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Research article

Comparative analysis of printed electronic circuits applying different printing technologies in the endurance test

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Abstract: The aim of the study is the question, whether printed electronics circuitry from low-cost printers can be improved in quality by certain ink combinations or after-treatments in order to achieve acceptable results in comparison with circuitry from higher-quality printers. For this purpose, the six samples circuitries from different printers (professional and semi-professional) and different ink combinations (PEDOT: PSS, Silver and Carbon) were subjected to an endurance test of 5 million switching cycles under varying climatic conditions. For this purpose test number of N = 5 experimental evaluations were carried out for each samples (in total of 30 experimental evaluations were done). The results show that, respectable results could be achieved with corresponding ink and post-treatment combinations. This opens up new possibilities for future developments in the field of printers, inks and post-treatments under the aspect of "low-cost".

Keywords: low-cost printers; conductive ink; switch; electronic circuits; printed electronics

1. Introduction

Low-cost inkjet-printed technology is a promising approach for the future. In recent years we could see a significant progress in printing technology as well as pre/post treatment methodologies, different inks formulations [1–4] and substrates. Despite the progress in this field, problems exists in regard to lifetime [5, 6], e.g. cycle bending reliability of printed silver electronics: "…traces of a

125 µm-thick on a polyethylene terephthalate substrate were found to have the lowest characteristic lifetime of about 1000 bending cycles, whereas the traces with the same geometry with a thickness of 50 µm endured hundreds of thousands of bending cycles" [5]. As mentioned in previous articles beside the geometry, the ink itself is a challenge: "When flexed, PEDOT: PSS remained conductive for a lower radius of curvature (10 mm) than silver. Among the printed patterns, the sinewave pattern was observed to be superior for flexible electronics applications" [6]. Furthermore the fact that environmental stability of conductive inks should also be considered [7,8] (e.g. caused by the hygroscopic behavior or pH-value [9]). Next problem which has a high impact is the printing technology itself. Without hesitation we could state that we could see lot of progress in this area, but the cost associated with the high cost printing technology is always high and the low cost solutions are not available in this area. The price ranges of these printers are in the range between € 30,000 for semi-professional and up to more than € 100,000 for professional use. Another challenge could be seen in post and pre-treatment, which is also an investment for the user [10–12]. In addition to the high cost associated with these printing technology there exit other restrictions. Because of the thermoplastic nature of plastic flexible substrates, the high sintering temperature of NP inks restricts the choice of substrates. To lower the ink sintering temperatures on plastic substrates, researchers have tried to make use of other sintering processes, such as microwave, laser curing, plasma treatments, and chemical sintering. Although these alternative methods have given fruitful and plausible results, however thermal post-treatment is still extensively used in printing commercial metal inks because of its wide availability and easy access [13]. To sum up, a lot of research has done regarding inks, printing technology, substrates and pre and post treatment. But recently published articles illustrate, that there is still room for improvement [14–17]. In other word, it is an ongoing topic which needs to be investigated.

Aim of the paper is to figure out, whether it is possible to printout long-lasting (higher lifetime) switching circuits with non-professional printers. This should be realized by combination of different inks (PEDOT: PSS, Silver and Carbon) or post-treatment of the printed switching circuitries. After this phase of study, the aim is to compare these samples with samples (circuits) printed with a professional printer without post-treatment. For the industrial application of these circuits, it is necessary that they withstand millions of switching cycles. For this study, following key performance indicator was selected for the test: (1) Number of switching cycles was fixed by 2 million (Switching on/ off for the printed switch circuits) for the first run and 5 million for the second run. These performance indicators were selected based on the expected lifetime of well-known producers of tactile membrane switches which ranges from > 1 million cycles [18] up to < 5 million cycles [19] as mentioned in a previous article; (2) The applied force on these circuities (printed switching circuits) were fixed to approx. 32 N; (3) Environmental conditions like temperature and humidity also needs to be considered for the test runs. Based on these assumptions the following hypotheses were formulated:

H1: If printed circuits are produced by means of a professional / semiprofessional printer using silver inks, then the life span (measured by electrical resistance) is significantly higher under standard conditions (= 20° room temperature / 50% humidity) than for low-cost printers of non-professional use.

H2: If printed circuits are produced by means of a professional / semiprofessional printer using silver inks, then the life expectancy (measured by electrical resistance) is significantly higher under test conditions in a climate chamber (= 85°C room temperature / 85% humidity alternating with 10% room temperature / 40% humidity) than for low-cost printers for non-professional use.

H3: If printed circuits are produced by means of a low-cost printers for non-professional by using silver inks, then the service life (measured by electrical resistance) under test conditions in a climate chamber (85°C room temperature / 85% humidity alternating with 10% room temperature / 40% humidity) is comparable to those from professional printers, provided the silver ink is protected by an additional conductive carbon monoxide.

2. Method

For the test, six different samples were used in the form of circuits (as shown Figure 1). Three out of six circuits were produced with a low-cost printer and other three with a professional or semiprofessional printer. The following Table 1 gives an overview of the six circuitries. These are to be tested:

Table 1. Overview test samples

Technology	Substrate	Ink	Printer	Treatment
Low-Cost-Printer				
Sample A	PET	Silver	Brother MFC-J6710DW	No
Sample B	PET	Silver and PEDOT	Brother MFC-J6710DW	No
Sample C	PET	Silver and Carbon	Brother MFC-J6710DW	No
Professional-Printer				
Sample D	PET	Silver	Dimatix Materials Printer DMP-2850	No
Sample E	PET	Silver	Novacentrix-Ink mit Epson C88+	YES
Sample F	PET	Silver	Meyer Burger Pixdro LP 50	No

*Note: Sample A–C was produced by means of own pressure, Sample D–F was produced externally.

