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# Sustainable substrate for printed electronics

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## Abstract

*This paper focuses on evaluation of paper as a substrate for printed flexible electronics. The motivation for using paper as a substrate comes from its attractive recycling and composting properties. In electronics manufacturing the majority of the materials used come from the substrate (>>50%). Thereby, the use of a sustainable substrate is the first step towards sustainable flexible electronics. The next step would be evaluation and development of the other materials required, such as inks and adhesives, for minimal environmental impact. This paper presents results on evaluation of existing commercial printed semiconductor and conductor ink performance on different paper substrate grades. The ultimate goal was to evaluate the potential of semiconductor and conductor materials as part of an NFC powered electrochromic display [1].*


## Introduction

Modern electronics are filled with circuit boards on which various metals and plastics are soldered together. Some of these materials are toxic or break down into toxic substances. Electronics made of paper are viewed as a potentially cost-effective alternate in various applications. Use of this flexible and foldable substrate material enables the use of high-speed printing methods, such as inkjet and screen printing, that both are roll-to-roll compatible. The semiconductor industry is capital intensive and materials represent 10 to 15% of the full value. For printed electronics the situation is different and the material share increases to considerable range making a potential overall market for the printed electronics materials industry highly interesting, also from material sustainability and recyclability perspective [2]. It is estimated that additive manufacturing processes, powered by electricity generated from renewable energy, uses one tenth of the materials of traditional factory production, resulting in a dramatic reduction in $CO_2$ emissions and the use of the earth's resources [3].

At the moment printed flexible devices are primarily being fabricated by utilizing polymeric substrates and circuit boards are manufactured on stiff substrates (e.g. FR4). Flexible printed circuits offer several advantages compared to rigid circuits, including reduced package dimensions, reduced weight, and optimization of component real estate [4], thus making sustainable substrates attractive. Paper's low cost and many applications make it an attractive substrate, but its high roughness and absorbency has been mentioned to create challenges for printed electronics [5].

## Materials and methods

Six substrates were used for the experiments: 5 paper substrates and one plastic substrate:

- Paper 1 - Arjowiggins Creative Papers Powercoat XD 125 (125 µm thick)
- Paper 2 - Arjowiggins Creative Papers Powercoat HD-95 (95 µm thick)
- Paper 3 - Arjowiggins Creative Papers Powercoat HP HN230 (230 µm thick)
- Paper 4 - Stora Enso Lumisilk 130 g/m2 (111 µm thick)
- Paper 5 - Photographic paper from Intelicoat Technologies (254 µm thick).
- Plastic substrate - PET (PolyEthylene Terephthalate) DuPont Teijin Films Melinex ST506 (125 µm thick)

The paper manufacturer has tailored the three Arjowiggins paper grades for printed electronics (Papers 1-3). Paper grade from Stora Enso (Paper 4) was selected since it has been found suitable for printed electronics in earlier studies [6-7]. PET is used in printed electronics extensively and it was used as a reference. Photographic paper was selected for inkjet printing since it has been found compatible with many functional inks in earlier studies [8-9].

Printings were done with screen printing and inkjet printing so different inks were used for the methods.

For inkjet printing two conductive polymer inks and two silver nanoparticle inks were used:

- Inkjet ink 1 (IJ1): Clevios P Jet 700, PEDOT:PSS (Poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate)) based conductive polymer ink from Heraeus
- Inkjet ink 2 (IJ2): IJ-1005, PEDOT:PSS based conductive polymer ink from Agfa Materials
- Inkjet ink 3 (IJ3): ORGACON SI-J20x, silver nanoparticle ink from Agfa Materials
- Inkjet ink 4 (IJ4): ANP DGP40LT 15C, silver nanoparticle ink from Advanced Nano Products

For screen printing one conductive polymer ink and five metal conductive inks were used:

- Screen ink 1 (Screen1): ORGACON EL-P5015, PEDOT:PSS based conductive polymer ink from Agfa Materials
- Screen ink 2 (Screen2): LS-411AW, silver paste from Asahi Chemical Research Laboratory
- Screen ink 3 (Screen3): CI-1036, silver ink from Engineered Conductive Materials
- Screen ink 4 (Screen4): IPC-114, 100 % solids silver paste from Inkron
- Screen ink 5 (Screen5): XCMB-590, silver-carbon blend from PPG Industrial Coatings
- Screen ink 6 (Screen6): Smart'Ink S-CS21303, silver nano-particle ink from Genes'Ink

Inkjet printing was carried out with laboratory scale multinozzle inkjet printer based on single use printhead cartridges (DMP-2850 from Fujifilm Dimatix) with 10 pl drop size. During inkjet printing drop formation was optimized by modifying the waveform, and multiple ink layers were used in order to achieve sufficient thickness with PEDOT inks. With nanoparticle inks one inkjet printed layer was sufficient.

