Printing Transparent Grid Patterns with Conductive Silver Inkwith Flexography

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Abstract

The printed electronics market, according to market research firm IDTechEx, is slated to grow from \$16.04 billion in 2013 to \$76.79 billion in 2023. Printing transparent grids with a known printing process such as flexography is attractive to this market because transparent grids are part of the fast-growing touch screen market (amongst others), and flexography offers a lowcost/high-volume print platform that can handle a variety of substrates, high press speeds, roll-toroll (R2R) printing, and the ability to handle different ink rheologies. However, flexography has print characteristics that can be problematic for printed electronics such as high image (feature) gain and the ability to hold fine images (features) in the (relief) printing plate.

This study used a "banded" anilox roll, which has multiple volumes and cell counts separated into bands, conductive nano-silver, water-based ink, and a PET substrate and printed on a commercial flexo narrow- web press. The process parameters studied included types of sticky back, anilox cell count/volume, plate imaging, and plate surface morphology. The printed transparent grid patterns were evaluated for conductivity, which was measured with a Digital Multimeter; and transparency, which was measured with a transmission densitometer.

The results of the study showed that when the photopolymer plates were imaged at a resolution of 8000dpi and with a "flat top dot" (1:1 file – image) exposure technique, the flexo plate was able to hold a minimum of 6.35 microns line width. The 6.35 microns lines were successfully printed on the press and produced the conductive lines. Types of sticky back and different anilox cell volumes have an impact on measured resistance of the grid pattern. A high modulus, or "firm" sticky back tape with a low screen count/high-volume anilox (800 cpi/2.85bcm) resulted in the lowest (<15 ohms) resistance. In addition, the introduction of plate surface texture during

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the platemaking lowered resistance. The grid pattern shapes of diamond/square and hexagon yielded the best transmissivity (above 92%).

The study proved that commercially available flexographic materials and optimized process parameters could improve the printability and performance of transparent grid patterns with conductive inks.

Introduction

Transparent conductive films (TCFs) produced with transparent conductive oxides (TCOs) and transparent polymers are used for the manufacture of a variety of applications including touch screens, flat panel displays, and solar cells. They have high optical transmission and electrical conductivity (Materion Microelectronics & Services).

Liu, et al (2014) concluded that TCFs can be manufactured with both subtractive and additive processes. Arguably, the most common way to produce TCFs is by using chemical vapor deposition (CVD) with Indium Tin Oxide (ITO), or tin-doped indium oxide. CVD is a subtractive process, depositing the ITO completely on the substrate. Any patterning for TCF can be created either with a laser, plasma, or chemical etch. The benefits of ITO are that it has very high optical transmission and electrical conductivity. The material is extremely transparent and can be deposited in thin layers. However, the material is brittle and is easily damaged, resulting in decreased performance and limits its use on flexible substrates. Since Indium is mined, it is in limited supply and its price fluctuates based on availability.

There are additive processes for TCFs as well. Silvernanowire inkshave been developed as an ITO replacement; ultra-fine wires are suspended in a clear fluid to create a random network of conductive wires that provide high conductivity. Because the wires are tens of nanometers in diameter and tens of microns in length, they form a randomly-distributed conductive grid with high transparency (Gupta, and Peruvemba, 2013).

In addition to silver nanowire, organic polymers such as PEDOT: PSS (Poly 3,4-ethylenedioxythiophene) polystyrene sulfonate are conductive polymers, which can be applied to substrates such as PET (flexible) using processes such as flexography and screen-printing. Although there are several manufacturers of this material, the challenge is that the conductive polymers are more resistive and not as transparent as other solutions.

Carbon Nanotubes (CNTs), graphene films, silver nano wires, and silver nanoparticles are all materials that also have been explored as ITO replacements. Printed Electronics Now indicates silver nanoparticles in particular are attractive as they are high in conductivity but are less expensive than silver nano wires, and don't have issues such as potential moiré (Sun, 2015).