Afterwards, all the above mentioned samples were subjected to a 5 million switching cycles as a long-term test (with and without climate control).

2.1. Materials and methods – Low-cost-printer technology

As mentioned in previous studies [20–23] the following materials and methods were used to test different printed electronics circuits.

(a) Printer: The switching circuits were printed with the low-cost printer from Brother (Typ: MFC-J6710DW);

(b) Switch Design: The switching pattern was created by using Microsoft Word 2013. The following figure shows the design of the upper and lower switching layer;

(c) Layer Substrate: *Novele*™ from *NovaCentrix* is used as substrate. AgIC Printing System Start Guide recommendations with below mentioned Brother Printer setting [24]: Media Type “Brother Photo Paper BP61”; Print Quality “Best”; Color Mode “Active”; Orientation “Portrait”; were used for printing;

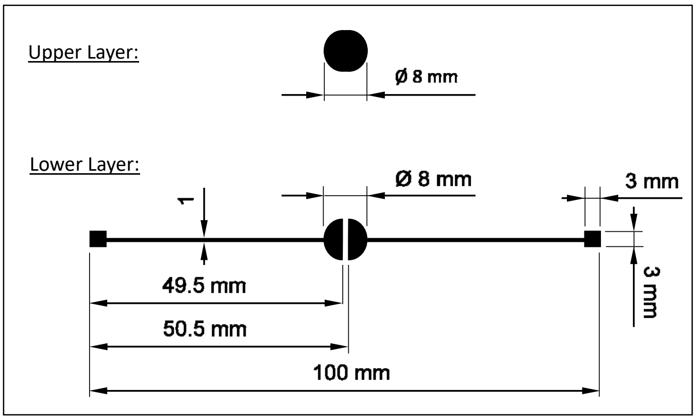

Figure 1. Design of the switching cycle test pattern (upper and lower layer).

(d) Connection: The lower switching layer was connected with two connecting wires, which were glued with standard electro-conductive glue onto the contact pads;
(e) Ink: For printing AgIC Circuit Printer Cartridge Set were used. In this set the silver ink has already been filled into a set of 3 Brother LC71 (US) / LC1240 (Europe) cartridges. For the protection of the printed silver ink on the substrate, one of the following options were employed (i) Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) – 0.8% in H_2O from the company Agfa-Gevaert N.V. (PEDOT:PSS) (ii) Resistive Inkjet Ink #3800 Series from the Methode Development Company as carbon ink;
(f) Printout: The printed layer was not pre-treated or post-treated;
(g) Test device: The membrane switches were tested in a special test device which was developed to create a practical test conditions. The device consists of two short-stroke pneumatic cylinders (= No. 0822406401 from *Aventics Rexroth Pneumatics)* mounted of an aluminum frame with cylinder bore diameters of 12 mm, strokes of 10 mm and a operation pressure of 1–10 bar, a software package to save the data [25] and an Arduino Uno as well as two breadboards for the sensors and the connections were employed for the test;
(h) Test parameter: This includes Electrical resistance, pressure of the compressed air and number of cycles. The measured average force by applying pressure of 3 bar from the test arrangement is 32 N. The number of switching cycles for the initial test run was 2 Million and respectively 5 Million for the second test run;
(i) Optical control: For the measurement of the surface a 3D-Laserscanningmicroscope from Keyence (VK-X100k / X200K) was used;
(j) Test environment without climate chamber was approx. in the range of 20°C and 50% humidity;
(k) Test environment with Climate Chamber: An installation of the company Vötsch type $VC^3$4018 [26] with two test cycles was used: Cycle 1 with a temperature of 85°C and a humidity of 85% with a test duration of 48 hours and cycle 2 with a temperature of 10°C & a humidity of 40% with a test duration of 24 hours. The total test time for a complete cycle was approximately 1,000 hours.

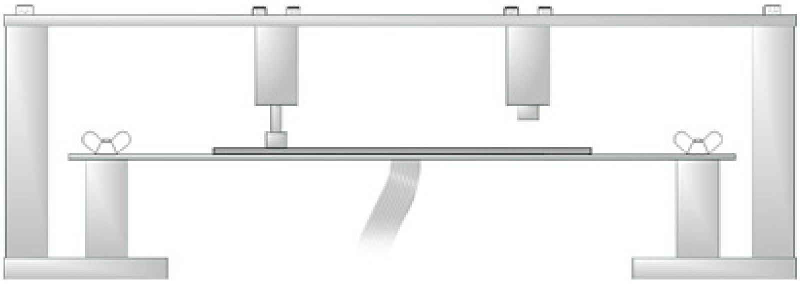

Figure 2. Test arrangement of the switching cycle test.

2.2 Materials and methods – Semiprofessional / Professional-printer technology

Samples from external suppliers were requested for samples D–F, which had to be prepared according to the same specifications as of sample A–C. This means the same geometries and the same substrates: Novele™ from NovaCentrix was used for the fabrication, so that electrical characteristics of all the samples can be compared. For the purpose of testing, sample number of N = 5 for each test pieces (Sample A–F) were used. In total of 30 experimental evaluations were made for the complete cycle of test.