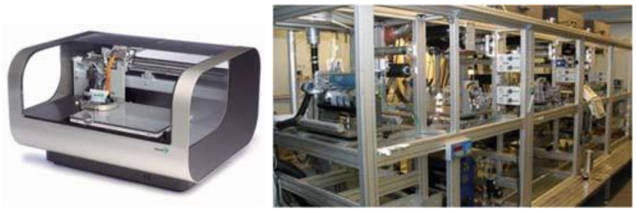

*Figure 1. Printing equipment used: on left laboratory-scale inkjet printer and on right rotary screen printer.*

Screen printing was carried out with roll-to-roll rotary screen printer ROKO with up to 10 m/min printing speed. Different mesh sizes were used to variate the achieved print quality, layer thickness and resistivity. The mesh sizes used were 125, 215 and 305 mesh/inch for PEDOT:PSS inks and 215 and 305 mesh/inch for metal inks. Two different mesh types for 125 and three for 305 meshes were used (Table 1).

**Table 1. Screen types** and their properties **from SPGPrints** used in rotary screen printing trials.

| Screen type | Mesh count - Open area (%) - Wet ink layer thickness (µm) |
|---|---|
| RotaPlate® 125V | 125L - 42 % - 26 µm |
| RotaPlate® 125W | 125L - 43 % - 45 µm |
| RotaPlate® 215V | 215L - 29 % - 18 µm |
| RotaPlate® 305S | 305L - 21 % - 11 µm |
| RotaPlate® 305V | 305L - 22 % - 15 µm |
| RotaPlate® 305M | 305L - 17 % - 8 µm |

Print layout consisted of lines of different width and compact areas for both printing methods.

Optical and electrical characterization of the printed areas was carried out. Optical characterization included visual observations from microscopic images, line width measurement to determine ink spreading, roughness measurement and layer thickness measurement. Electrical characterization was carried out by measuring sheet resistance.

## Results

Conductive polymer inkjet inks (IJ1 and IJ2) were inkjet printed on all substrates. The printing frequency was 1 kHz and resolution 1270 dpi. The amount of ink layers printed were 1, 3 and 5. Inkjet printing of these commercial PEDOT:PSS based inks presented challenges in drop formation so the waveform was carefully fine-tuned. Substrate compatibility also was not very good and the best printed surfaces were achieved on photographic paper (Paper 5) and on Lumisilk paper (Paper 4) (Figure 2 and 3). With IJ2 ink only areas on Lumisilk paper were conductive due to layer unevenness and cracks on the other substrates. With IJ1 ink conductivity was measured only on PET and Paper 1 substrates.

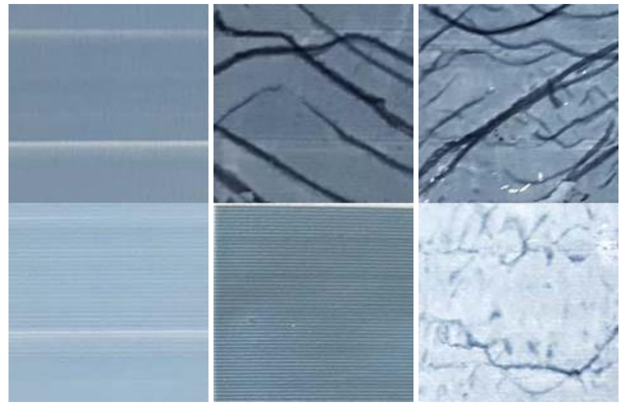

*Figure 2. Five layers of Clevios ink (IJ1) on different substrates. On top from left Paper1, Paper 2 and Paper 3. On bottom from left Paper 4, Paper 5 and PET.*

