Conductive inks’ performance is affected by the printing conditions and the substrate’s properties. In this study, one graphite-, one polymer-, and two silver-based conductive inks were printed on four primer-coated metal substrates by screen printing. The compatibility and wettability between the inks and the primers were evaluated by infrared spectroscopy and surface energy measurements. The printed structures were characterized by laser confocal microscopy, peel-off tape testing, and four-point probe electrical resistivity testing. In general, silver inks exhibited the best performance in terms of printability and electrical conductivity. The graphite ink presented the worst printing, adhesion, and functional properties. The polymer-based ink revealed poor wettability but good adhesion and functionality. The surface roughness, energy, and polarity of the primer coating had no significant influence on the electrical conductivity of the printed inks.

1. Introduction

Conductive inks are mainly used in the printed electronics industry to produce printed circuits, organic light-emitting diodes, radio-frequency identification tags, and battery test strips [1–5]. The main advantages of such products are their flexibility, lightweight design, and lower costs [6]. Conductive inks are based on complex formulations with different components. The most important is the conductive component, which may be based on metal or carbon particles as well as conductive polymers. Resins are used to disperse the conductive particles and provide the mechanical and adhesive properties. Solvents are added to dissolve the resins and to control the rheological properties of the ink, and additives are used to adjust the processability or the functional properties [7–10].

Specific challenges in printing conductive inks are achieving continuous and uniform ink deposition and achieving compatibility of the ink to various substrates, both of which have a significant impact on electrical conductivity and other functional properties. Printability is dependent on the ink formulation, content, particle size distribution, and shape of the conductive filler, as well as the surface tension and energy of both ink and substrate. Besides the printing conditions, including the drying/curing process, the substrate’s properties such as permeability, surface roughness, porosity, or surface energy also affect the quality of the printed structures [7, 8].

Conductive inks have been processed by screen printing, gravure printing, flexography, or inkjet printing [2, 7, 11, 12]. Previous research focused on analysing the conductive inks’ performance on substrates, such as paper, glass, and polymer films [2, 11–14]. So far, the printability and functional properties of conductive inks on anticorrosive primer coatings have not yet been investigated. Hence, the main objective of this study is to evaluate the printability and properties of various conductive inks on selected primer coatings for potential use in the printed electronics industry. Furthermore, potential correlations between the structure of the substrate and the functional properties of the printed inks should be evaluated.

2. Materials and Methods

Four commercially available conductive inks based on silver (two different suppliers), graphite, and PEDOT:PSS were
selected (see Table 1). Graphite and silver inks are paste-like solvent inks. The silver ink formulations presented the highest conductive particle fraction of between 60 and 70%. The polymer ink was a gelatinous water-based ink with a relatively low amount of PEDOT:PSS polymer. Accordingly, the sheet resistance was higher for the polymer ink compared to the silver inks.

The substrates were primer-coated 0.5 mm metal sheets. The primers are anticorrosive coatings which also improve the printability of the inks. These were applied by wire-wound rods on the metal substrates and were differentiated by the type of binder and content of fillers (silica particles as roughness modifiers) in their formulations. The coating thicknesses varied from 9 to 16 μm. Due to the silica roughness modifiers with particle diameters of around 5 μm, the thickness of the layer was higher for the rough primers. In agreement with the gloss values ranging from 5 to 84%, the root mean square roughness was about 0.6 μm for the scattering primer coatings Pr_1m and Pr_2m and 0.07 μm for the specular primers Pr_3s and Pr_4s (see Table 2).

The conductive inks were printed by flat-to-flat screen printing, in which both the printing plate and substrate are flat. The printing machine RokuPrint SD 05 was equipped with an adjustable table and a pneumatic squeegee mechanism (see Figure 1). The mesh size and the thread diameter of the polyethylene terephthalate- (PET-) based screen were 120 threads/cm and 34 μm, respectively. According to the data sheets of the ink suppliers, the selected inks are recommended for flat-to-flat screen printing with PET-based screens.