The inks, printers and pre-treatment and post-treatment processes used are described in the following Table 2:

Table 2. Detailed overview sample D–F

Overview	Description
Sample D	Printer: Dimatix Material Printer DMP-2850 with Dimatix-print-head (Professional Printer); Ink: ANP 40LT15C; After treatment: None; Link: http://www.fujifilmusa.com
Sample E	Printer: Epson C88+ with special CISS for color prints (semi-professional use); Ink: JS-B25P from Novacentrix especially developed for Epson C88+; After treatment: Yes, by overnight drying and curing by using PulseForge-technology. Link: http://www.novacentrix.com
Sample F	Printers: Meyer Burger Pixdro LP 50 (Professional Printer) with a print head from Konica Minolta (KN-512 SH); Ink: unknown; After treatment: None; Link: http://www.meyerburger.com

*Note: Specifications based on the information of the external supplier.

2.3 Testing hypotheses H1–H3

Hypotheses H1 was tested under standard conditions by printing an individual silver switching circuit (upper and lower layer) on a PET-foil according to Figure 1. After that for the samples from A–F, upper layer and lower layer are glued together forming a functional switching circuit. Sample B was overprinted with PEDOT: PSS (PEDOT: PSS was coated over functional switching layer printed using silver) and sample C with the organic conductive ink. Sample A was not overprinted. These are connected to the microcontroller device measuring the electrical parameter as well as the switching cycle. They are then installed in the test device mentioned in Figure 2. and tested under standard conditions described above for the test cycle of 2 Million and 5 Million switching cycles. Sample D, E and F were used for the test as supplied from the external supplier, whereas sample A–C was printed with the Brother printer with AgIc ink [24]. For hypotheses H2 and H3 the same procedure was used for the fabrication, but was tested under the conditions of the climate chamber as described above.

3. Results

For the experimental evaluation, total number N = 5 sample piece from each type of sample test piece (from Sample A–F) were taken. In total 30 experimental analyses were carried out in the whole cycle of the experiment. The results from the five sample test pieces were quiet similar (for Sample A–F) and best sample has been used in this section for better understanding and illustration.

For hypothesis H1 the samples A–F were subjected to 2 and 5 million switching cycles under standard conditions in the first step. The results are shown below:

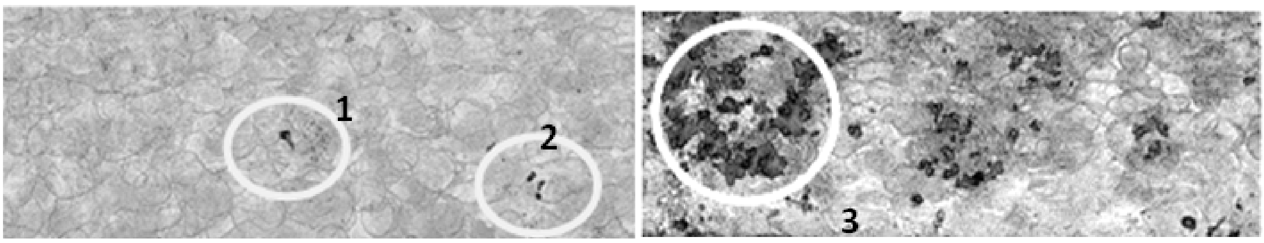

Figure 3. 3D-laser-scanning microscope-recording of the surface of sample A (pure silver ink) after 2 and 5 million switching cycles under standard conditions.

As seen in the Figure 3, there is flaking for Sample A in various areas on the silver surface indicated through white rings 1 and 2. Continued test of the Sample A for 5 million cycles resulted in large flaking in the silver surface indicated through white ring 3 was visible. At the end of the investigation, even though with the surface flaking (silver) the circuit was still functional.

After 2 million switching cycle, Sample B showed (misproportion observed in the surface through 3D-laser-scanning when compared to the initial structure before testing) of overprinted PEDOT: PSS layer indicated through the white ring 1 in the Figure 4 over the entire surface. Continued testing under the standard test condition for 5 million switching exhibits considerable deformation indicated through the white ring numbered 2 in the Figure 4. The silver surface showed no additional damage in both cases, the circuit was functional.

Sample C shows no visible flaking on the silver surface, but deformation (misproportion observed in the surface through 3D-laser-scanning when compared to the initial structure before testing) on the carbon layer, after 2 million and also after 5 million cycles indicted through white rings in the Figure 5 were observed. The circuit remained functional throughout and after the test cycle. The samples D and E showed no significant wear / damage and remained functional, as the following Figure 6 illustrate.

The Sample F from the professional printer Pixdro LP 50 from Meyer Burger showed adhesion problems in the initial stages itself (on the delivery of sample this was observed). This could be possibly caused by transport & storage of the sample or could also due to the incompatible PET substrate film with the ink being used by the manufacturer. Due to this reason the Sample F was omitted for fairness reasons from the test procedure. In context to Hypothesis 2 and 3, the behaviors of the test samples under of 5 million switching cycles were investigated in the climate chamber.

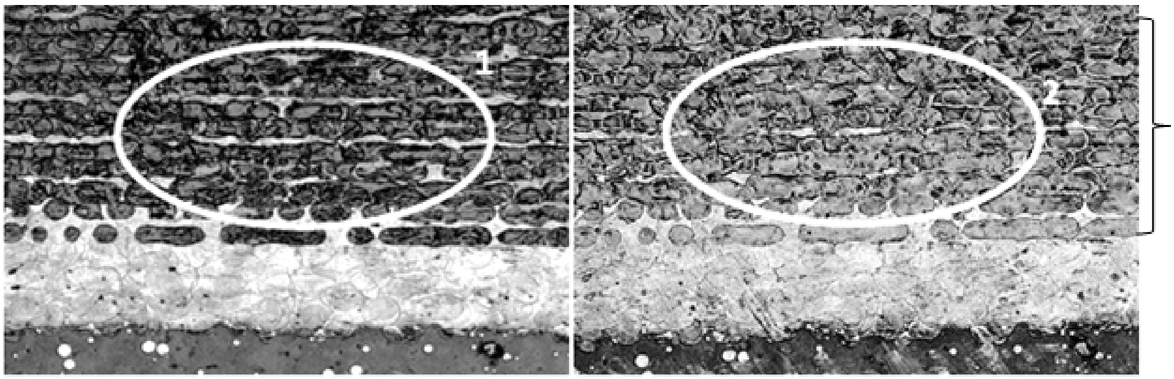

Figure 4. 3D-laser-scanning microscope-recording of the surface of sample B (silver ink overprinted with PEDOT: PSS) after 2 and 5 million switching cycles under standard conditions.