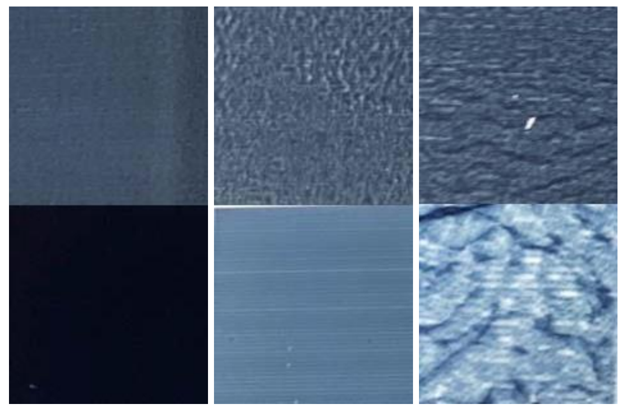

*Figure 3. Five layers of IJ-1005 ink (IJ2) on different substrates. On top from left Paper1, Paper 2 and Paper 3. On bottom from left Paper 4, Paper 5 and PET.*

Due to the high roughness of the printed surfaces and inks absorbed into the paper pores, it was not possible to measure reliable thickness values required for resistivity calculations. As a result, only sheet resistance values were measured and those were in the range of 25-7000 Ohm/square depending strongly on type of substrate and substrate-ink combination. The smallest sheet resistance was on PET with IJ1 ink and the highest on Paper 1 with IJ2 ink. The results indicate that with conductive polymer inks the properties of the paper substrate are crucial.

Inkjet printing of the commercial silver nanoparticle inks was successful after fine-tuning printing settings, such as resolution and waveform. For IJ3 ink the optimal print resolution was 1690 dpi, and for IJ4 ink either 1270 dpi (Paper 2 and Paper 4) or 2540 dpi (other substrates). The properties of the paper surface affected strongly the achieved print quality and resistance, but on all paper grades printed structures were conductive (Figures 4 and 6). The effect of oxygen (O2) plasma was also evaluated with IJ3 ink and it had a significant effect on observed visual quality (Figure 5). The same resistance was achieved on some of the paper substrates than on PET substrate, but detail rendering was much better on paper substrates than on PET (Figure 8). Thereby, paper substrates enable printing of smaller details than PET.

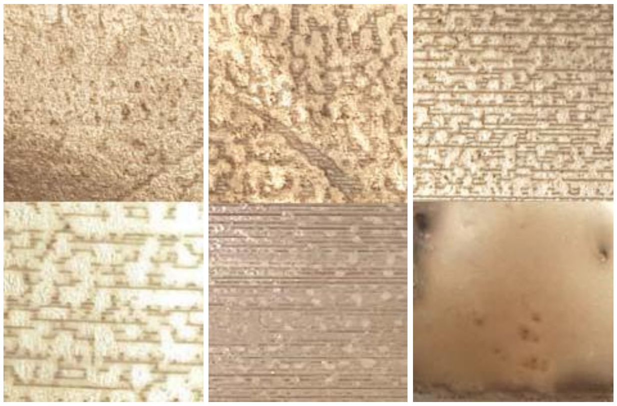

*Figure 4. Orgacon silver nanoparticle ink (IJ3) on different substrates. On top from left Paper1, Paper 2 and Paper 3. On bottom from left Paper 4, Paper 5 and PET.*

*Figure 5. Orgacon silver nanoparticle ink (IJ3) on different plasma treated substrates. On top from left Paper1, Paper 2 and Paper 3. On bottom from left Paper 4, Paper 5 and PET.*

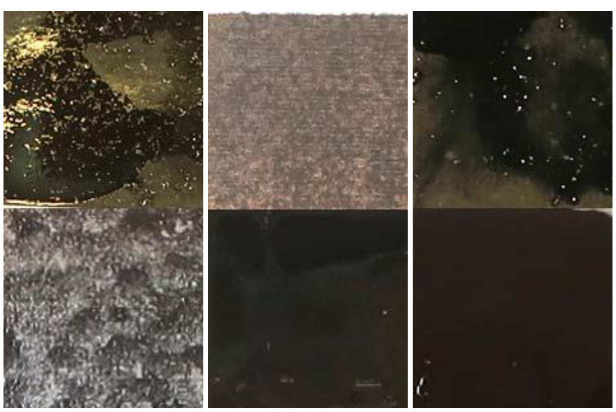

*Figure 6. ANP silver nanoparticle ink (IJ4) on different substrates. On top from left Paper 1, Paper 2 and Paper 3. On bottom from left Paper 4, Paper 5 and PET.*

