

Screen-printed passive matrix displays based on light-emitting polymers

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Due to their outstanding properties, e.g., good contrast, wide viewing angle, low power consumption, and self-emission organic light-emitting (OLE) displays on the basis of conjugated polymers are on the verge of commercialization. Two major disadvantages of the current processing technique for the polymers—spin coating—are the material waste and the difficulties involved in patterning multichrome or even full-color displays. Therefore, we investigated the screen-printing technique for the production of OLE displays. In this letter, we present performance data and images of screen-printed OLE diodes. They are already comparable to spin-coated ones. We observed luminance of 10 000 cd/m² at 8 V and peak efficiencies exceeding 10 cd/A for green diodes. These data indicate that printed organic displays have the potential to replace “classical” spin-coated devices. © 2001 American Institute of Physics. [DOI: 10.1063/1.1379594]

The graphical presentation of information becomes more and more important in our everyday life. A significant role as an interface between man and machine play low information content displays that are used, e.g., in mobile phones or car radios.

Certain display properties are of particular interest: good contrast and brightness, wide viewing angle, availability of different colors, small depth, low power consumption, and long lifetime. All these conditions—apart from the last—are rather well fulfilled by organic light emitting (OLE) displays on the basis of conjugated polymers like poly-(2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV) or polyfluorenes (PF).¹

The classical coating technique for polymers is spin coating of polymer solutions, as it allows fabrication of very thin and homogeneous films. Nevertheless this technique also features a number of disadvantages: Since only a very small amount of polymer forms the actual film, more than 99% of the polymer solution is wasted. Furthermore the substrate size is limited to approximately 40×40 cm². Lateral patterning of different polymers—and thus various colors—is only possible in a costly process.² A further drawback of spin coating for mass production of displays is the necessity of removing the polymer film from the electrical contact pads. Typically, this removal is performed by laser ablation, a complex, time-consuming, and expensive process.

Therefore we decided to investigate a printing technique for the coating of the polymers in the production of passive matrix displays. Among many others we chose screen printing as it offers some appropriate features: First, it is a well known, inexpensive, and highly parallel technique. Second,

it allows high printing speed on large substrates up to m² range, with good resolution below 100 μm. Besides, it is suitable for glass plates as well as flexible substrates. Recently, results that prove the feasibility of screen printing technique in the fabrication of OLE diodes were published by Pardo *et al.*³ They succeeded in producing OLE diodes with a screen printed hole transport layer and a thermally evaporated emitting layer.

For all our experiments we used a commercially available semi-automatic screen printer with an optical positioning system. Our screens featured a size of 350×350 mm² and consisted of polyester fabric with 165 meshes per cm. They were direct-coated with poly+HV—a water proof, photosensitive emulsion—and structured by means of photolithography. An important parameter in screen printing is the so-called snap-off distance, the gap between the screen and the substrate. We chose a rather small snap-off distance of 0.5 mm.

At first we printed test diodes and compared performance data—efficiency, luminance, rectification ratio, and onset voltage—with spin coated diodes. Our test layout allowed us to produce 8 diodes on one 50 mm square substrate. We used glass substrates that are coated with ~100 nm indium tin oxide (ITO). The roughness of the sputtered ITO was in the range of 2.2 nm and the sheet resistance was below 20 Ω/□. The first process step was an UV/ozone treatment. Immediately afterwards we printed the first polymer layer—a barrier layer that prevents indium diffusion from ITO into the emitting polymer. For this purpose we used a commercially available poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) aqueous dispersion (PEDOT) from Bayer AG, Leverkusen.