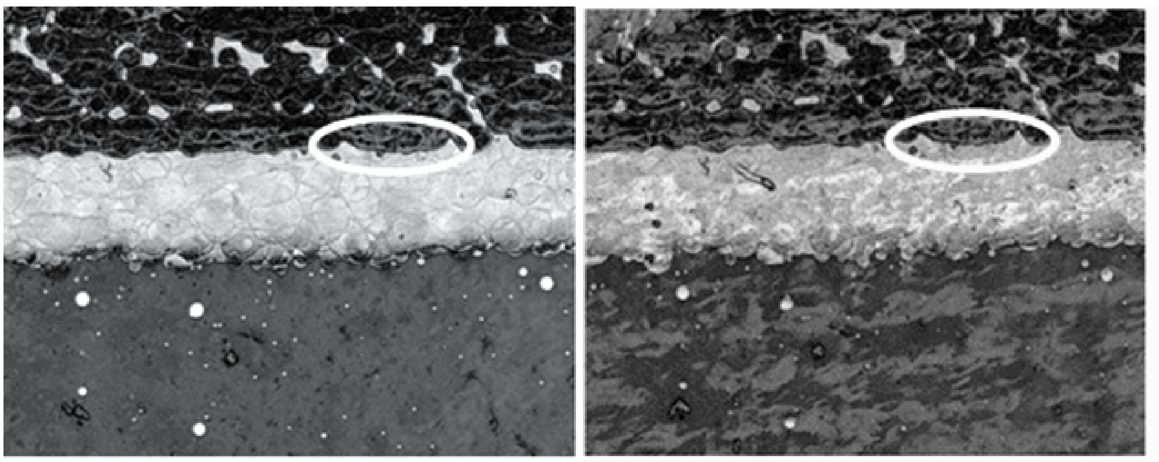

Figure 5. 3D-laser-scanning microscope-recording of the surface of Sample C (silver ink overprinted with conductive carbon ink) after 2 and 5 million switching cycles under standard test conditions. The carbon layer was not applied over the entire surface of silver layer. Due to the force from the test device the silver ink partially shines through.

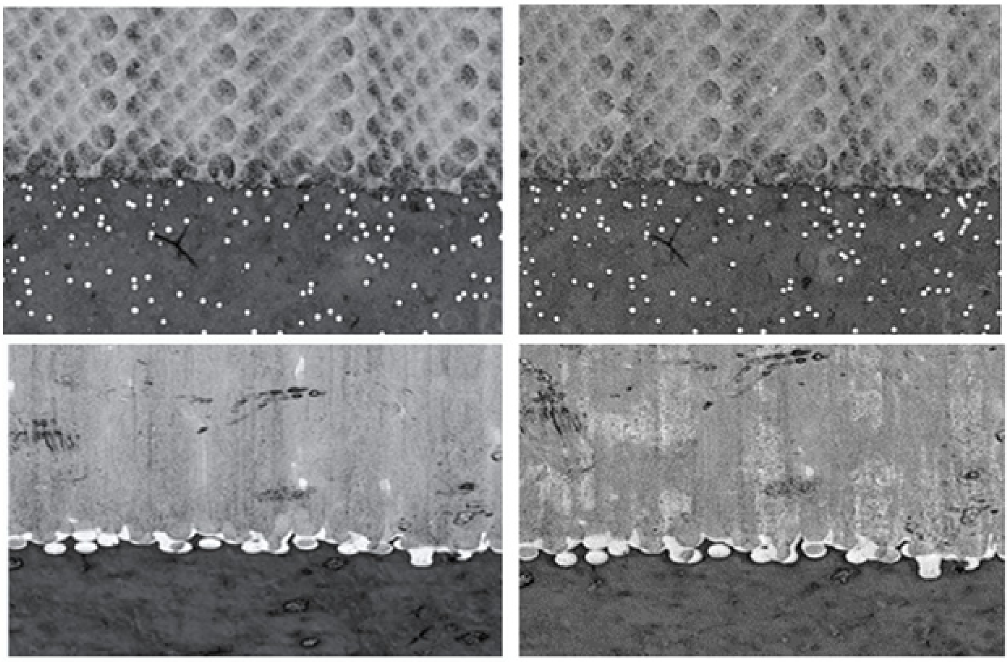

Figure 6. 3D-laser-scanning microscope-recording of the surface of Sample D - Dimatix - and Sample E - Novacentrix - after 2 million and 5 million switching cycles under standard conditions.