The challenge of additive printing methods and materials such as silver for TCFs is that the more material that is deposited on a substrate, the higher the conductivity – but due to the opacity of the silver, the lower the transparency. There is typically an inverse relationship between transparency and conductivity when printing conductive materials on clear substrates. Liu, et al concluded that in a flexographic printing environment, the grid pattern shape and printing parameters had an effect on performance of a printed transparent conductive film. The results showed that a hexagonal shape was optimum compared to another tessellated shapes such as triangles and squares, that printing pressures influenced the result (150N), and that sintering influenced conductivity. In addition, as line width for the hexagon increased, transparency decreased, but as line length increased, creating larger open areas. transparency increased. Liu et al chose a 300 CPI (cells per inch) anilox roll for higher volume ink metering as the line segments had lower resistance. Since high transmittance and low resistance are required in TCF applications, Liu et al characterized the comprehensive quality of these two aspects as "O" (the ratio of light transmittance to total resistance). The hexagonal grid had the highest "Q" value (Liu, et al 2014).

Based on the research of Liu, et al, hexagonal grid patterns, along with a few other shapes were explored with the goal of achieving the best conductivity and transparency from commercial printing process. Some of the challenges of flexographic printing of TCFs were revealed in their research (2014). Very high ink volumes can affect ink transfer and "flooding" of ink around the features (300 CPI anilox roll). Surface roughness of the plate can affect the way ink lays on the substrate.

The purpose of this paper is to investigate the variables in the flexographic printing process that affect the way ink is transferred to the substrate from the printing plate. The research sought to determine if the following variables could be optimized with commercially available technology to impact and exploit better reproduction of fine features and conductivity/transparency for TCFs: plate imaging, exposure, and processing, plate material, mounting tape modulus, and anilox line screen and volume. This research was conducted by a collaborative team from Cal Poly and Clemson University, with the support of DuPont™ Cyrel® (plates), NovaCentrix (inks), and DuPont™ Teijin Films.

Experimental Procedure

This TCF printing experiment was designed to take advantage of commercially available consumable materials, in addition to commercially available technologies.

The printing press used was an OMET VaryFlex flexographic press. This press is a narrow web, seven-station flexographic press with a heat tunnel overhead dryer that makes it ideal for printing and curing the ink. The substrate used was DuPont

Teijin Melinex 502 PET. The conductive ink was Pchem/NovaCentrix PF1-722 nanoparticle water-based silver ink. The mounting tape materials used were 3M 1920 (light medium), and 3M 1820 (firm) mounting tape. The digital photopolymer plate material used was DuPont DPR, processed in solvent – 0.045" thickness with .010" - .015" relief. DPR is a hard (~60 shore A) plate material that tends to deform less during printing than softer plates, thus providing a better potential reproduction of features. In addition, this digital plate was processed in the presence of nitrogen, as opposed to air (oxygen), in order to form "flat top" features. These "flat top features", which have essentially a 1:1 ratio of file image to plate feature, are known to hold fine detail better such as the thin grid lines, and print with less deformation than a traditional digital plate exposed in the presence of oxygen (Kahn, et al 2012).

The plates were imaged at both 4000 dpi (dots per inch) and 8000 dpi resolution on an Esko SecuFlex digital plate imager.

The press run used a banded anilox roll with the following format: A: 1200 cpi (cells per inch)/1.03 bcm (billion cubic microns), B: 1100 cpi/1.53 bcm, C: 1000 cpi/ 1.75 bcm, D: 900 cpi/ 2.22 bcm, E: 800 cpi/ 2.85 bcm, F: 700 cpi/ 3.58 bcm. Table 1 illustrates the configuration of the banded anilox roll.

Anilox Band	Line Screen (CPI)	Cell Volume (BCM)	
A	1200	1.03	
В	1100	1.53	
С	1000	1.75	
D	900	2.22	
Е	800	2.85	
F	700	3.58	

Table 1: Banded anilox roll configuration

A test target was created in Adobe Illustrator Creative Cloud with screening details applied in Esko PackEdge, and contained twelve transparent grid patterns with six different hexagon patterns, one square, one pentagon, one octagon, one circle, one triangle, and one diamond pattern. These grid patterns had various ratios labeled in the convention of shape, line width (), size, and number of sides. 1: Hexagon, $6.35 - 500\mu - 6x$, 2: Hexagon, $9.25\mu - 500\mu - 8x$, 3: Hexagon, $9.52\mu - 510\mu - 6x$, 4: Hexagon, $6.35\mu - 1mm - 6x$, 5: Hexagon, $15.875\mu - 1mm - 6x$, 6: Pentagon, $9.52\mu - 900\mu - 5x$, 7: Square, $12.70\mu - 1mm - 4x$, 8: Hexagon, $9.525\mu - 1mm - 6x$, 9: Triangle, $12\mu - 6\mu - 1mm$, 10: Circle, $9.525\mu - 1mm - Circ$, 11: Diamond, $12.70\mu - 1mm - 4x$ (b), 12: Hexagon, $12.70\mu - 1mm - 6x$. Table 2 illustrates the configuration of the test target.

