Inkjet Printing as a Roll-to-Roll Compatible Technology for the Production of Large Area Electronic Devices on a Pre-Industrial Scale

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Abstract

Inkjet printing is a promising approach towards the solution processing of electronic devices on an industrial scale. Of particular interest is the production of high-end applications such as large area OLEDs on flexible substrates. Roll-to-roll (R2R) processing technologies involving inkjet printing have especially high potential, since they allow a continuous production of large volumes with high throughput.

Here we report on our research activities to scale up printed OLED production on foils to an industrial scale. When building up the device, necessarily deposition will have to be done on very different types of surfaces, some of which are highly prone to damage upon mechanical load. Inkjet printing as a non-impact technology is therefore expected to have decisive advantages compared to e. g. screen printing. We have evaluated both methods for silver shunt line deposition on a barrier layer which protects the OLED against humidity. Screen printing on the barrier layer resulted in a significantly higher number of defects than did inkjet printing. For post-deposition treatment we have used photonic flash sintering as a highly efficient and R2R compatible method.

We were thus able to demonstrate that a core step of OLED production can be carried out by R2R processing. Finally, our efforts resulted in the production of fully functional large area OLED devices, with inkjet printed silver shunt lines. Our future plans include moving towards fully integrated R2R production of OLEDs to demonstrate this concept's feasibility for industrial scale manufacturing.

Introduction

The integration of electronic functionalities like light emission, photovoltaic power generation, communication into everyday consumer products has highly promising economic prospects [1]. In order for this vision to become commercially viable on a wide scale, fast and inexpensive mass production technologies for electronic devices must be available. Printing and other solution processing methods are highly attractive in this respect, as they can easily be up-scaled to large manufacturing volumes and operate under ambient conditions [2-3]. A crucial component of any electronic device are highly conductive structures (Fig. 1), and those can be prepared by the printing of metal based inks [4-5]. In addition to its many other advantages, inkjet printing as a non-contact deposition technique is

especially attractive for substrates which are highly sensitive to damage by mechanical load, such as the barrier foils necessary for flexible OLED and OPV devices [6]. Mass production using inkjet printing is a well-established technology in the graphics industry and is currently also starting to be applied for electrode manufacturing in the area of printed electronics [7,8]. When used for conductive inks, it is especially attractive in combination with a fast and efficient post-deposition treatment, like photonic flash sintering [8,9]. Both technologies have been demonstrated to be roll-to-roll compatible and are thus highly promising candidates for development towards mass production.

In this contribution, we report on the processing of silver based conductive inks by inkjet printing and photonic sintering. We demonstrate that functional foils coated with fragile barrier layers can be processed using these techniques on a roll-to-roll pre-industrial production line, and that they can be successfully applied in functional OLED devices.

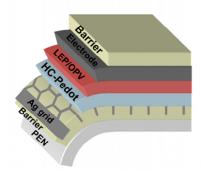


Figure 1. Schematic representation of a typical OLED device architecture.

Experimental Part

Sheet-to-sheet printing was carried out using a commercial materials inkjet printer (DMP from Fujifilm-Dimatix) with disposable cartridges (DMC-11610; droplet size approx. 10 pl). The print head contains 16 parallel squared nozzles with a diameter of 30 μm . The ink was printed at a voltage of 25 V, using a frequency of 10 kHz and a customized waveform. The printing height was set to 1 mm, while using a dot spacing of 20 μm . Roll-to-roll inkjet printing was carried out with a customised industrial inkjet module with a Xaar 1001 recirculating print head. All inkjet experiments were carried out with commercially available silver nanoparticle dispersions.

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Screen printing was done using a DEK Horizon 03i screen printer equipped with a vacuum substrate holder which can handle substrates up to 30 x 30 cm². For the experiments two types of commercial screen meshes were used (calendared to 17 μ m thickness with a 5 μ m thick emulsion and electroformed nickel 355 meshes with emulsion thicknesses between 6 and 20 microns). The used squeegees had a shore hardness of 70 with an angle of attack of 45°. The material used was a commercial conductor paste based on a highly concentrated dispersion of micron sized silver flakes in an organic solvent mixture.