Sample A: The results shows that the Sample A (pure silver ink as functional layer) did not withstand the 5 million switching cycle under the climatic conditions (85°C / 85% humidity alternating with 10°C / 40% humidity after approx. 1000 hours). Large area flaking of the entire silver ink surface was visible. This can be visualized in the Figure 8 through the comparison of the 3D-detail-laser image to the 3D-detail-elevation-profile. This difference in the detachment is indicated through the white ring 1 in the laser scanning image in Figure 8. 3D-detail-elevation-profile view of the same affected area indicated by the white ring 2 in Figure 8 showing variance in the colour (green indicating the detachment of silver layer instead of the reddish brown which indicates the silver surface) justifies the detachment of the Silver surface layer after the experimental test run.

From the Figure 8 the detachment of the silver from the surface has only visualized in a few regions indicated by the white ring numbered 2. The detachments were not uniform and this can be confirmed from the scanning images of 3D-detail-laser image (white spacing indicated by white rings indicating the detachment) and 3D-detail-elevation-profile (change in the colour profile from green (indicating the detachment) to reddish brown showing the silver surface which was not affected). The initial electrical resistance measured before the test procedure for the Sample A was 10.414 Ω and the final resistance was 36.354 Ω. The circuit was not functional after the test procedure and the detailed study pointed to a permanent contact between the primary and secondary switching circuit, hence it was no more working as printed switching circuit.

Sample B: For sample B (silver with PEDOT: PSS over coating), certain detachment phenomena were evident, which have been further intensified by the separation of the upper from the lower layer. This is visualized from the Figure 9 through 3D-detail-laser image (deformation in the surface indicated by white rings (1) and 3D-detail-elevation-profile (blue colour indicated in the white ring numbered (2) showing the detachment. It can be assumed that the lower silver layer has completely peeled off at the affected area indicated by the rings. The initial electrical resistance measured before the test procedure for the Sample A was 16.096 Ω and the final resistance was not measureable due the deformation & detachment phenomenon as indicated above. The circuit was not functional after the test procedure.

Sample C: The study of the Sample C (silver ink with carbon over coating) shows the marginal flaking of the surface in the unprotected silver ink surface area similar to the experimental results of the Sample A. This can be visualized in the in the Figure 10 in the 3D-detail-elevation-profile for the difference in the height profile. The Figure10 shows that there is no significant flaking or complete outbreaks in the protected carbon overpressure areas.

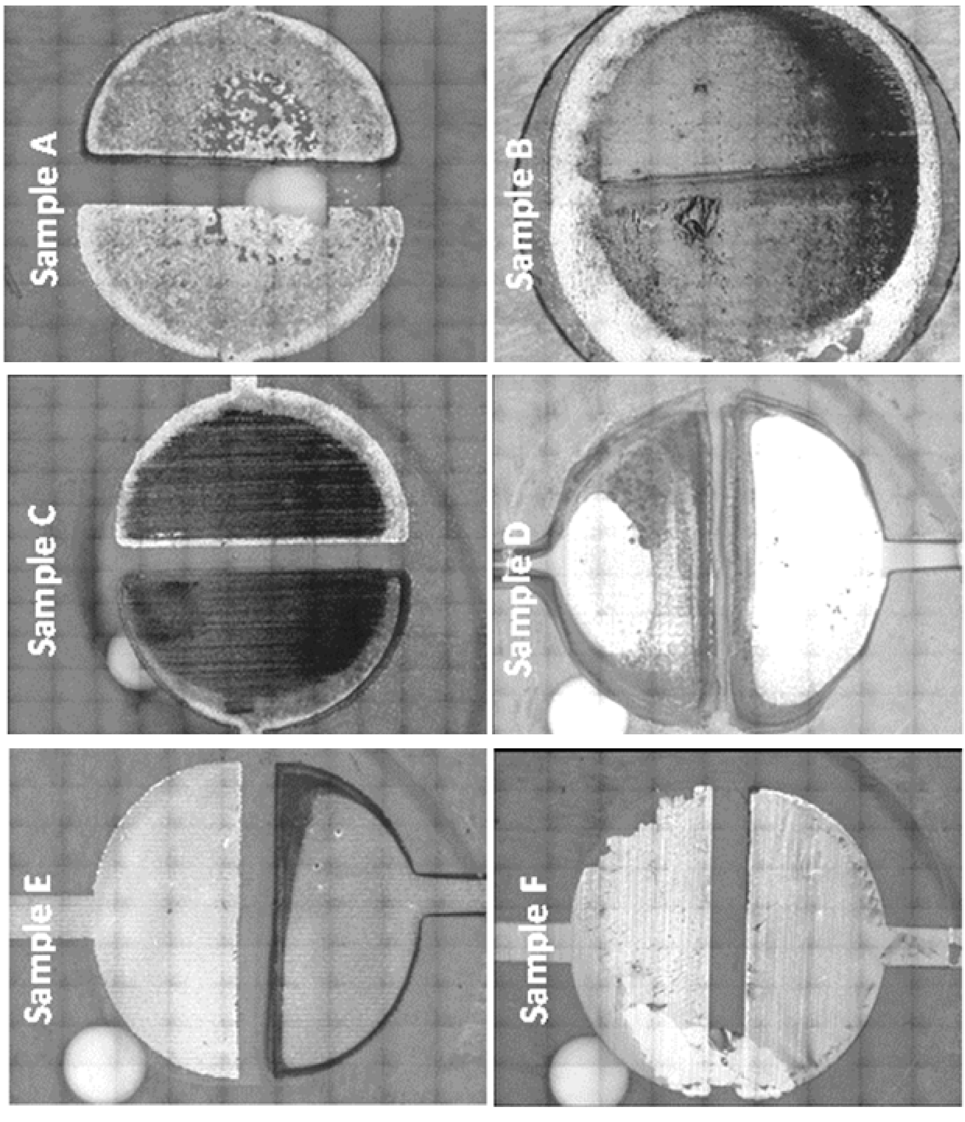

Figure 7. 3D-laser-scanning microscope-recording of the surface of sample A - F after 5 million switching cycles under climatic chamber conditions (= 85°C / 85% humidity alternating with 10°C / 40% humidity after approx. 1000 hours).

