Influence of Printing Parameters in Fully Pad-printed Electroluminescence Panels on Curved Surfaces

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Extended Abstract

Precision printing of particulate slurries with particle sizes of 5 to 20 µm using an indirect gravure printing, specifically with pad-printing technology, offers interesting application of functional printing on 3D-shaped surfaces. We present our results for printed electroluminescent panels (EL), where printing inks containing luminescent and dielectric particles of such comparatively huge size are transferred from a gravure printing form with gravure patterns not much larger than the particles. Compared with screen printing the benefit of pad printing is that fairly complex and corrugated surfaces of 3D-printed bodies can be endowed with a particulate functional surface. We demonstrated this using commercially available materials for electroluminescent panels. From the electrical point of view EL panels are capacitors with a luminescent and insulation layer which enable a careful investigation of the printed surface by the characterization of their electrical and optical performance. We emphasize that the printing process we developed is not restricted to EL panels but could also be used for printing protective, electrochemically active, or radiation absorbing metal or metal oxide coatings to 3D-printed fused-deposition-modeling polymer bodies. Printing parameters such as pad hardness, printing velocities (Bodenstein, 2018), pad surface tension are investigated. Further, we consider the relation between the printing process, the thickness of the insulation layer, the volume fraction of particulate $BaTiO_3$ in this layer, and EL panel luminance. Our pad-printed EL panels, using adapted ink formulations, achieved average luminance's of 140 cd/m^2 on curved surfaces which also corresponds to the performance standards of conventional screen-printed panels.

1. Introduction and background

In graphical printing pad-printing is frequently used to print items of limited size on curved surfaces, e.g. technical markings on medical products, advertisement materials, characters on computer keyboards or faces of toy puppets. As this can be achieved with particular high resolution is appears natural to use pad-printing in functional printing or printed electronics. In this way one could apply conductive patterns, electrical sensors, or lighting elements on curved, specifically on 3D printed surfaces, and open new applications here. However, the challenge is that many functional inks also involve particulate dispersion containing particle of considerable size of order of few 10 µm. This requires a partially new process concept. Specifically, in direct and indirect gravure printing the use of particulate dispersions is limited by the size of the gravure cells in the printing form. Gravure cells should be considerably larger than the largest particles in order to achieve reliable transfer of the dispersion to the substrate. On the other hand, using pad-printing forms with enhanced gravure cells may give rise to printing defects, loss of printing resolution, and insufficient control on the deposited material. We have therefore designed novel laser-engraved pad printing forms with optimized gravure pattern. Our specific goal was to understand the effect of separation of the particulate and the liquid phase of the printing dispersions in the pad printing process. Any change of particle concentration in the insulation and in the luminescent layer of an EL panel will immediately affect its electrical properties, i.e. its capacity and its optical performance. Together with measurements of the respective layer thicknesses this yields insight into the ink transfer mechanism, and on the degree of a possible phase separation cause in the pad printing process.

Electroluminescence lighting (EL) technology is based on light emission from a luminescent phosphor which is exposed to an alternating electric field. Figure 1 shows the schematic layer structure. The phosphor, usually a particulate layer of doped zinc sulfide particles, as well as a highly dielectric, insulation layer, are embedded between two electrode layers one of which is transparent. The electrodes are connected to an AC power supply. It is possible to reproduce large quantity of customized EL panels where each of the layers can be patterned according to the specific application. Nowadays, EL panels are printed with screen printing technology by default. However, screen printing is limited to its variety of substrates (flat or rotative symmetric shape of objects). Pad-printing technology offers high flexibility and endless variety of surfaces and shapes to print on.

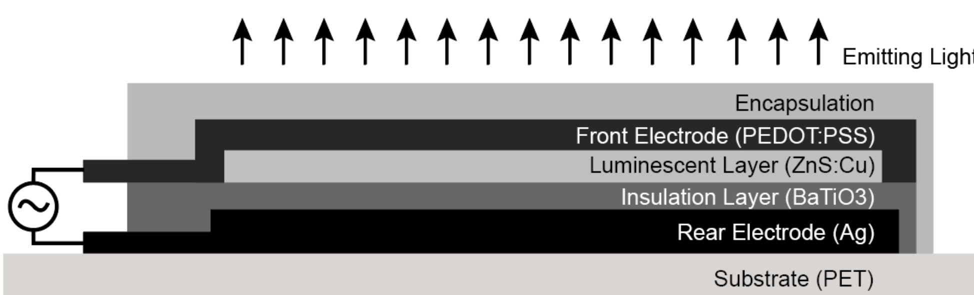

Figure 1: Schematic Layer structure of a printed electroluminescent device (permission from Hirmer, K.)

Contrary to screen printing, where thick layers of 10 µm and higher can easily be reached and particulate dispersions are easy to handle pad-printing usually creates layers of 2-3 µm in thickness. This is mainly the consequence of the much lower ink viscosities. The challenge in EL printing is, however, to provide a stack of dense layers of hard ZnS and BaTiO3 particles, respectively, as the particles, not the soft carrier phase in the interspaces, are responsible for the desired electrical features such as electrical insulation between the electrode layers, high dielectric polarizability, and continuous light emission.

