Highly performant organic top-emitting light-emitting diodes (OLEDs) by solution process

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Abstract: This works aims to develop an inverted top-emitting solution-processed OLED. The early results obtained are promising to achieve similar efficiencies than bottom-emitting solution-processed OLED structures.

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Organic Light-emitting diodes (OLEDs) research has been focusing on the simplification of the fabrication process to reduce the cost. Printing technologies to make thin films of conjugated polymers or small molecules have been shown to be attractive for that purpose. For a few years, research on inverted bottom-emitting OLEDs (BOLEDs) fabricated by solution-process has been widely conducted. Inverted BOLEDs show similar or even higher performance than conventional structures [1–3]. The main problem of BOLEDs is the emission through the glass substrate: the refractive index of glass is around 1.5, whereas the refractive index of organic layers is around 1.8. This difference results in a typical light extraction of only 25 % [4,5]. On the other hand, top-emitting OLEDs (TOLEDs) show interesting properties for displays and lighting. They are more suitable for active-matrix OLED displays, using a thin-film transistor backplane. The TOLED structure allows the design of a large aperture for high-resolution displays without any blocking from the wired metal line and the opaque TFT [6,7]. Besides, inverted TOLEDs are highly promising in the case of a CMOS addressing circuit because they are easier to use than direct TOLEDs via a common anode addressing scheme. Nevertheless, research effort mostly focuses on evaporated inverted TOLEDs and only a few studies have been conducted to develop TOLEDs by solution process, whether direct or inverted structures [8]. One of the major issues of TOLEDs is the optical effects of the microcavity: to reduce them, the organic layer thickness has to be controlled, which is more difficult to realize by solution process than by thermal evaporation. The optical cavity can be optimized at different orders. For order k=0, the total organic layer thickness has to be below 100 nm[9]. Additionally, each layer has to be thin (< 20 nm), which is difficult to achieve by solution process. Here, we present a study of TOLEDs with order k=1 for varying organic-layer stack thickness of 200-300 nm. The performance of the TOLEDs is compared to BOLED performance.

BOLEDs and TOLEDs based on the same emissive material (Super Yellow, PDY-132 Merck) were fabricated. Both architectures are shown in Fig. 1. The whole fabrication process takes place in a clean room environment. The BOLED structure has been studied in a previous work [3,10]. OLEDs are built on 25 by 25 mm^2 glass substrates, covered by ITO for BOLEDs and covered by an evaporated Al/Ag bilayer for TOLEDs. Qian et al. showed for a direct TOLED that this bilayer increases the reflectance compared to an Al monolayer, leading to a higher current efficiency (+15 %) [6]. Besides, depositing Ag on Al can avoid short circuits in OLEDs: Deposition of Ag directly on glass forms a rough layer because of the poor infiltration between pure Ag and glass. The same bilayer is used in this work for inverted OLEDs, even if the work function of Ag is higher than the work function of Al. Indeed, the PEIE layer deposited on top of the cathode can effectively reduce the cathode work function to keep an efficient electron injection in the LUMO level of Super Yellow (-3.0 eV). Zhou et al. measured WF_{Ag}= -4.60 eV and WF_{ITO} = -4.62 eV by Kelvin probe [11]. After spin-coating PEIE, the new work function for both materials are -3.70 and -3.60 eV, leading to a similar electron injection. For TOLEDs and BOLEDs, the same first layers were spin-coated for the same thickness: ZnO, PEIE:TPBi and Super Yellow. Then an NPB layer was evaporated at different thicknesses in the TOLED structure to adjust the optical cavity and increase the light extraction from the device.

Figure 1 shows that the turn-on voltage is lower for TOLEDs, around 2.2 V, showing that the charge injection into the device is efficient with Al/Ag cathode. However, lower current densities are shown for TOLEDs, more particularly for TOLEDs with NPB, due to the high thickness of this layer. For 20 mA/cm², the BOLED luminance is around 1220 cd/m² whereas the TOLED luminance is 375 cd/m² and the NPB-TOLED is 930 cd/m² (Table 1). Adding NPB in the TOLED structure efficiently increases the luminance and therefore the current and luminous efficiencies compared to TOLED without NPB.