Grid Pattern	Shape	Line width (µ)	Size (diameter of in circle)	Number of sides
1	Hexagon	6.35	500μ	6
2	Octagon	9.25	500μ	8
3	Hexagon	9.25	510μ	6
4	Hexagon	6.35	1mm	6
5	Hexagon	15.875	1mm	6
6	Pentagon	9.25	900μ	5
7	Square	12.7	1mm	4
8	Hexagon	9.25	1mm	6
9	Triangle	12/6	1mm	3
10	Circle	9.525	1mm	Circle
11	Diamond	12.70	1mm	4
12	Hexagon	12.70	1mm	6

Table 2: transparent grid patterns

Figure 1 illustrates the grid patterns described above, and Figure 2 illustrates the positive and reverse lines.

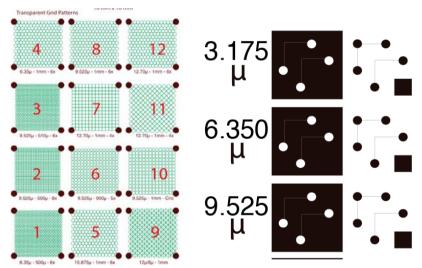


Figure 1: transparent grid patterns

Figure 2: 45-degree lines at various thicknesses

Fine positive and reverse line features were created at forty-five degree angles with the following thicknesses: 3.175 (one pixel at 8000 dpi) μ , 6.35μ (one pixel at 4000 dpi), 9.525μ , 12.7μ , and 20μ .

Resistance was measured with a Mastech MS8268 digital multimeter, and transmission (transmissivity) was measured with an X-Rite 301 transmission densitometer and mathematically converted from density to % transparency.

Results

1. Resolution in the imaging device is critical for the ability to hold fine features for TCF printing. The finer the feature that can be printed, the more transparent the grid pattern will be on the TCF. An 8000 dpi plate-imaging device can create a 3.175 feature in the plate, which is one pixel in size at that resolution. A 4000 dpi plate-imaging device can image a 6.35 feature on the plate, which is one pixel in size at that resolution. When the features are on a bias relative to the drum rotation, however, the way the pixels align themselves can create issues with forming robust features in the processed plate. As seen in Figure 3 (a,b), a line imaged off the bias may produce variation of features on a plate.

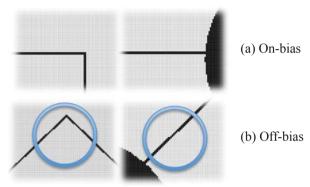


Figure 3 (a,b): line integrity based on orientation in imager

2. For this experiment, the most robust plate imaging was done at 8000 dpi, with 2 pixels; a 6.35μ line fully formed in both positive and reverse (Figure 4). Even though the 4000 dpi imaged a 6.35μ line on the plate carbon mask, it was not robust enough at one pixel, nor did the 3.175μ line at 8000 dpi form in the plate or print. The most robust combination of feature size and resolution was 8000 dpi and 6.35μ (2 pixels). As mentioned already, these plates were exposed without the presence of oxygen, thus the photopolymerization, or curing, of the plates was not inhibited (Kahn, et al 2012). The features in the plate had a "flat top" and sharp definition between image and shoulder—no rounded corners.