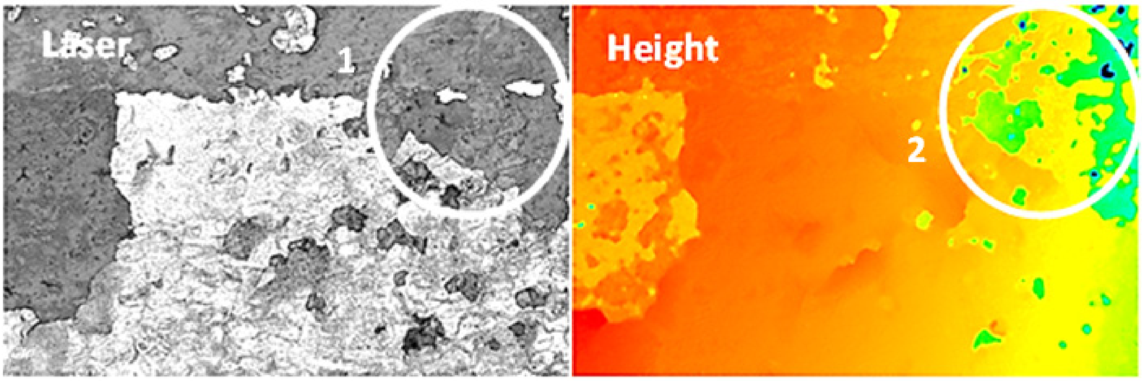

Figure 8. Detachment of the silver inks visualized from 3D-detail-laser image and 3D-detail-elevation-profile for Sample A - Lower switching layer.

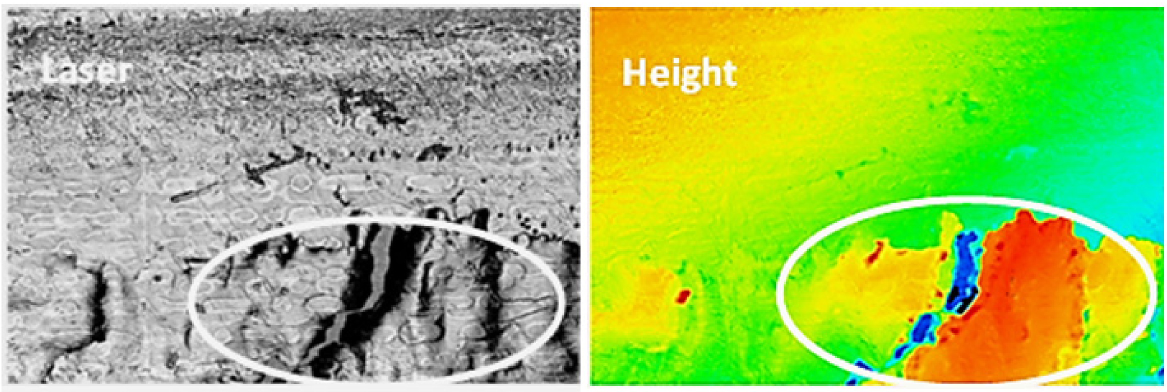

Figure 9. Detachment of the silver-PEDOT: PSS -ink as a laser image and as a height image of Sample B - Lower Layer.

Only in the unprotected area (lower area of the dashed lines, Figure 10) has partial holes indicating the flaking. The protected area with carbon over coating over the silver showed only small superficial flaking. Therefore, two further detail examinations were made as depicted in the Figure 11.

Slight changes in the surface profile can be visualized from the Figure 11 indicated through the white rings. The change in the colour (dot of dark blue) indicates the small superficial flaking of the protected area in the Figure 11. The initial resistance was 12.593 Ω and reduced to a final value of 10.743Ω. The circuit was fully functional.

Sample D: (Dimatix DMP 2850): it can be seen that, according to Figure 12, no significant flaking occurs on the surface, but the printed ink clearly shows changes in the structure (indicated by white rings in Figure 12). This can be due to the temperature changes or the humidity that has penetrated into the composite layer. A 3D-detailed view of the surface confirms that this is largely intact:

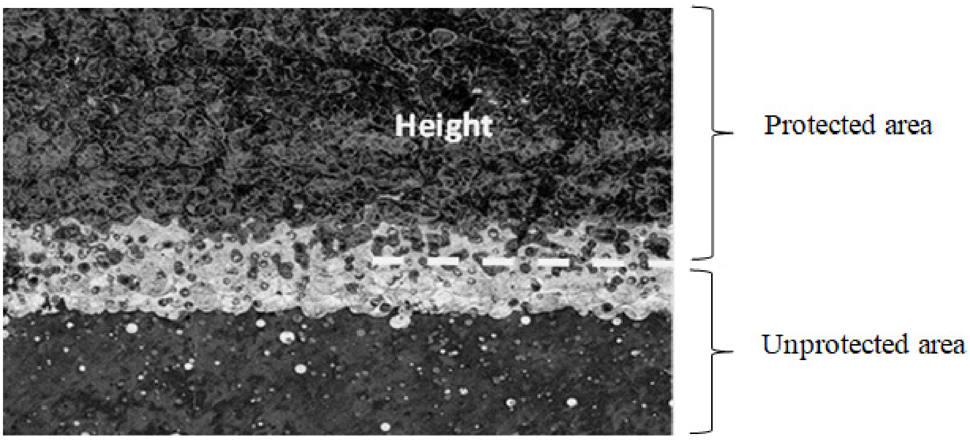

Figure 10. Detachment of the silver ink with carbon coating as laser image and height image of Sample C - Lower Layer.