The measured sheet resistance of silver nanoparticle inks on different substrates were in the range of 0.01-1 Ohm/square (Figure 7), except one ink-substrate combination had a significantly higher resistance (IJ4 on Paper 3). Detail rendering was defined by measuring line width of the printed lines (Figure 8). With ANP ink (IJ4) the lines were of poor quality and discontinuous thus resulting in no conductivity. With Orgacon ink (IJ3) the lines were mostly of good quality and resistance values are presented in Figure 9. However, even with this ink not all lines were conductive. The best lines with visual observation were on photographic paper (Paper 5) and on Lumisilk paper (Paper 4). However, the best detail rendering was on papers tailored for printed electronics (Paper 1 and Paper 2) although the lines were not conductive (Figure 9). The achieved line width is much closer to targeted line width compared to all the other substrates (Figure 8). This might be due to other print quality defects observed visually on the printed surfaces (Figure 4-5) because of ink-substrate incompatibility.

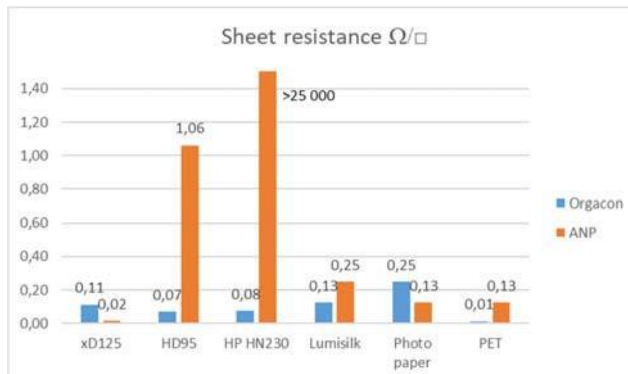


*Figure 7. Sheet resistance of inkjet printed silver nanoparticle areas on different substrates.*

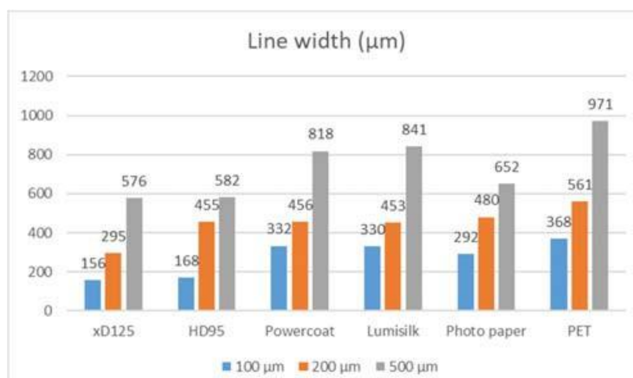


*Figure 8. Detail rendering of inkjet printed silver nanoparticle inks on different substrates.*

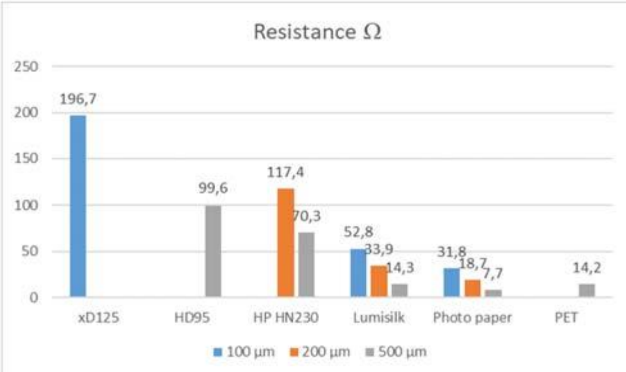


*Figure 9. Resistance of inkjet printed silver nanoparticle lines on different substrates.*

For rotary screen printing only substrates Paper 2, Paper 4 and PET were used due to only those available in roll format. With PEDOT:PSS ink all different mesh sizes and types were used with Lumisilk paper (Paper 4). With the other substrates only one screen type was used to check the printability of Screen 1 ink on other substrates: 215V with HD-95 paper (Paper 2) and 305S with PET

Rotary screen printing of the commercial PEDOT:PSS paste (Screen 1) was successful onto different paper grades and PET substrate (Figure 10 and 11). The printed layers had well-defined edges and rather nice coverage. On PET, the ink had some transfer issues at the trailing edge of the printed patterns. The layer thickness increased and the sheet resistance decreased as more ink was transferred onto the substrate, as seen in Figure 12 and 13. The sheet resistance values ranged from 60-400 Ohm/square depending on the screen type and the substrate. The lowest sheet resistance values were achieved on the smooth PET substrate although the print quality was poorer. On HD-95 paper (Paper 2), the sheet resistance was higher than on Lumisilk paper (Paper 4). Detail rendering of the PEDOT ink on different substrates is presented in Figure 14. The minimum achievable gap size increased when more ink was transferred onto the substrate. On smooth PET, the ink could spread more, thus increasing the minimum gap size. The minimum conductive line was 100-200 µm.