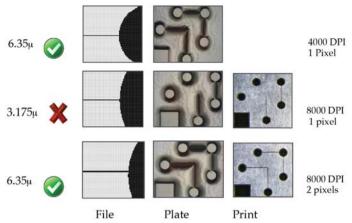


Figure 4: example of image resolution on feature forming and printing

3. Sheet resistance measured from the conductive traces from each anilox band was as follows: A: 1200cpi/1.03bcm measured ±3.06 Ω/square, B: 1100cpi/1.53bcm measured ±2.11 Ω/square, C: 1000cpi/1.75bcm measured ±1.17 Ω/square, D: 900cpi/2.22bcm measured ±0.62 Ω/square, E: 800cpi/2.85bcm measured ±0.37 Ω/square, and F: 700cpi/3.58bcm measured ±0.26 Ω/square. As anilox volume increases, and more ink is delivered to the plate and substrate, sheet resistance goes down. Figure 5 illustrates this trend graphically.

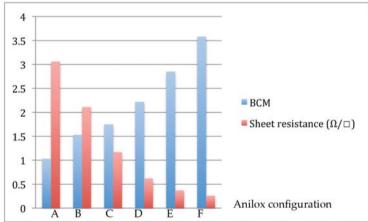


Figure 5: anilox volume vs. sheet resistance

4. As was noted in Liu's research (2014), the grid patterns of squares, triangles, and hexagons are the only polygons that will tessellate the Euclidean plane. With more sides touching each other, there is opportunity for more light to transmit, thus increasing transparency.

This experiment confirmed that the hexagon pattern is the best choice for overall transparency and conductivity.

5. The plate-to-print analysis revealed that the lines on the processed plate were 11μ (growth from 6.35μ). Figure 6 shows the printed vertical lines grew to 25μ and the printed diagonal lines grew to 30μ. As the plates are printed, the hexagon shape is influenced by the press direction. There is more image growth, commonly referred to as "line gain," against the press direction and less where the line is oriented in the press direction.

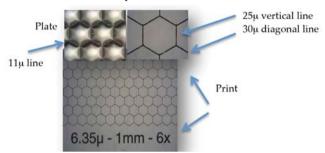


Figure 6: example of plate feature size compared to print

6. The firmer of the two types of mounting tape tested in this experiment provided better results than the light medium mounting tape. The 3M 1820 foam tape is a PE foam structure with a density of 35 lb./ft3. The compression deflection at 10% is 35 lb./in2. The durometer (hardness) is 72 Shore A. The 3M 1920 foam tape is a PE foam structure with a density of 26 lb./ft3. The compression deflection at 10% is 10 lb./in2. The durometer is 60 Shore A (3M Company, 2010).

The 3M 1820 tape printed the grid patterns with the water-based silver ink more robustly than the 3M 1920 tape. This is logical, as in graphic printing with flexography there tends to be less image gain with lighter or medium tape, and solid areas of the plate tend to print with fewer pinholes and visual defects with firm tape than lighter tape. So, although the grid lines printed with the 3M 1920 have less image gain, the firm tape provided a more stable platform for image integrity. The plate printed with more growth with the 3M 1820, but there were no image skips or pinholes, and the ink film was thicker. As seen in Figure 7, in graphic printing, the visual effect of the 3M 1920 would be preferred, but that printing result would affect the functionality of the grid pattern—conductivity. Figure 7 illustrates the difference in mounting tape selection.

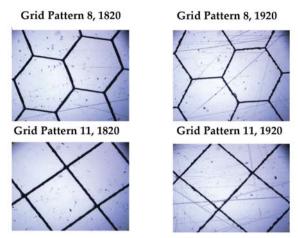


Figure 7: comparison of mounting tape material

Transmission analysis of the grid patterns indicates that grid shape and line thickness/length in addition to mounting tape were the main variables that influenced results. The hexagons and the square/diamond shape grids provided the highest levels of transmissivity (~ 94%), with their shared walls. For this research, transmissivity was calculated from the measurement of solid ink density (SID) and film density (the PET substrate) – density = log 10 1/T, where T = Transmittance. Transmission density is a function of the amount of light, which passes through the film (X-Rite, 2003). Transmissivity is defined by T^10 . The grid patterns 4 (6.25 μ - 1mm - 6x), 8 (9.525 μ - 1mm -6x), 7 (12.70 μ - 1mm - 4x square) and 11 (12.70 μ - 1mm - 4x diamond) were the best performing grid patterns overall. Anilox line screen and volume did not influence the transmissivity of these four grid patterns. These grids had high transmissivity no matter what anilox roll band was used. Liu, et al demonstrated that a course anilox line screen of 300 cpi - which is much courser than the line screens in this experiment - showed transmission numbers around 95% with a hexagonal pattern (2014). However, mounting tape firmness had a significant impact. For the number 4 grid pattern, the firm tape (3M 1820) provided higher transmissivity readings than the medium soft tape. The Grid Pattern 8, 1820 Grid Pattern 8, 1920 Grid Pattern 11, 1820 Grid Pattern 11, 1920 Figure 7, comparison of mounting tape material four highest transparency grid patterns are two hexagonal patterns and the square and diamond patterns. Figure 8 illustrates the graphical analysis of transmissivity for each mounting tape, grid pattern and anilox roll band.