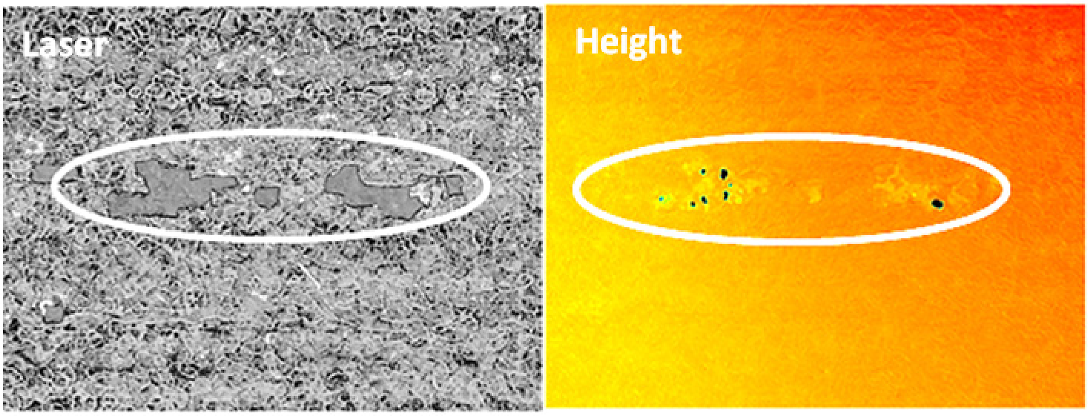

Figure 11. Detachment of the silver ink with carbon coating as a laser image and height image of Sample C - Upper Layer.

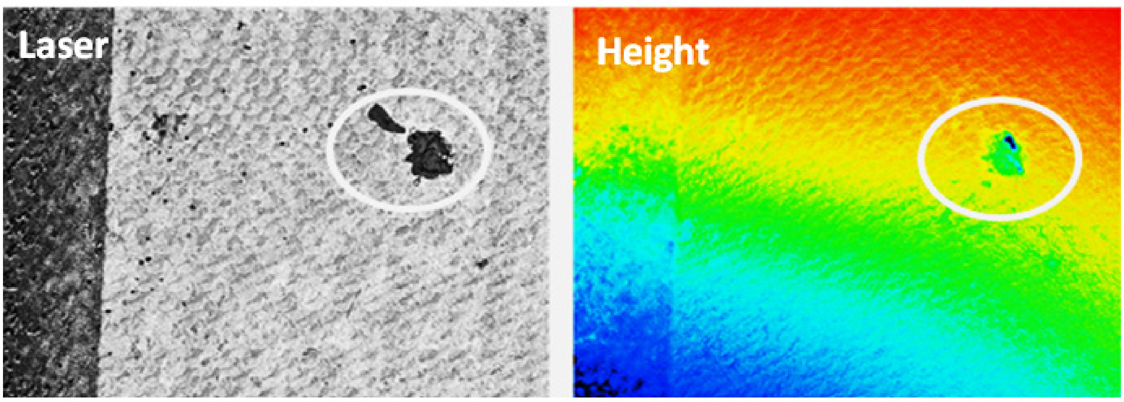

Figure 12. Detachment of the ink for the sample printed using professional Dimatix printer: 3D-detail-laser image and 3D-detail-elevation-profile of Sample D - Lower Layer.

As visualized in Figure12, there are (except for one area indicated by white ring) no significant flaking or eruptions that have occurred on a surface area. The initial resistance was 25.090 Ω, the final value was not measurable since the circuit was not working. There were significant changes in the material which could be due to either the effects of heat or moisture, or both. This change can be visualized in the Figure 7 for Sample D (please refer Figure 7 Sample D).

Sample E: (Novacentrix): The surface of the Sample E shows much significant flaking, but only limited changes in the material. This can be visualized in the Figure 7 in Sample E (please refer Figure 7 Sample E). There is a limited change of the material, as can be seen on the right side of the image in Figure 7 for the Sample E. It is to be assumed that these changes have been caused by humidity since the left side can be regarded as completely intact. To examine this changed area again separately, following detailed 3D-image of the modified region was also carried out here:

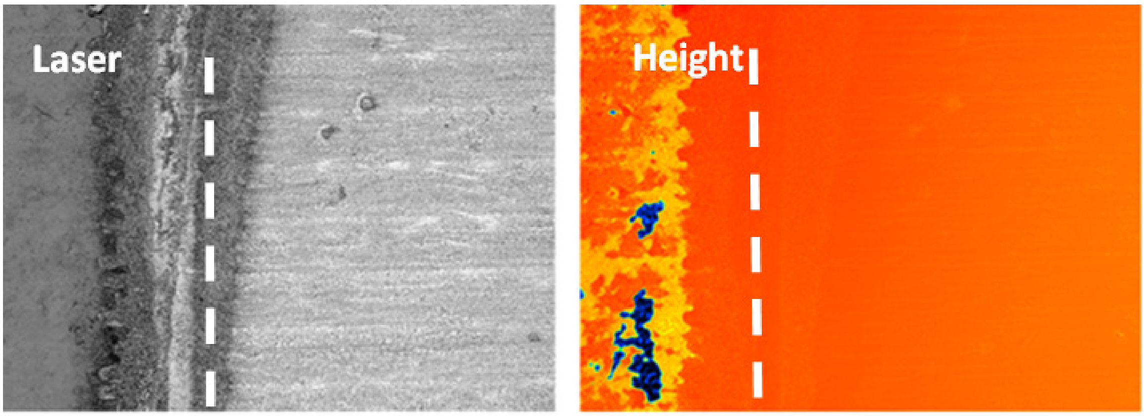

Figure 13. Detachment of the ink from sample by Novacentrix as a laser image and as a height image of Sample E - Lower Layer.