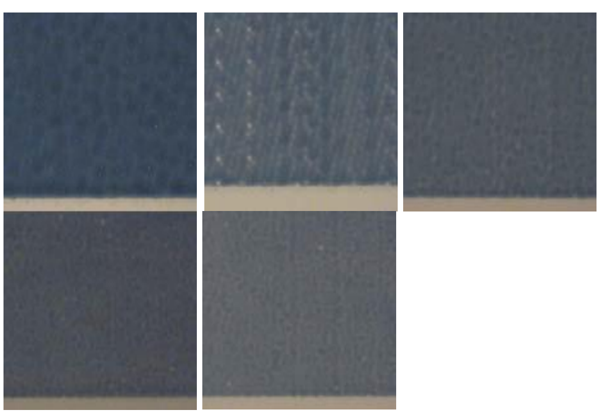

*Figure 10. Rotary screen printed PEDOT layers (Screen 1) on Paper 4 substrate using different screen types. On top from left 125W, 125V and 215V. On bottom from left 305S and 305V.*

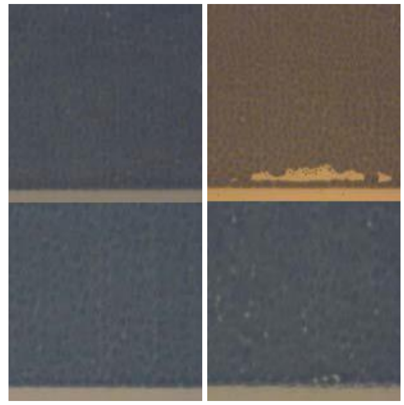

*Figure 11. Rotary screen printed PEDOT layers on different substrates. On top from left Paper 4 and PET using 305S screen. On bottom from left Paper 4 and Paper 2 using 215V screen.*

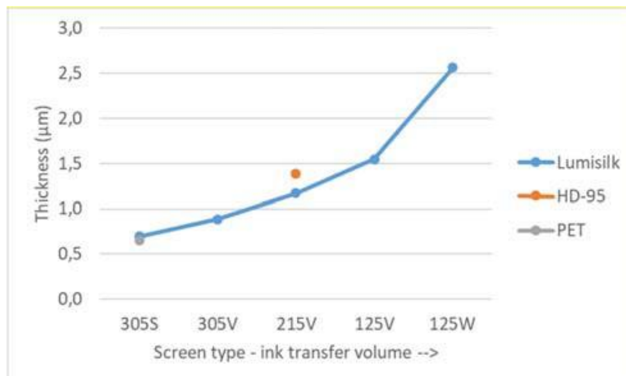


*Figure 12. Layer thickness of rotary screen printed PEDOT layers on different substrates and using different screen types.*

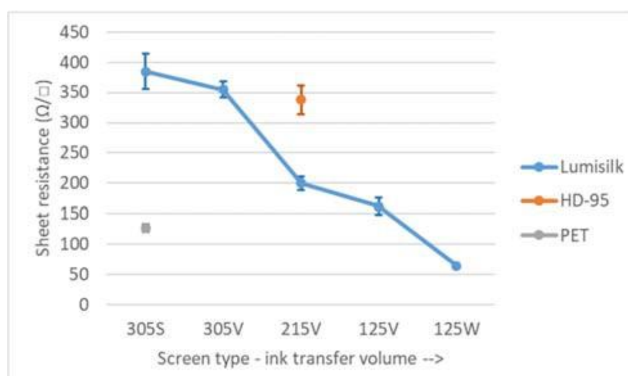


*Figure 13. Sheet resistance of screen printed PEDOT:PSS ink on different substrates with different screen types.*