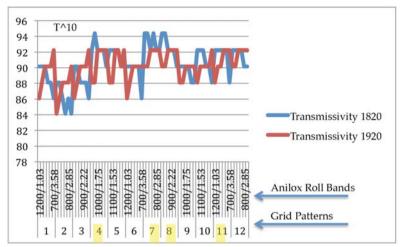


Figure 8: trasmissivity readings comparing stickyback and anilox configuration

8. The resistance of the grid patterns was influenced predominantly by anilox volume and mounting tape firmness. The "heavier" volume of 800 cpi/ 2.85 bcm anilox band and the 3M 1820 (firm) tape provided resistance numbers less than 15 ohms. Again, the four grid patterns of 4, 8, 7, and 11 (hexagonal and diamond/square) showed lowest mean resistance compared to the other grid patterns. The graph in Figure 9 shows the differences in resistance with high volume anilox and firm tape to grid pattern.

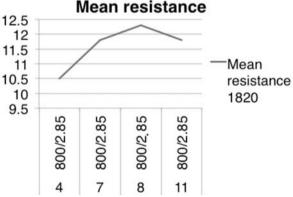


Figure 9: resistance by grid shape and stickyback type

9. The most successful grid pattern was a hexagon pattern with its shared "walls", and hexagons, more so than squares or diamonds, maximize open area. Optimization of all parameters in the flexographic printing process is required in order to print fine conductive lines reliably. Maximizing the open area of the grid is necessary for high transmissivity.

Liu, et al used a transparency-to-resistance ratio (Q ratio) to partly demonstrate the efficiency of a printed grid. The hexagon shape had the best Q ratio of low resistance and high transmission. In addition, it was shown that transmission increased as line length increased, but transmission decreased as line width increased (2014).

To determine the best line length to width ratio, we calculated the transmission of grid pattern number 4 (6.25μ - 1mm – 6x), the best performing hexagon pattern in the experiment, as it relates to conductivity and transmissivity. The calculated transmission on a 1.5 cm2 grid indicated that with a 577 μ length line (1mm smallest width across hex pattern) and the line width at 6.35μ , the ratio of line length/width is \sim 90:1 and the calculated transmission is 95.20%. Figure 10 shows the calculation of the ideal line length of our experiment, comparing line lengths to the same 6.35μ line width of grid pattern number four. Comparing 100, 250, 400, 500, 577, and 1000 μ length lines, the 577 μ line length at the constant 6.35μ width line provides the ideal parameters. Figure 11 illustrates the pattern visually.

R _s	Line length (µm)	Line width (µm)		Transmission (T)	Estimated resistance (R) [Rs=L/W]	Q ratio (T/R)
0.37	100	6.35	15.7480315	64.10%	5.826771654	0.110007615
	250	6.35	39.37007874	88.18%	14.56692913	0.060537583
	400	6.35	62.99212598	92.93%	23.30708661	0.039871448
	500	6.35	78.74015748	94.42%	29.13385827	0.032409669
	577	6.35	90.86614173	95.20%	33.62047244	0.02831667
	1000	6.35	157.480315	97.29%	58.26771654	0.016696486

Figure 10: theoretical Length/width ratio

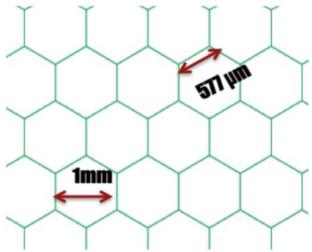


Figure 11: 6.35μ line width – 1mm in width across (diameter of in circle) hex – line length of $577\mu = 90:1$