The printed structures were dried/cured in a forced air convection heating oven according to the recommendations of the ink’s suppliers. The conditions were 150°C for 15 minutes for I_p ink, 160°C for 30 minutes for I_g, and 150°C for 4 (I_m2 ink) and 5 minutes (I_m1 ink) for the silver inks. The printed structure was designed to measure the electrical resistance using the four-point probe method. The overall length of the structure was approximately 21 cm. The width varied from 0.4 to 3 mm.

To evaluate the compatibility of the inks and the primer-coated substrates, infrared spectroscopy and surface energy measurements were performed. The cured primers and inks were characterized by FTIR-ATR spectroscopy. The spectra were recorded in a wavenumber range from 4000 to 600 cm⁻¹ using a PerkinElmer 100 FTIR-ATR. Due to the high viscosity of the inks, the surface energy was measured for solidified primers and inks. The conductive inks were spin coated on circular silicon wafers using a POLOS Spin 200i device. The surface energy was examined with an optical contact angle-measuring OCA 20 device from DataPhysics. The contact angles of 3 test fluids (water, formamide, and glycerin) were measured. The surface energy values were calculated with the contact angle data as input parameters using the Wu method and the Owens-Wendt method implemented in the software package of the OCA 20 device.

To evaluate the thickness, width, ink distribution, and surface roughness of the printed structures, an Olympus LEXT OLS 4000 Laser Confocal Microscope was used. For surface roughness measurement, a 405 nm laser with a 50x objective was employed. The distribution of the ink was analysed using a LED white light with a 10x objective.

Regarding the main properties, the electrical resistance and adhesion performance were characterized. The resistance of the printed structures was examined with a Jandel RM3000+ four-point probe device. The voltage was measured for a given constant current. For the polymer and graphite inks, a current of 10 nA was used; for the silver inks, a current of 10 mA was used. The bulk resistivity and conductivity were deduced using the average thickness and width values obtained by laser confocal microscopy. The adhesion of the printed ink patterns was evaluated by peel-off testing using a Tesa 4104 tape. The tape was applied and peeled off manually. By visual inspection, ink residues were examined. If no residues were discernible, the adhesion was classified as a “pass”; otherwise, it was classified as a “fail.”

**Table 1: Designation and properties of the investigated inks.**

<table>
<thead>
<tr>
<th>Primer ID</th>
<th>Type</th>
<th>Type of solvent/ink</th>
<th>Conductive agent content (%)</th>
<th>Sheet resistance (Ohm/sq.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I_g</td>
<td>Graphite</td>
<td>Solvent/paste</td>
<td>16.5</td>
<td>Not provided</td>
</tr>
<tr>
<td>I_m1</td>
<td>Silver</td>
<td>Solvent/paste</td>
<td>68.5</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>I_m2</td>
<td>Silver</td>
<td>Solvent/paste</td>
<td>61</td>
<td>0.007</td>
</tr>
<tr>
<td>I_p</td>
<td>PEDOT : PSS</td>
<td>Water/gel</td>
<td>2.5</td>
<td>400</td>
</tr>
</tbody>
</table>

* Determined experimentally.

**Table 2: Designation and properties of the investigated inks.**

<table>
<thead>
<tr>
<th>Primer ID</th>
<th>Thickness of primer (μm)</th>
<th>Gloss 60°</th>
<th>Surf. roughness (Sq) (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pr_1m</td>
<td>15</td>
<td>6</td>
<td>0.59 ± 0.03</td>
</tr>
<tr>
<td>Pr_2m</td>
<td>16</td>
<td>5</td>
<td>0.67 ± 0.06</td>
</tr>
<tr>
<td>Pr_3s</td>
<td>16</td>
<td>80</td>
<td>0.07 ± 0.01</td>
</tr>
<tr>
<td>Pr_4s</td>
<td>9</td>
<td>84</td>
<td>0.07 ± 0.01</td>
</tr>
</tbody>
</table>

