



Efficient transparent small-molecule organic light-emitting devices adopting laminated transparent top electrodes



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ABSTRACT

We report efficient transparent small-molecule organic light-emitting devices (OLEDs) with laminated top transparent electrode. This was made possible via the aid of the transparent conductive adhesive based on conducting polymers. With use of small-molecule materials and efficient phosphorescent emitters, rather high external quantum efficiency and current efficiency of up to (11.4%, 43.1 cd/A), much higher than those in previously reported OLEDs using laminated top electrodes, were obtained. The transparent OLEDs exhibited rather balanced electroluminescent intensities and spectra from both sides of the devices, a broad optical transmission band, and a high optical transmittance of up to 77%. It provides a simple and yet effective approach for fabrication of efficient transparent OLEDs and can be extended to flexible devices or even roll-to-roll processes in future.

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1. Introduction

Due to their various merits, organic light-emitting devices (OLEDs) have posed as promising candidates for the next-generation display and lighting technologies [1–4]. The possibility to make transparent OLEDs renders possible even more interesting and fascinating applications, ranging from transparent (see-through) displays, transparent (see-through) lighting panels, head-mounted displays, smart windows for architectural or advertising purposes, to navigation/warning displays on car windshields [5–12].

To realize transparent OLEDs, the highly reflective and opaque top metal electrodes of conventional OLEDs need to be replaced by transparent or semi-transparent electrodes. For instance, the top metal electrodes may be replaced by transparent conducting oxides (TCOs), such as indium tin oxide (ITO), indium zinc oxide etc. [5–8]. However, the sputtering process generally used for deposition of TCOs often causes damages to the organic layers/interfaces below, rendering it a very difficult process for transparent OLEDs [5–8].

Alternatively, the thick and opaque top metal electrodes may be replaced with thin and semi-transparent metal electrodes [9–12]. Yet, the transparency may be limited by the absorption loss and residual reflectivity of thin metal electrodes [9–12]. In addition, thin metal electrodes often cause strong spectral effects to transmission and emission spectra of transparent OLEDs thus made, that may distort the colour of the transparent OLEDs [9–12]. As such, a more convenient and effective method for preparation of top transparent electrodes of transparent OLEDs is still highly desired.

In devising an alternative approach for transparent top electrodes of transparent OLEDs, an earlier work by Yang et al. came to our attention [13]. In this work, Yang et al. used the modified transparent conducting polymer poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) as the transparent conductive adhesive (TCA) to laminate a top electrode onto the luminescent polymer (at elevated temperatures) to complete the polymer light-emitting device (PLEDs). The PEDOT:PSS was modified by adding a compound D-sorbitol having a high boiling point (~300 °C) and multiple polar (hydroxyl) groups as an additive, or by depositing a thin D-sorbitol layer on the surface of PEDOT:PSS [13]. D-sorbitol itself has a melting temperature of 95–100 °C [13]. Thus it is solid at room temperature, but it becomes highly viscous and sticky when heating at a temperature of ~50 °C to a temperature

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higher than its melting point [13]. Such characteristics make it useful as the adhesive additive to PEDOT:PSS or as the adhesive interlayer to bond things together. Although this early work successfully demonstrated a working PLED by this approach, the device exhibited a very low electroluminescent (EL) efficiency of 0.017 cd/A. In addition, the demonstrated device was opaque, not transparent. Whether a similar approach can be adapted for small-molecule OLEDs that can provide much higher efficiency than PLEDs and for transparent light-emitting devices, however, had not been explored. In recent years, there were also other reports of transparent or non-transparent OLEDs with direct lamination of top electrodes of different compositions, such as metals [14,15], carbon nanotubes [16–18], and graphenes etc. [19] Still, most of these works were conducted for polymer light-emitting devices and EL efficiencies achieved by these devices were very low, with external quantum efficiencies (EQEs) of only 0.1%–2%, current efficiencies of only up to 3 cd/A, and maximal brightnesses of a few hundred cd/m² at most [14–19].

In this work, we report an efficient transparent inverted small-molecule phosphorescent OLED adopting the laminated transparent top electrode via the aid of the PEDOT:PSS-based transparent conductive adhesive (TCA). The device exhibited rather high optical transmission of up to 77% and balanced emission intensities from both sides, with rather high total external quantum efficiency of up to 11.4% (corresponding to total current efficiency of 43.1 cd/A), current efficiency of up to 23.1 cd/A for top-side emission and 20.0 cd/A for bottom-side emission. The total EL efficiencies are much higher than those of previously reported devices using laminated top electrodes.