10. This theoretical calculation of ideal line length/width ratio for the hexagon grid pattern to achieve a high transmission and low sheet resistance was borne out in the actual conductivity measurements of the various grid patterns printed for this experiment. Figure 12 illustrates that when the "high" volume anilox band of 800 cpi/ 2.85 bcm (E) was used with firm tape (3M 1820) and grid pattern number 4, the resistance measured in the horizontal/vertical/diagonal areas of the grid (10, 10.4, 11.1 ohms) resulted in a transmissivity reading of 92.2%. The transmissivity readings are lower than the predicted calculation, and the resistance readings are also lower. Some of this may be explained by the fact that the calculation does not take into consideration printing characteristics such as image gain and ink opacity. For example, the printed silver conductive inks are actually translucent and not entirely opaque. If we use 25+u as our line width, the resistance lowers, but transmissivity also is lowered. The data in our actual measurements also indicates that grid pattern number 8 (9.525u -1 mm - 6x) also performs well at that anilox configuration (E), but the O ratio is lower, as resistance is higher. Grid pattern number 1, by comparison (6.35µ - 500u - 6x) has a poor O ratio—lower resistance, but transmissivity was lower (line length smaller).

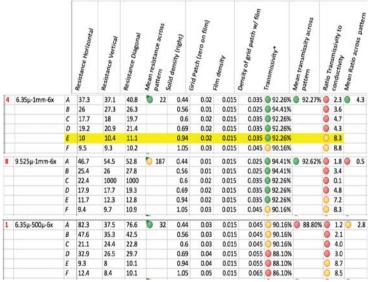


Figure 12: actual transmissivity readings

11. In flexographic printing, many types of ink are not entirely opaque, which can affect transmission readings. Line widths do not "gain" uniformly, and this will impact resistance. Resistance varies with line uniformity variations. Any artifacts in the printing process such as scratches on the film will further impact performance. As illustrated in Figure 13, slight marring or surface scratches caused by dirty rollers or other mechanical impacts on the PET film may not negatively impact graphic printing, but can impact the functionality of the grid by causing shorts in the TCF.



Figure 13: scratches in PET

Conclusions

The purpose of this paper was to investigate the variables in the flexographic printing process that affect the way ink is transferred to the substrate from the printing plate. The research sought to determine if the following variables could be optimized with commercially available technology to impact and exploit better reproduction of fine features and conductivity/transparency for TCFs: plate imaging, exposure, and processing, plate material, mounting tape modulus, and anilox line screen and volume.

We found that high resolution plate imaging, "flat top dot" plate exposure, mounting tape hardness and anilox volume were all important variables which influenced the ability print TCFs with high transmissivity and low mean resistance. The shape of the grid pattern was also an important element, as the hexagon pattern yielded the best overall measurements. For the press variables that were tested, an anilox volume of 2.85 bcm and firm tape proved to give lowest mean resistance. The firm tape was the key for transmissivity, in addition to the shape of the grid pattern – hexagon.

For the printing of TCFs by flexography using water-based, nano-particle silver ink on PET, the following are recommended parameters:

- Hexagonal shape with minimum line width as determined by the flexo plate image setter. 4000 dpi is more common in graphic printing environments, but the higher resolution image setter, the finer feature can be held on the plate.
- After determining the minimum line width achievable on the plate, create hexagons with a line length/width ratio of 80-100.

- Control press parameters to maintain line integrity. This experiment was run on a very fine-tuned press with excellent hygiene. Minimal impression, and impression variances, good ink metering, and tension control are essential.
- "Heavier" volume anilox (~ 2.85 bcm) provided lower resistance.
- "Firm" mounting tape (3M 1820) proved to provide more "gain" in features, but the printing results were more robust, bereft of pinholes and skips that would cause shorts.

The optimization strategy for printing grid lines, given that light transmission and low resistance are opposing variables, should be as follows:

- Determine the minimum line width able to be reproduced in print on various axes. Control imaging of the photopolymer plate and exposure to get as close to a 1:1 file: feature in the processed plate, and on press.
- Determine the ideal "open area" (length and width of lines) based on resolution required if a touch-screen, resistance requirements, and redundancy to eliminate an open circuit.
- Optimize the transparency-to-resistance ratio (Q ratio)

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