Fleming and Gururaj Neelgund, "Effect of Substrate properties on Conductive Traces Printed With Silver-Based Flexographic Ink", *NPPRJ*, 2009, 24, p.101-106.

[9] Ramesh Kattumenu, Marian Rebros, Erika Hrehorova, Paul D. Fleming, Margaret Joyce, Bradley J. Bazuin, Alexandra Pekarovicova and Gururaj Neelgund, "Evaluation of Flexographically Printed Conductive Traces on Paper Substrates", TAGA Proceedings, San Francisco, March 2008.