Photonic sintering was carried out in a home-made roll-to-roll compatible apparatus, the details of which are disclosed in [8] and [9]. Shortly, it consists of a near-IR drying unit (Adphos NIE 120-M3) and a set of four Xenon stroboscope lamps (XOP-15, Philips, The Netherlands) with elliptic reflectors. The flashing parameters like intensity, frequency and the pulse sequence were controlled by a software programme written in Labview. The reflectors projects an image of the lamp on the second focus line, through which the foil is moving, resulting in a bell-shaped intensity distribution with a full width at half maximum of ca. 12 mm. The rather broad intensity distribution of the focused light ensures a homogenous illumination of the ink line over its entire width, as long as the test line is significantly narrower than the FWHM. The lamps have a maximum electrical power consumption of 1000 W, an emission spectrum ranging from 350 to 900 nm, a flash frequency of up to 17 Hz, and a pulse duration which varies between 3 and 8 ms, depending on the used intensity.

A typical bottom emissive OLED stack consists of a hole injection layer and a light emitting polymer sandwiched between a transparent anode and a full-area non-transparent cathode of evaporated aluminium. As the electrical conductivity of the transparent electrode material (Pedot:PSS) is not sufficient, an additional high conductive metal grid is required, which is inkjet printed on a barrier coated substrate. Due to the orientation of the electrical field in the OLED stack the silver of the metal grid tends to migrate over time resulting in a defective OLED device. To prevent this, an additional isolation layer is printed on top of the silver anode grid. This isolation layer is typically 2 microns thick and in the current experiments around 200 μm wide to make sure all silver is fully covered.

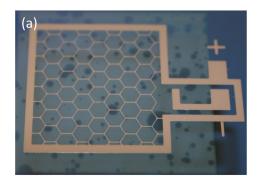
Results and Discussion

Screen vs. Inkjet Printing

Shunt line structures for transparent electrodes have been deposited on substrates coated with a barrier layer, which consists of a multiple stack of organic and inorganic layers. The purpose of this barrier layer is to retard the diffusion of oxygen and moisture from the environment in to the device, and therefore prevent degradation of the functional layers and impoverishment of the device performance. The inorganic material is highly brittle and sensitive to mechanical forces. The location of pinholes can be conveniently monitored by using it to seal a calcium layer, which is destroyed by contact with water, whereupon clearly visible holes appear after storage in a moist environment. When a silver paste was screen printed on such a test device, defects were clearly visible after a few days (Fig. 2a), whereas no such damage was observed for inkjet printed samples. This indicates that the strong

mechanical forces exerted on the surface during screen printing break the inorganic layers, forming pinholes through which water can penetrate into the device architecture. As a non-contact deposition technique, inkjet printing is much more gentle and does not cause any damage.

Another difference between the two printing techniques was found when the profiles of silver lines where measured: Screen printed lines were not only significantly higher, but also showed a much rougher surface topology and a lot of spikes (Fig. 2b). This is expected to negatively affect the performance of such structures in electronic devices, as it increases the risk of local short circuiting through the functional layers. By contrast, inkjet printing of silver inks resulted in very smooth surface profiles, much more suited for device integration (Fig. 2b).



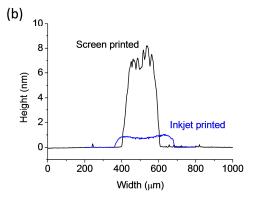


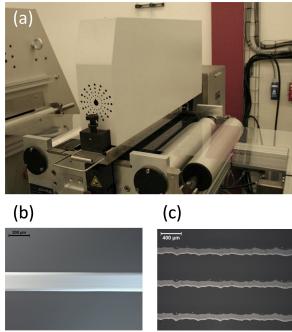
Figure 2. Screen printed silver grid on a barrier-coated calcium layer, demonstrating pinhole formation by local degradation of the calcium (a). Typical surface profiles of a screen and inkjet printed silver line (b).