After having printed the first organic layer, we baked the substrate for drying purposes in the oven. Then we printed

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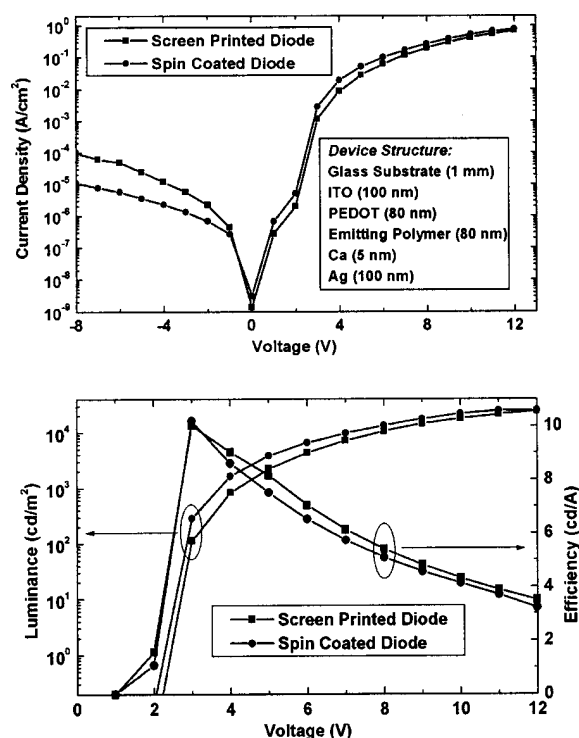


FIG. 1. Comparison of a screen printed and a spin coated OLE diode with the same device structure (coating techniques refer to the two organic layers only). Efficiencies as well as luminances and onset voltages are comparable. Rectification ratio of the screen printed device is approximately one order of magnitude lower. The diodes emit green light.

the emitting polymer, a Polyfluorene derivative (PF) that emits in the red or green region. Molecular structures and electroluminescence (EL) spectra are reported in Ref. 4. As solvent benzene derivatives, such as xylene and toluene, were used. This second organic layer was dried first under ambient conditions for 1 min and then in the oven at 80 °C for 5 min. We are able to adjust the polymer thickness between 70 and 100 nm by varying the concentration of the polymer solution. All the steps described so far were carried out in a clean room. In a last step we thermally evaporated a Ca (5 nm)/Ag (100 nm) cathode. The evaporator as well as our test equipment for the luminance and the I - V measurement were situated in a glove box filled with argon. For a detailed description of the process conditions, see Ref. 5.

By visual inspection, the luminance of our test diodes were absolutely homogeneous. Unlike reported elsewhere³ no pattern caused by the screen fabric was visible. In order to achieve perfect homogeneity, the drying conditions after the screen printing of the polymers play an important role. A fast drying in the oven causes a “cloudy pattern,” whereas slow drying generates coffee stain like rings on the diodes. Therefore, we used a combination of drying under ambient conditions and baking in the oven. In addition to the appropriate thickness, the lateral resolution is of interest if one intends to print adjacent pads of different polymers, e.g., for multicolor displays. While the thickness homogeneity is adequate, the width of the edge zone of our printed areas is still pretty large: depending on the polymer 0.5–1.0 mm.

Figure 1 shows performance data of a typical screen printed test diode that emits in the green range, and a spin coated reference diode. Both diodes had the same structure,

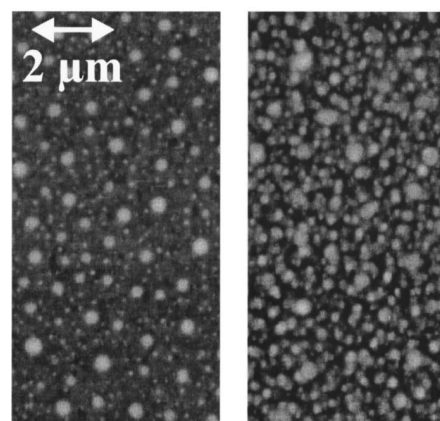


FIG. 2. AFM images of a spin coated (left) and a screen printed (right) emitting polymer (PF). The morphology is similar. Roughness of the spin coated film is 3.6 nm, of the screen printed film 6.2 nm.

but were structured by different techniques. The differences in luminance, onset voltage, and efficiency are in the range of uncertainty of our diodes. Remarkably, the backward current of the printed devices is about one order of magnitude higher. This is probably caused by particles accumulating on the substrate during printing and thus creating local shorts in the diode.