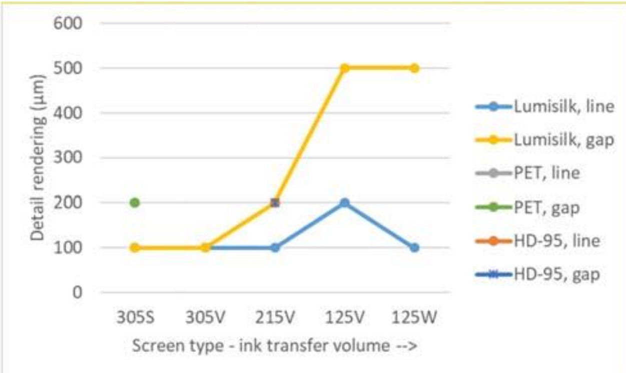


*Figure 14. Minimum line and gap widths of rotary screen printed PEDOT layers on different substrates and using different screen types.*

Silver inks were screen printed with 305 mesh, except Screen 4 ink was printed with 215 mesh because of its larger particle size

The rotary screen printing of commercial silver pastes was successful onto both PET and paper substrates, as shown in Figure 15 and 16. The layers had good coverage and good detail rendering. The minimum gap width was 100-500 µm and the minimum conductive line width was 100-200 µm depending on the ink, screen type, and the substrate. Better detail rendering was achieved on papers than on PET due to the smaller ink spreading (Figure 17 and 18). On PET, the print quality was also poorer due to ink transfer issues at the trailing edge of the printed patterns.

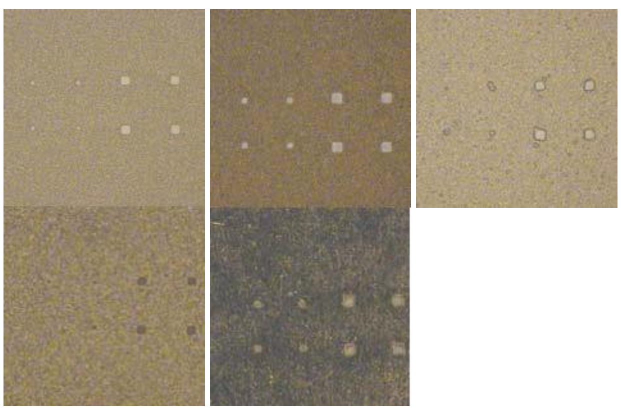

*Figure 15. Rotary screen printed silver layers on Paper 4 substrate. Five different inks are printed. From top left Screen 2, Screen 3, and Screen 4 and from bottom left Screen 5 and Screen 6. The size of the square gaps are 100 µm and 200 µm.*

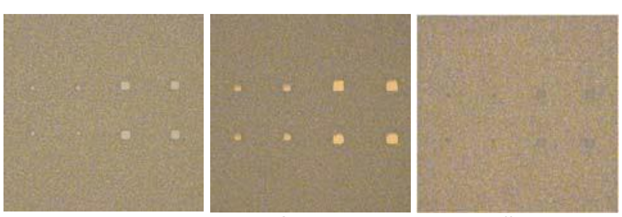

*Figure 16. Rotary screen printed Screen 2 silver layers on different substrates. From left Paper 4, PET, and Paper 2. The size of the square gaps are 100 µm and 200 µm.*

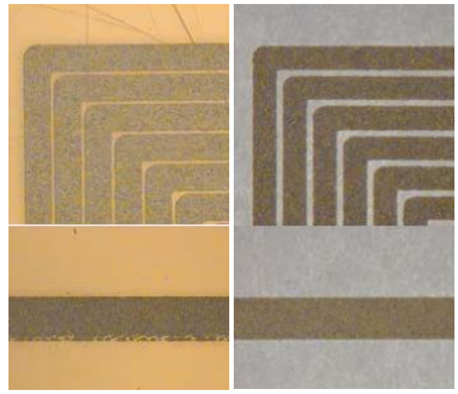

*Figure 17. Rotary screen printed silver layers on PET (left) and Paper 4 (right) substrates using Screen 3 ink.*

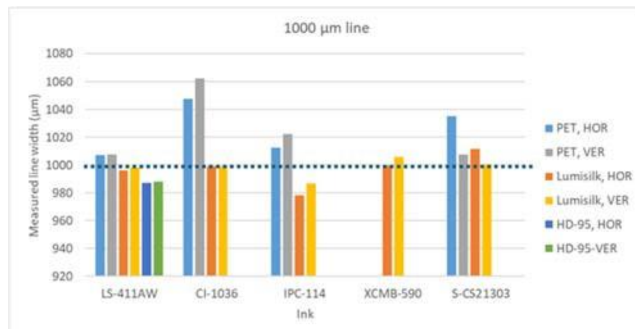


*Figure 18. Line with of 1000 µm screen printed lines with different inks on different substrates.*

The sheet resistance values were 10-140 mOhm/square depending on the substrate and the ink (Figure 19). On papers, lower sheet resistance values and better print quality and detail rendering were achieved than on PET. The lowest sheet resistance was obtained with Screen 4 ink. This resulted from the 100 % solids content of Screen 4 ink and the use of coarser screen, thus leading to higher layer thickness. Screen 5 ink contained also carbon which increased the sheet resistance of the printed layer. The conductivity of the nanoparticle Screen 6 ink was good when taking into account its low thickness.