As described above, the layer structure of EL technology is basically a plate capacitor where the capacity is defined by

$$C = \varepsilon_0 \cdot \left(\frac{d_1}{\varepsilon_{r1} A} + \frac{d_2}{\varepsilon_{r2} A}\right)^{-1} \quad [1]$$

where ε_0 is the vacuum permittivity, $\varepsilon_{r,1}$ and $\varepsilon_{r,2}$ the material permittivities of insulation and luminescent layers. *A* is the area size and *d_1, d_2 are* the thicknesses of the two layers. Note that the permittivities significantly of a particulate printing ink significantly depends on the particle versus binder content of the formulation as transferred to the substrate. We were able to demonstrate that this is not always identical with the particle content of the raw formulation but changes with the amount of ink that is transferred to the respective layer by the pad. This could be explained by an effect the different adhesions of particulate and liquid binder phases the printing form, and to the pad, giving rise to a partial segregation of the particulate phase. In order to study this effect, we created sets of EL panels which had undergone different numbers of 1 to 6 subsequent pad-printing steps of the insulating printing suspension. We then measured the average thickness of the layer, its electrical capacity per area, and also the electroluminescent light emission. In view of eq. (1) this yields the effective permittivity of the layer as a function of the process steps, and allows for some conclusions on the effect of the printing process on particle segregation and transfer rate. Changing *d_1 or d_2* leads to a change of the capacity. With eq. (1) we can draw conclusions on the permittivities of the respective layers of the panels, and on the dependence of the particle content on the processing scheme. The aim of this study is first to realize a fully pad-printed EL-panel and second to understand the influence of printing parameters to electrical and optical properties. The specific focus is on the role of the printed insulation layer.

2. Materials and Methods

Pad-printing

In pad-printing, which is an indirect gravure printing technology, the printing pattern is engraved as a raster of microscopic cells on a planar printing form. The size of these cells, usually a few tens of microns in width and depth, determines the amount of ink transferred to the surface of the substrate. In the pad-printing process the engraved pattern is first flooded with ink. After the engraved cells are filled with ink and excess material is removed from the walls by blading, a soft silicone pad is pressed against the printing form. When the pad is lifted from the printing form, it takes the ink out of the cells and subsequently deposits it on the surface of the substrate.

The pad-printing machine used in the experiments is described by Hakimi (2016). For each layer a separate printing form have to be created. As shown in Figure 1 in the schematic layer structure, the first printed layer is the rear electrode with conductive material followed by the insulation and luminescent layer. To complete the functionality of the plate capacitor another conductive layer must be printed on top. In the case of electroluminescence this electrode must be conductive and transparent at the same time, so that the emitting EL-light is not shaded. An encapsulation is printed and is isolating the electrical component.

Since a very low variety of functional inks for pad-printing are available the ink for the insulation and luminescent layer were self-formulated by blending the functional particles with a common pad-printing solvent based transparent ink. The EL multilayer is processed with polymer printing forms with a depth of 30 microns, a resolution of 100 L/cm. The raster has an area coverage of 86 % respectively. The samples were processed under clean room conditions with a humidity 50 % and temperature of 23.5 °C.

We first evaluated printability and then functionality of pad-printed EL with respect to each of these printing parameters on a flat PET substrate in order to optimize the printing conditions to print in a next step onto a curved surface.

For measuring the layer thicknesses of the different amounts of insulation layers (1-6) the layers were printed separately onto a PET substrate with the same printing conditions. The measuring system is a tactile profilometer DektatXT® with a pin radius of 2.5 µm and a stylus force of 3 mg.

For characterization the pad-printed EL samples were connected to an 'EL sheet inverter LF3012 12 VDC 250 mA 320 m²'. The luminance in cd/m² was measured on each sample with a Gossen Mavo-Monitor.

3. Results and Discussion

It turned out that the printed EL-samples with only one layer of insulation between the electrode layers was insufficient, and short circuits were detected. This can be explained by the porosity of the insulating layer, and by the imbibition of the front electrode printing fluid into the remaining pores. However, panels with 2 to 6 printed insultaion layers did not show this effect and luminescent properties of the panels could be determined. Figure 2 shows the measured average layer thicknesses of the insulation layers created by 1 to 6 consecutive pad-printing runs. With increasing number of printing runs layer thickness was increasing linearly. If the dielectric constant would be unaffected by this procedure the capacity as well as the luminance of the panel should drop, as implied by eq. [1]. The influence of printed insulation layers to the luminance is shown in Figure 3. The efficiency by regarding the luminance is from 40-50 cd/ m². Contrary to this we did not observe any significant influence of the increased insulation layer thickness on the luminance, also regarding the statistical deviation of our measurements.