**Figure 1: Screen printing machine RokuPrint SD 05.**
3. Results and Discussion

3.1. Chemical Structure and Surface Energy of Inks and Primer-Coated Substrates. Characteristic IR bands identified in the primers’ and conductive inks’ transmission spectra are summarized in Table 3. Aliphatic (−CH2), carbonyl (−C=O), and ester (−C(=O)O−) bands were found in all the primers and inks. Except for the polymer-based ink, the primers and other inks exhibited characteristic bands related to polyester resins. The aliphatic H-C stretching vibration bands were distinguished at 2950 and 2850 cm−1. For the majority of the samples, a strong band attributed to a carbonyl in the ester groups was distinguished at 1720 cm−1. Medium intensity bands were obtained for aromatic ring stretching at 1600 cm−1 and out-of-plane bending at between 900 and 670 cm−1. Ester and ether group bands at 1260-1150 cm−1 and 1125-1020 cm−1, respectively, were found in all of the samples [15, 16].

The identified amine (−CN and −NH) and isocyanate (N=C=O) bands were the second characteristic of the spectra. At 2270-2250 cm−1, bands related to the asymmetric stretch of the isocyanate group were detected [17]. Furthermore, stretching carbonyl bands in a urethane group were found at 1690 cm−1. Also, peaks at 1550-1500 cm−1 were attributed to the bending of secondary amine groups [16, 18]. The primers Pr_2m and Pr_3s revealed weak peaks in the three mentioned wavenumber regions. For Pr_1m, the secondary amine peaks were not discernible. Pr_4s and I_g presented peaks just in the amino region. Both silver inks, I_m1 and I_m2, exhibited a weak isocyanate peak but differed in the secondary amine band, which was perceived just for I_m2. Presumably, polysiocyanates were used as the crosslinking agent for the polyester-based inks and residues of the crosslinking agent were found for the silver inks [19].

All primers and binders from the conductive inks, except I_p, were polyester and polyurethane based, maybe as a blended resin system or as polyurethanes. These binders are commonly used for coatings applied on metal substrates and for metallic conductive inks [17, 20]. Due to the chemical nature, a good compatibility between the primers and the solvent-based inks was expected.

For the polymer-based ink, the obtained spectrum did not present a good resolution. However, some bands in the hydroxyl group region at 3500-3300 cm−1 and in the aliphatic H-C stretching vibration range at 2900 cm−1 were identified. Some weak peaks were observed at 1380, 1270, 1120, and 1030 cm−1. These peaks were attributed to the aromatic and ether groups from PEDOT:PSS, and those at 1180 and 1020 cm−1 were attributed to the sulfonate group in PSS [21]. The chemical nature of the polymer-based ink differed significantly from the investigated primer systems. Hence, the compatibility may be critical.

In Figure 2, the surface energy and polarity (evaluated using the Wu method) of the primers and the conductive inks as solid layers are depicted. The surface energies of the primers ranged between 22 and 28 mN/m with low standard deviation, while the surface polarities varied between 33 and 43%. Both primers with a smoother surface, Pr_3s and Pr_4s, exhibited lower surface energy and polarity values. Compared to the IR spectra, this is in agreement with the weak polar group absorption peaks of these primers.

The surface energy of the inks was slightly higher for I_g, I_m1, and I_m2 and significantly higher for I_p compared to that of the primers. The I_g and both silver inks revealed a surface energy of about 30 mN/m. In contrast, the value for the water-based I_p ink was 46 mN/m. This is in agreement with findings in the literature [9]. In terms of surface polarity, significant differences with values between 19 and 69% were determined. I_p showed the highest value which can also be attributed to the water-based formulation. I_m1 was characterized by the lowest polarity percentage. Using the Owens-Wendt method, similar trends were obtained. In general, the surface energy and especially the surface polarity values were lower if evaluated by the Owens-Wendt method (except for the surface polarity of the water-based polymer ink I_p).

It was observed that all primers had lower surface energies than the inks, which could represent bad wettability [9]. By comparing the primers as well as the graphite and silver inks, a low variation of surface energy was exhibited with a coefficient of error below 10%. These materials also had surface polarities below 50%. Hence, a good chemical compatibility was expected. On the contrary, the compatibility of the I_p ink and the primers should be lower due to the higher significant differences in surface energy and polarity.