2. Experiments

Since in this work the TCA to laminate a top electrode was based on PEDOT:PSS and it is mainly hole-conducting, thus an inverted OLED configuration having the top electrode as the anode was adopted. The transparent OLED to be constructed by lamination was based on an inverted phosphorescent OLED (reference device 1 – Ref. [1], Fig. 1), having the structure of: glass/ITO cathode/Cs₂CO₃ (1 nm)/B3PYMPM (30 nm)/TPBi (15 nm)/CBP:Ir(ppy)₃ 8 wt% (15 nm)/TAPC (40 nm)/HATCN (20 nm)/Al anode (150 nm). The

bottom ITO and the top Al electrode served as the cathode and anode, respectively. Cs₂CO₃ was the electron-injection layer (EIL) [20]. B3PYMPM [4,6-bis(3,5-di(pyridin-3-yl)phenyl)-2-methylpyrimidine] and TPBi [2,2',2''-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole)] were electron-transport layers (ETLs) [20,21]. CBP [4,4'-bis(N-carbazoyl)-1,1'-biphenyl] doped with 8 wt% Ir(ppy)₃ [fac-tris(2-phen-ylpyridine)iridium(III)] was used as the emitting layer [20]. 4,4'-cyclohexylidenebis[N,N-bis(4-methylphenyl)benzenamine] (TAPC) was the hole-transport layer (HTL) [22], and dipyrzino[2,3-f:2',3'-h]quinoxaline-2,3,6,7,10,11-hexacarbonitrile (HATCN) was the hole-injection layer (HIL) [20]. All layers above the ITO cathode were deposited by thermal evaporation. The active area of devices was 1 × 1 mm² as defined by in-situ shadow masking.

To construct the transparent device by lamination, the device structure was divided into two separate substrates (lamination device, Fig. 1). The vacuum-deposited top Al anode was replaced with an adhesive transparent anode, that was initially prepared on another plastic PET [poly(ethylene terephthalate)] substrate, herein named as the top plate. The adhesive transparent anode was formed by coating a TCA layer over the PET/ITO substrate, in which TCA was the PEDOT:PSS having D-sorbitol as the additive (details of TCA to be discussed later), followed by annealing at 130 °C for 15 min. As for the other substrate (herein named as the bottom plate), it contained the layer structure of: glass/ITO cathode/Cs₂CO₃ (1 nm)/B3PYMPM (30 nm)/TPBi (15 nm)/CBP:Ir(ppy)₃ 8 wt% (15 nm)/TAPC (40 nm)/HATCN (20 nm)/D-sorbitol (15 nm), which was almost same as the layer structure in the reference device 1, except for the 15-nm D-sorbitol on top of HATCN. The thin layer of D-sorbitol on top of HATCN was deposited by thermal evaporation together with other layers beneath. Evaporated D-sorbitol films was highly transparent and homogeneous. A thin layer of D-sorbitol on top of HATCN helped to firmly bond modified PEDOT:PSS (the TCA) and the HATCN side mechanically and electrically. Initially, the structure without the D-sorbitol layer was tested for lamination. Yet without D-sorbitol, it was not possible to directly bond modified PEDOT:PSS and HATCN together, a situation very different from that in polymer devices in which modified PEDOT:PSS (the TCA) can effectively bond the polymer layer surface on the other substrate to from mechanical and electrical contacts. In this work, the top plate

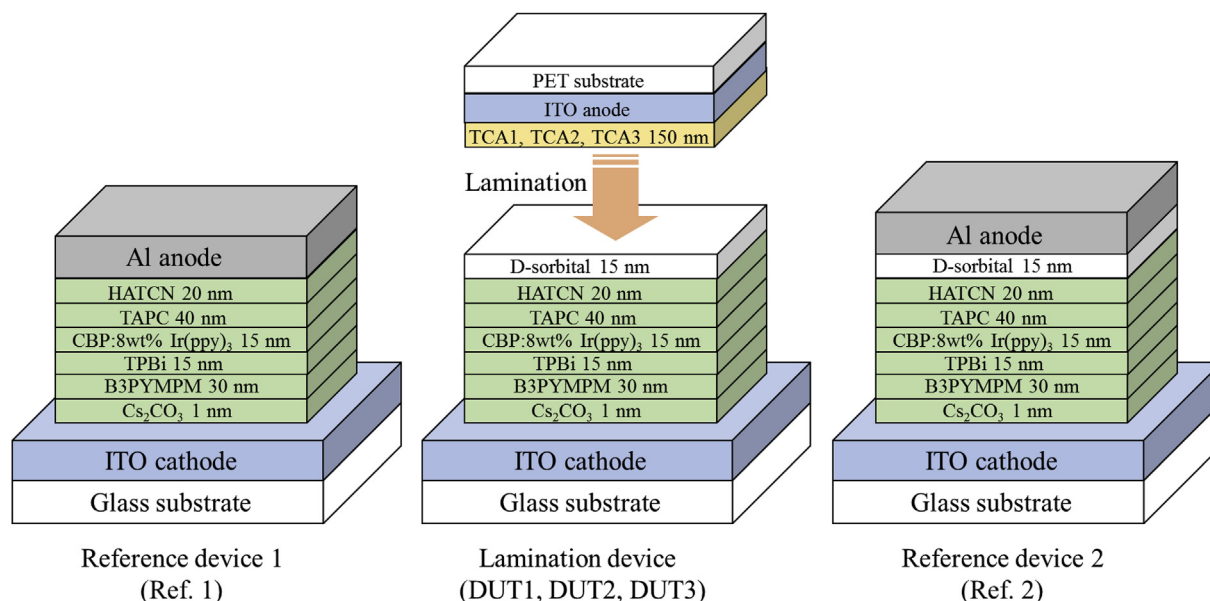


Fig. 1. Schematic device configuration and fabrication of the reference device 1 (Ref. [1]), the reference device 2 (Ref. [2]), and lamination devices (DUT1, DUT2, DUT3).

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