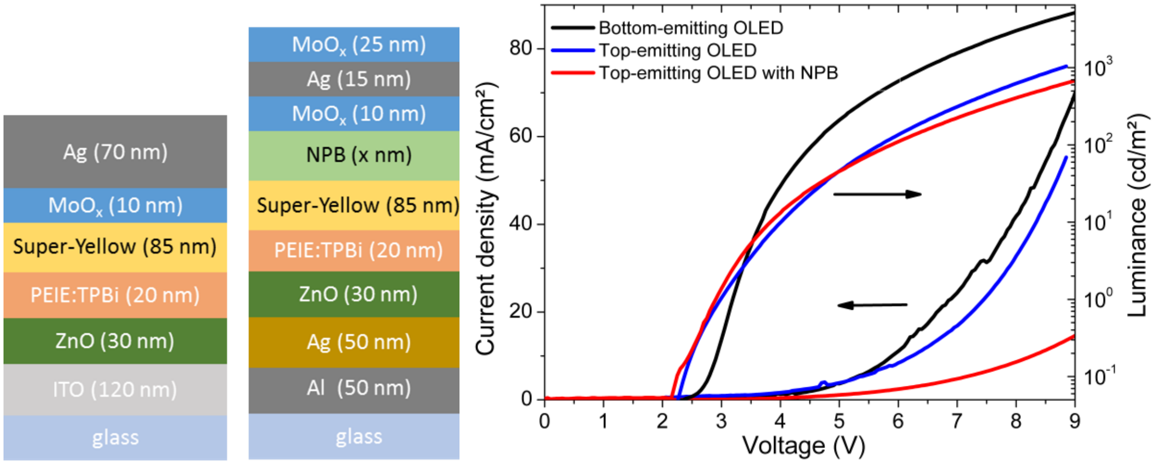

Fig. 1. Bottom-emitting inverted OLED (left) and top-emitting inverted OLED (right) fabricated by solution process and their IVL characteristics.

Table 1: Performances of bottom-emitting OLEDs (BOLEDs) and top-emitting OLEDs (TOLEDs).

Structures	**Max. Current efficiency (cd/A)**	**Max. Luminous efficiency (lm/W)**	**Luminance (cd/m²) at 20 mA/cm²**
BOLEDs	7.6 ± 0.8	4.0 ± 0.4	1220 ± 180
TOLEDs	1.8 ± 0.2	0.9 ± 0.1	375 ± 40
TOLEDs with 80 nm-thick NPB	4.3 ± 0.7	2.5 ± 0.2	930 ± 120

The electroluminescence (EL) spectrum is measured for TOLEDs at different NPB thicknesses. Fig. 2 (a) shows that the maximum electroluminescence peak is around 600 nm for TOLED without NPB, showing that the optical cavity is not optimized for the Super Yellow emission spectrum, which has its peak intensity around 550 nm. Fig. 2 (a) shows also that adding an NPB layer leads to a spectral change in the emission spectrum. For thicker NPB layers, the device emission better matches the SY emission spectrum. This result is confirmed by optical simulations which show the spectrally integrated out-coupled intensity for different HTL and ETL thicknesses in Fig. 2(b). The optimum layer thickness of 100 nm NPB and 10 nm extra ETL are close to those of the fabricated device with the best overlap of the SY emission spectrum and the measured EL spectrum.

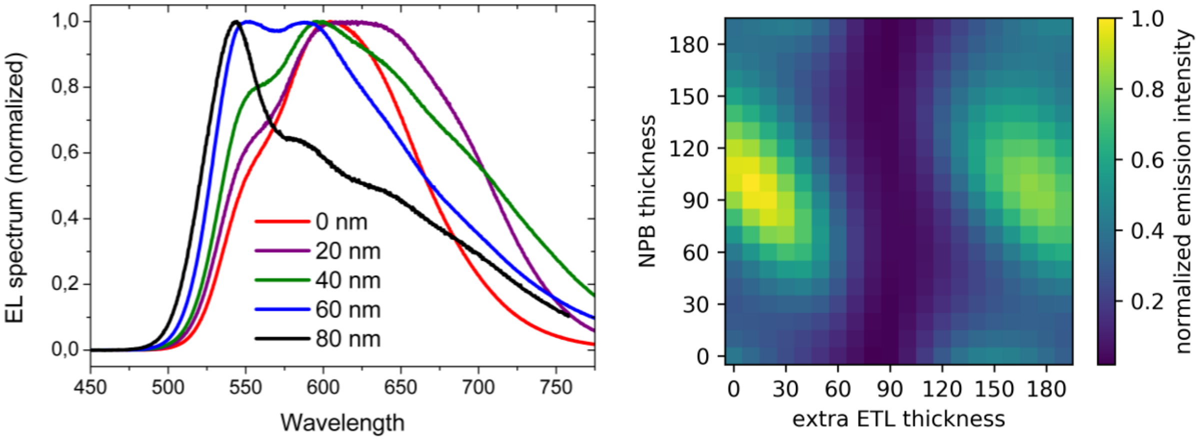

Fig. 2. (a) Electroluminescence spectra for TOLED with different NPB layer thicknesses. (b) Calculated emission intensities for different ETL and NPB thicknesses.

More tests will be conducted to increase the TOLED efficiency. The NPB-TOLED current density can be increased by doping the thick NPB layer, leading to higher luminances. PEIE and TPBi materials will also be included into the ZnO solution leading to a ternary blend deposited in one process step [12]. Instead of PEIE and TPBi, PEI could be used in a binary blend with ZnO. Indeed, PEI can also block the holes escaping from Super Yellow [13]. The process will be greatly simplified, improving also the reproducibility of OLEDs performance.

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