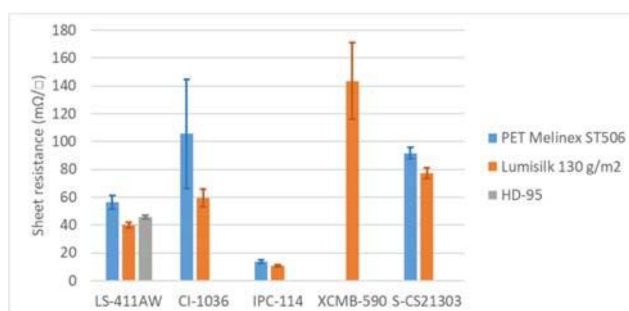


*Figure 19. Sheet resistance of rotary screen printed silver inks on different substrates.*

**Table 2. Layer thickness of rotary screen printed silver layers on different substrates.**

| Ink | Thickness (µm) |
|---|---|
| Screen 2 | 10.9 (PET)<br>12.8 (Paper 4)<br>9.2 (Paper 2) |
| Screen 3 | 4.4 (PET)<br>5.3 (Paper 4) |
| Screen 4 | 26.7 (PET)<br>24.2 (Paper 4) |
| Screen 5 | 3.2 (Paper 4) |
| Screen 6 | 0.9 (PET)<br>1.2 (Paper 4) |

## Conclusions

Both screen and inkjet printing are potential methods for semiconductor and conductor printing on paper. Since screen printing provides thicker layers than inkjet printing, it seems like a more potential manufacturing method, because paper surface non-uniformity does not affect the electrical performance as much as in inkjet printing. However, inkjet printing would be ideal method for certain applications where small material usage, possibility to tailor the printed structure and process flexibility are required. To achieve this it would be necessary to further optimize the ink and substrate compatibility for higher conductivity and better detail rendering. Use of surface pre-treatments, such as plasma, could be one solution.

In this paper specifically PEDOT:PSS inkjet inks presented challenges in achieving conductive structures on paper substrates. The inks themselves are not as high conductive as e.g. metal inks. It is specifically demanding to achieve conductive structures with such low conductive inks on porous and rough substrates, such as paper.

## References

[1] www.supersmart-project.eu

[2] Das, R. and Harrop, P. (2013) Flexible Electronics Forecasts , Players & Opportunities 2013-2023.

[3] Rifkin, J. (2014) Beyond Obama's Plan: A New Economic Vision for Addressing Climate Change, Huffington Post, available at: www.huffingtonpost.com.

[4] Gagliardi, M. (2016) GLOBAL MARKETS FOR ROLL-TO-ROLL TECHNOLOGIES FOR FLEXIBLE DEVICES. Available at: www.bccresearch.com.

[5] Sullivan, M. (2018) Printed Electronics: Global Markets to 2022, BCC Research LLC.

[6] Hakola, L., Gaspar, C., Ilmonen, A., Lehtinen, K., Smolander, M., (2014), Printed intelligence for consumer products.

[7] Sandberg, H., Gaspar, C., Hakola, L. (2014), Integrated Printed Hybrid Electronics on Paper. Proceedings of the 5th Electronics System-integration Technology Conference, ESTC 2014. Institute of Electrical and Electronic Engineers IEEE.

[8] Hakola, L., Maaninen, T., Tuurala, S.; Vaari, A. (2016), Inkjet printable anode ink for fuel cell applications. Proceedings of Printing for fabrication 2016 conference, the 32nd International Conference on Digital Printing Technologies, Manchester, UK (2016). p. 149-152.

[9] Hakola, L., Vehmas, K. Functional ink formulation for individualized Smart Tags. In Proceedings of the International Conference on Digital Printing Technologies, Dresden, Germany, 24–28 September 2018; pp. 211–214.

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