3.2. Dimensional and Topographical Features of Printed Structures. In Table 4, the morphological and surface properties of the printed structures are depicted and summarized. The light microscopic images of the lines indicate a pattern structure for the I_g and I_p inks. This was attributed to the screen grid of the printing process. I_p also showed limited wettability. The amount of voids and nonwetted areas were quantified between 9 and 25%. In contrast, a uniform and homogeneous print quality was obtained for the silver-based inks I_m1 and I_m2. The edges of the lines were jagged for the particle-based inks. The quality of the edge improved with increasing line width. The best edge definition was obtained for the I_p ink, which had few width variations.

Regarding the thickness of the lines, average values of about 4.5 μm were obtained for the I_g and I_m1 inks. The thickness variations were lower for I_m1, which did not reveal a pattern morphology. Interestingly, a lower thickness of 3 μm with a deviation of about 10% was determined for the I_m2 ink. The thickness of the I_p printed structures was detected to approximately 400 nm, with significant deviations. The low accuracy is also attributable to the measurement uncertainties of the laser confocal microscope. All the thickness values were independent of the structure’s width and the topography of the primer coating. This is in contrast to information in the literature where a significant impact of the substrate’s surface on the ink transfer and printability was described, specifically for substrates with roughness values up to the millimeter range. In this study, the average surface roughness of the investigated primer coatings was below 1 μm. These results were in agreement with previous data, where the applicability of conductive inks on surfaces with roughness values below 1 μm was evaluated [22].
Table 3: Identified FTIR bands of the commercial primers and conductive inks.

<table>
<thead>
<tr>
<th>Functional group (wavenumber)</th>
<th>Pr_1m</th>
<th>Pr_2m</th>
<th>Pr_3s</th>
<th>Pr_4s</th>
<th>L_g</th>
<th>L_m1</th>
<th>L_m2</th>
<th>L_p</th>
</tr>
</thead>
<tbody>
<tr>
<td>ν&lt;sup&gt;b&lt;/sup&gt; CH&lt;sub&gt;2&lt;/sub&gt; (3200-2700 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Weak</td>
<td>Weak</td>
<td>Med&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Med</td>
<td>Med</td>
<td>Med</td>
<td>Med</td>
<td>Weak</td>
</tr>
<tr>
<td>ν N=C=O (2275-2250 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Weak</td>
<td>Weak</td>
<td>Weak</td>
<td>—</td>
<td>—</td>
<td>Weak</td>
<td>Weak</td>
<td>—</td>
</tr>
<tr>
<td>ν C=O (1750-1700 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Strong</td>
<td>Strong</td>
<td>Strong</td>
<td>Strong</td>
<td>Strong</td>
<td>Med</td>
<td>Strong</td>
<td>Weak</td>
</tr>
<tr>
<td>ν C=O (urethane) (1690-1680 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Weak</td>
<td>Weak</td>
<td>Weak</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>ν Ph (1600 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Weak</td>
<td>Weak</td>
<td>Weak</td>
<td>Weak</td>
<td>Weak</td>
<td>Weak</td>
<td>Weak</td>
<td>—</td>
</tr>
<tr>
<td>ν CN and δ NH (1550-1500 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>—</td>
<td>Weak</td>
<td>Weak</td>
<td>Med</td>
<td>Med</td>
<td>—</td>
<td>Med</td>
<td>—</td>
</tr>
<tr>
<td>ν C(=O)O and δ&lt;sup&gt;c&lt;/sup&gt; =CH (1260-1150 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Strong</td>
<td>Strong</td>
<td>Strong</td>
<td>Strong</td>
<td>Weak</td>
<td>Weak</td>
<td>Med</td>
<td>—</td>
</tr>
<tr>
<td>ν O-C and δ=CH (1125-1020 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Weak</td>
<td>Weak</td>
<td>Med</td>
<td>Med</td>
<td>Weak</td>
<td>Strong</td>
<td>Strong</td>
<td>Weak</td>
</tr>
<tr>
<td>ν Si-O (1100-900 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Strong</td>
<td>Strong</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>γ&lt;sup&gt;d&lt;/sup&gt; Ph (900-670 cm&lt;sup&gt;-1&lt;/sup&gt;)</td>
<td>Strong</td>
<td>Med</td>
<td>Med</td>
<td>Med</td>
<td>Weak</td>
<td>Med</td>
<td>Weak</td>
<td>Weak</td>
</tr>
</tbody>
</table>

<sup>a</sup>Medium intensity. <sup>b</sup>Stretching. <sup>c</sup>In-plane deformation. <sup>d</sup>Out-of-plane deformation.