R2R Processing

In order to scale up the production of shunt line patterns for OLEDs to an industrial scale, we have designed and built a roll-to-roll manufacturing line, which a. o. comprises an inkjet tool (Fig. 3a). At current state, it contains a single printhead with 1000 nozzles, but work us being done to integrate more heads, so that higher printing speeds and web widths can be achieved. Optimisation of the printing conditions was carried out for a number of relevant substrates (coated and uncoated PET, PEN) by variation of drop ejection waveform and timing, resolution, and drop volume [8]. It was found that printing lines parallel to the

substrate movement resulted in much better defined structures than printing in the orthogonal direction (Fig. 3b,c)

Figure 3. Industrial inkjet printing module for conductive inks, integrated in a roll-to-roll production line (a). Inkjet printed silver structures on plastic foil,



printed on the roll-to-roll line in the substrate movement direction (b) and orthogonal to it (c).

In order to render the printed silver ink deposits on the plastic foils conductive, a post-printing treatment for drying and sintering is necessary. For a high overall production speed to be feasible, the conventional approach of thermal treatment by hot air ovens, which is a very slow process, was replaced by the recently developed method of photonic flash sintering [9]. This technique is based on the selective and localised heating of the silver inks by the absorption of intense, short pulses of visible light, for which the substrate is transparent. It is able to reduce the processing times from several minutes to fractions of a second without causing foil damage and is therefore compatible with high speed roll-to-roll manufacturing. The design and construction of a photonic sintering tool (Fig. 4a) comprising a near-IR drying unit and several Xenon flash lamps and its integration into a roll-to-roll line has been reported earlier [8]. Process optimisation was carried out by varying a number of parameters, such as the flashing intensity and frequency. A combined approach of near-IR drying and Xe flash sintering yielded the best results (Fig. 4b), allowing processing speeds of 10 m/min and conductivities up to 12 % of the bulk silver value [8].



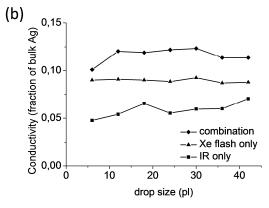


Figure 4. Flash sintering module for conductive inks on plastic foils, integrated in a roll-to-roll production line (a). Optimisation of the process condition for the photonic sintering.

OLED Fabrication

The compatibility of inkjet printed silver shunting line structures with plastic electronic devices was demonstrated by integrating them into an OLED architecture. Transparent electrodes were prepared by depositing a layer of PEDOT on top of the silver grids. Subsequently, the device was finished with a spin coated green electroluminescent emitter, a counter electrode and an incapsulation (Fig. 1). For comparison, also a set of devices were produced with identical architecture, except that there was no grid of shunting lines. Although both types of devices worked, the light output from the OLEDs with shunting lines was significantly more homogeneously distributed over the entire luminescent surface (Fig. 5). This is a result of the reduced resistive losses upon current transport through the transparent electrode. The devices prepared for this study had a surface area of 8 by 8 cm, but the beneficial effect of shunting lines is expected to become even more important for larger devices.

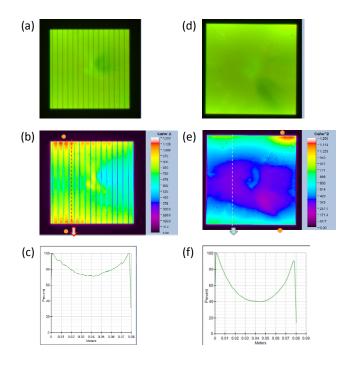


Figure 5. Photographs and light output homogeneity of OLEDs with (a-c) and without (d-f) inkjet printed silver shunting lines.

Conclusions

We have demonstrated that inkjet printing of silver nanoparticle inks is a viable method for the large scale production of plastic electronic devices such as OLEDs. As a non-contact technique and in contrast to e. g screen printing, it is compatible with mechanically highly sensitive surfaces, such as OLED barrier layers. Furthermore, in combination with a fast and efficient drying and sintering method like photonic flash sintering, it allows high process speeds and can be applied on a roll-to-roll line. Process speeds of up to 10 m/min are possible without the need for enormously long hot air ovens. Therefore, this approach is highly

promising for the manufacturing of printed electronic devices on an industrial scale.

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