It can be noted that the ink has dissolved in the damaged area and no flaking of the damage occurs in the undamaged area (= see dashed line in Figure 13), so this part is completely intact. The initial resistance was 4.232 Ω and the final resistance was 3.921 Ω. The circuit was fully functional.

Sample F: The last sample F from Meyer Burger shows considerable flaking (visualized in the Figure 7 for Sample F), which however can be resulted from the fact that the ink was not already strongly adhering to the PET film before the start of the test. However, it was striking that despite this disadvantage of the lack of adhesion at the beginning of the test (which may have originated through transport), the ink as such has proved to be relatively stable. In order to take a closer look at this aspect, a further 3D detail image was created as depicted below.

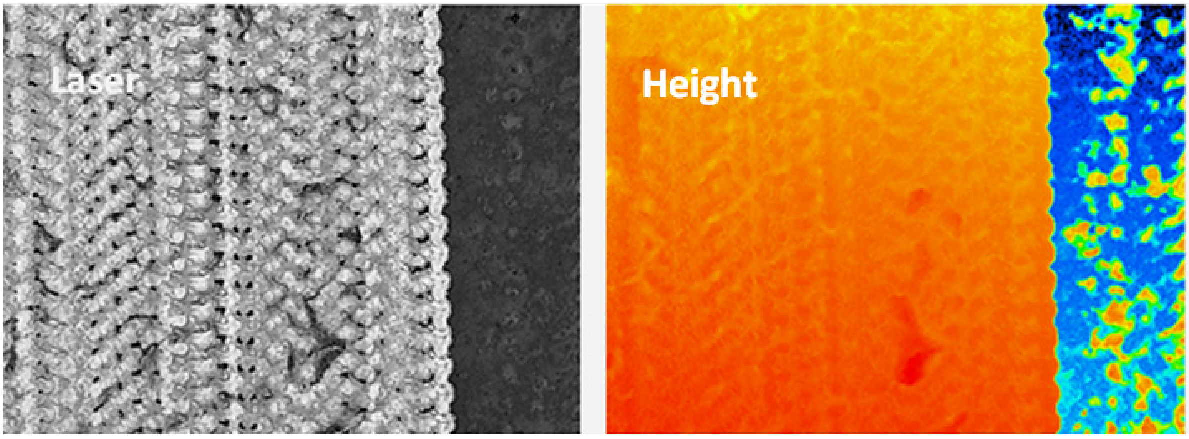

Figure 14. Detachment of the ink from sample by Meyer Burger Pixdro printer as a laser image and as a height image of Sample F - Lower Layer.

The results indicate that as per the Figure 14. the ink from Meyer Burger can be regarded as intact in partial areas which have not been flaked (no colour changes or dots available in the 3D-detail-elevation-profile of Sample F) , since no flaking or damage has occurred in the printed inks as such. The switch was not working at the end of the test.

4. Discussion

The results of the investigation can be summarized as follows:

Hypothesis 1 can be seen as confirmed by comparing samples D, E and F which is printed with professional printer /Post-treated circuits with Sample A which is printed with low-cost printers. Already at 2 million cycles under standard conditions first damage to the silver surface occur (for Sample A), which then significantly continues at 5 million cycles and thus no longer allow use these as a circuit. With regard to Sample B and C, it can be seen that even with a coating of the silver surface a significant improvement in the protection of the underlying silver surface can be achieved. With regard to **hypothesis 2**, it can be conclude that under climate camber conditions the sample A created with the non-professional-use printer completely fails, sample B also largely fails and the sample D and F (produced with professional printers) partly also fail. Only Sample E from Novacentrix silver inks with Pulse-Forge post-treatment and Sample C with silver with carbon coating are shown to be functional under these conditions. With this result also the **hypothesis 3** is confirmed. This assumes that it will be possible to achieve useful results with non-professional-use printers by compensating for the disadvantages of the printer technology by suitable ink combinations (Sample C with a Brother printer) or a surface improved by after-treatment E with an Epson C88 +).

The Limitations are the partially changes of the inks under temperature and humidity conditions. On the one hand the effects of these changes are visible; on the other hand the causes could not be explained due to missing detailed information about the ingredients of the inks. Furthermore, additional attempts with regard to further combinations with other inks or inexpensive after-treatment methods are expedient.

5. Conclusion

The investigations have shown that it is possible to print useful, low-cost circuits by combining suitable inks (in this work silver and carbon ink), while printers with higher quality (compared to the Brother printer used) from the middle price segment are certainly to be preferred for semi-professional / professional applications. High-end printers for highly professional applications and associated investments are definitely justified. However, they are not yet worthwhile in companies with smaller lot sizes. The outcome of the paper points to improvement of quality, lifetime and reduction of cost correlated to printed electronics with recent state of art printers by low-cost circuit printers. Furthermore, the combination of low-cost printers and the use of powerful after-treatment methods is also an interesting option. This will open up new opportunities in the area of flexible and cost effective printed electronic circuitry which will be driving force in future electronics industry.

Acknowledgments

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Conflict of interest

The author declares no conflict of interests in this paper.

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