Figure 2: Surface energy (a) and surface polarity (b) of primer coatings and conductive inks.
The width accuracy for most of the printed lines showed that the actual value was below the nominal one. This deviation was highest for the I_g ink and the thinner line with a nominal width of 0.4 mm. The width accuracy of the silver-based inks was significantly better and in a comparable range. The best width accuracy was obtained for the I_p ink. The differences in the width accuracy were presumably related to the size of the conductive particles. While the accuracy was worst for the I_g ink with the largest particles, it was best for the I_p ink without additional particles.

The surface roughness of the printed structures as a function of the line width is displayed in Figure 3 for a rougher primer Pr_2m and a smoother primer Pr_4s. A similar trend was obtained for all evaluated primers. The roughness was highest for the I_g ink with the biggest conductive particles. For this ink, a significant dependency on the line width was ascertained with lower roughness values for wider lines. For both silver inks, the root mean square roughness was lower and differed significantly. For I_m2, the lower value is presumably attributable to smaller silver particles.

The roughness values of I_g and I_m1 printed structures were higher than the ones of the substrates. For I_m2, the roughness was in a comparable range for the rougher primers but higher for the smoother ones. For all these ink structures, the roughness values did not vary significantly with the type of primer. In contrast, the polymer ink I_p showed a significant smoothing effect in the rougher primer coatings. For this combination, the polymer ink was mainly localized in the valleys of the surface associated with a decrease in surface roughness. For smooth primer surfaces, I_p did not affect the roughness significantly.

### 3.3. Electrical and Mechanical Properties of Printed Structures

The electrical conductivity values of the printed structures on different substrates are depicted in Figure 4. The lowest values ranging from 50 to 240 S/m were obtained for the graphite ink I_g. The obtained values are also lower compared to literature data for graphite inks [1]. The silver inks revealed significantly higher conductivity values of about $10^6$ S/m which were in accordance with literature data [1, 2, 23]. The electrical conductivity of the I_p ink was around $10^3$ S/m, which was slightly below the nominal value given in the datasheet. This deviation was attributed to the poor wettability and nonuniform printed layer. The results for this ink were in agreement with previous results for

---

**Table 4: Print quality and surface properties of conductive inks on primer-coated surfaces.**

<table>
<thead>
<tr>
<th>Ink</th>
<th>I_g</th>
<th>I_m1</th>
<th>I_m2</th>
<th>I_p</th>
</tr>
</thead>
<tbody>
<tr>
<td>Printed ink structure</td>
<td><img src="image1.png" alt="Image" /></td>
<td><img src="image2.png" alt="Image" /></td>
<td><img src="image3.png" alt="Image" /></td>
<td><img src="image4.png" alt="Image" /></td>
</tr>
<tr>
<td>Thickness (μm)</td>
<td>4.5 ± 1.1</td>
<td>4.5 ± 0.3</td>
<td>3 ± 0.3</td>
<td>0.4 ± 0.2</td>
</tr>
<tr>
<td>Width accuracy error (%) (decreasing with width)</td>
<td>23 to 3%</td>
<td>11 to 2%</td>
<td>10 to 4%</td>
<td>6 to 1%</td>
</tr>
</tbody>
</table>

**Figure 3:** Surface roughness (Sq) of printed structures on a rough primer surface (a) and a smooth primer surface (b).
PEDOT: PSS ink formulations containing alcohols and/or surfactants [24, 25]. The obtained conductivity values were increasing with the line width. This was more perceived at the lines with widths of 0.4 and 0.7 mm. Such effects were also reported in literature and were related to the different surface/bulk ratio [13]. As to the effect of the primer, the electrical performance was dependent on the ink/primer combination. The lowest conductivity values for all printed inks were detected for the smoother primer Pr_3s. On the contrary, the highest values for I_g and I_m1 inks were also obtained in combination with a smooth primer Pr_4s. I_m2 achieved the highest conductivity on the rougher primer Pr_2m and I_p on Pr_1m.