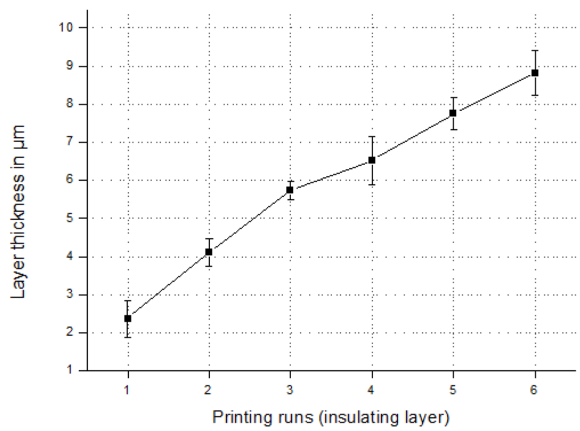

Figure 2: Measured layer thicknesses by increasing number of printing runs

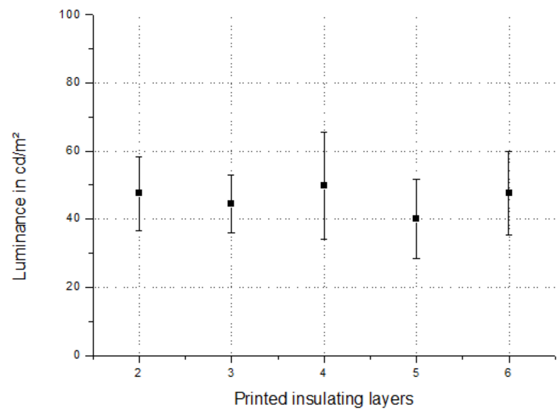

Figure 3: Influence of number of printed insulating layers to luminance

After characterizing the pad-printed EL-devices on flat PET substrate the whole process was applied to printing EL panels on a polycarbonate cup with curved and kinked surface features (as shown in Figure 4). Based on the results in this study, namely that a variation of the insulating layer thickness has no significant influence, the printing runs for the insulating layer were set to 4 times. When printing on curved PC cups the insulation layer was printed with a ratio 3:2 instead of 2:3 of $BaTiO_3$ particles and pad-printing ink. All other printing parameters were retained. With this variation of ink formulation, the efficiency was increased to an average luminance of 139 cd/m^2 (± 39.5) which is a threefold of the former results. By increasing the content of $BaTiO_3$ particles in the ink formulation, also the material permittivity ε_r increases which leads to changes in the capacity and thus to higher luminance.

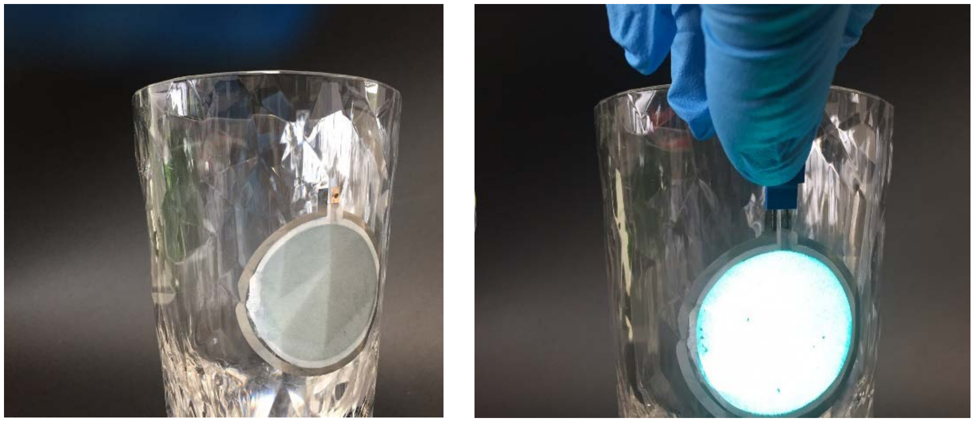

Figure 4: Pad-printed electroluminescence on an uneven and convex cup in off and on condition

4. Conclusions

Our conclusion is that the printing process as used in our study had significant tendency to induce a phase separation in particulate printing dispersions. The particle content in the deposited layer is increased with respect to the material in the ink reservoir. Even more important is that the printing pad appears makes a bidirectional transport of liquid and particulate ink components between reservoir and substrate. Our finding is, by repeating the transfer process several times in sequence, and under the condition that ink deposited on the substrate remains in the wet state, that the printing pad starts to return the liquid carrier phase back from the substrate to the printing form. On the other hand, the dispersed phase, $BaTiO_3$ particles, are more or less completely deposited, and are thus concentrated on the substrate with each repetition of the transfer step. This explains why luminance and capacity of the EL panels remains on the same high level in spite of the continuous increase of the optically inactive dielectric interlayer. If this was not the case, we would have expected that the luminance at a given operation voltage would decrease, but this was not observed in any of our samples.

Contrary to the expectations there was no significant influence of different insulation layer thicknesses to the luminance and reached 40-50 cd/m² on flat PET substrate. The ratio of insulation $BaTiO_3$ material to pad-printing ink was then changed from 2:3 to 3:2 and the EL was printed onto a curved PC cup. With this change in ink formulation the luminance was increased to 140 cd/m².

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