Peak efficiencies of more than 10 cd/A were observed. Onset voltage is less than 3 V, and luminance of 10 000 cd/m² was measured at 8 V. The rectification ratio—an important number if we consider the fabrication of passive matrix displays—was higher than 10³ at ± 5 V. These data clearly demonstrate that printed OLE diodes can be an alternative to conventional spin coated devices, which is a big step forward in technology.⁶

With an atomic force microscope (AFM) we investigated the morphology of the polymer film surface. We found that the roughness of the surface of spin coated emitting polymer films is a factor of nearly 2 lower than the roughness of screen printed films (3–4 nm vs 6–8 nm rms), whereas the morphology is similar (see Fig. 2). However, in our opinion this difference is not the major reason for the difference in rectification ratio for both kinds of diodes, because we observed that small area screen printed diodes tend to have higher rectification ratios. This indicates that not the morphology of the film, but rather defects cause the higher backward current of the printed diodes.

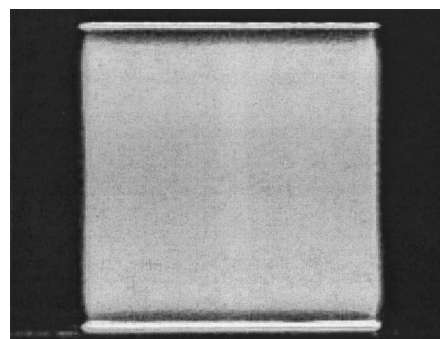


FIG. 3. Close up view of a screen printed electroluminescent pixel. There are no inhomogeneities caused by the screen. Close to the photo resist structures on the top and bottom side the polymer film is slightly thicker, therefore the luminance is less bright. Pixel size is 270 μ m $\times$ 270 μ m.

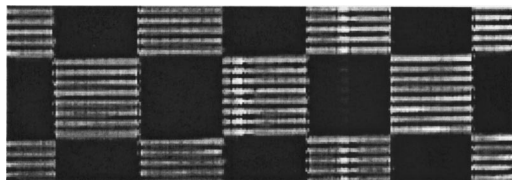


FIG. 4. Detail (33.5 mm $\times$ 12 mm) of a screen printed electroluminescent display. The size of the display is 43 mm $\times$ 26 mm. It shows a checkerboard test pattern. Each square consists of 8 $\times$ 8 pixels, pixel size is 640 μ m $\times$ 640 μ m, spacing is 30 μ m.

In order to produce a screen printed passive matrix OLE display we printed our polymers on an ITO-glass-substrate with pre-patterned photo resist structures. These structures have a small undercut and act as cathode separators.⁷ During the printing process they had to sustain the pressure of the squeegee. Therefore we had to strengthen our photo resist structures by modifying the photo lithography process steps. Further, we printed parallel to these structures and chose a small squeegee force, only 20 N, and a soft rubber squeegee to avoid damage to the photo resist. Figure 3 shows a close up view of a screen printed pixel. The electroluminescence is very homogeneous, no features of the screen are to be seen. However, scanning electron microscope (SEM) investigations prove that the polymers tend to assemble under the undercut of the photo resist structures. This implicates two negative effects: First, close to photo resist structures the polymer film is about twice as thick as in the center of the pixel. Therefore the brightness at the outer 15 μ m is reduced. Besides, the polymer partly fills the undercut whereby the cathode separation deteriorates.

A picture of a screen printed display is shown in Fig. 4. The checker board test pattern is easy to recognize, although

there is still some cross talk between the pixels. Every square consists of 8 $\times$ 8 pixels. The two brighter columns are due to shorted pixels in this row, so still some improvements have to be done.

In conclusion, we have fabricated high-efficiency organic light emitting diodes on basis of conjugated polymers, in which both polymer layers were screen printed. The performance data, as far as efficiency, luminance and onset voltage are considered, are the same as that of spin coated devices. Rectification ratio of printed devices is about one order of magnitude lower. The homogeneity of the printed layers is sufficient, whereas the lateral resolution is still improvable. We also demonstrated that it is feasible to produce screen printed passive matrix OLE displays.

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