To elucidate the potential effects of the substrates’ surface properties on the electrical conductivity, a one-way ANOVA was conducted for each conductive ink. It was deduced that there were statistically significant differences on the electrical conductivity performance of the inks I_g ($F(3, 4) = 31.1, p = 0.003$), I_m1 ($F(3, 4) = 7.6, p = 0.04$), I_m2 ($F(3, 4) = 26, p = 0.04$), and I_p ($F(3, 4) = 458.60, p = 1.6E-5$) at the $p < 0.05$ level. The LSD-Fisher post hoc test also revealed that for each ink, a different pair of primers did not have statistically significant differences. Therefore, no general tendencies were observed for all the inks, only ink-specific behaviours. Similar results were previously reported for silver-based inks, but for a polymer ink a correlation between the conductivity and the surface roughness of the printed structures was stated [7, 24]. However, in this research, the ink-primer combination showed a higher effect on the conductivity than the surface properties of the substrate and the printed structures.

The peel-off test revealed a good adhesion performance of the printed ink I_m1 and I_p on all substrates (see Table 5). No residues were detected on the tape. I_m2 passed the test in all primers except for the smooth primer Pr_4s. It
failed via delamination in the substrate-to-primer interface (see Figure 5). Most likely, the solvent of I_m2 was able to penetrate through the primer layer and weaken the interface by an environmental stress cracking mechanism [26]. However, this phenomenon was detected only for this primer/ink combination.

A poor adhesion performance was attained for the I_g ink on all primers investigated. As depicted in Figure 5, a significant amount of graphite had remained on both the tape and the primer. For this reason, this ink was characterized by a cohesive failure mechanism. Such failure types are characteristic for graphite-based ink formulations [1].

4. Conclusions

Four commercial conductive inks based on silver, graphite, and PEDOT:PSS were screen printed on different primer-coated substrates. A basic characterization of the primers, inks, and their printed structures was performed to determine their compatibility, printability, and functional properties. According to the identified IR bands, all primers and conductive inks, except the polymer-based ink, were based on polyester and polyurethane resins. Additionally, they exhibited low variations of surface energy values and a similar surface polarity percentage. Due to the similar chemical nature and surface properties, good compatibility was expected between graphite, silver inks, and primers. In contrast, the compatibility of the polymer-based ink was presumed to be lower because of the significant differences observed in its properties.

By laser confocal microscopy, the dimensional and topographical features of the printed structures were analysed. Both silver inks presented the best printability properties with uniform and continuously deposited ink layers. Graphite ink showed an average printability characterized by continuous lines but low uniformity and morphological properties. The worst performer was the polymer ink which exhibited nonwetted areas ranging from 9 to 25%. However, the polymer ink structures revealed the best width accuracy and edge definition. While the surface roughness of the graphite and the silver inks’ printed lines were independent from the substrates’ properties, a smoothing effect was ascertained for the polymer ink on rough primer surfaces.

The highest electrical conductivity was obtained by the silver inks’ printed lines with values of about $10^6$ S/m. The graphite ink structures presented the lowest conductivity with values ranging from 50 to 240 S/m. The electrical conductivity for the printed polymer ink was around $10^3$ S/m. This was attributed to the poor printability on the substrates. No significant correlations between electrical conductivity and the substrate’s surface properties were deduced. The peel-off test revealed a good adhesion performance of the printed polymer and silver inks. The silver ink I_m2 printed on the smooth primer Pr_4s failed the test via delamination in the substrate-to-primer interface. The graphite ink showed a cohesive failure mechanism with ink residues in both the tape and the primer.

Data Availability

The experimental data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

The authors kindly acknowledge the support of the Institute of Microelectronics and Microsensors at the Johannes Kepler University Linz for providing the devices for screen printing. The publication of this article was supported by the Johannes Kepler Open Access Publishing Fund.

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