

Zinc Oxide and Polyethyleneimine as an Electron Transport Layer Substitute Toward Efficient, Solution-Processed Phosphorescent OLEDs

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Abstract — The standard organic light-emitting device (OLED) structure typically produces more efficient devices with lower turn-on voltages compared to an inverted architecture. However, inverted devices are more stable and compatible with n-type backplane transistors. A ZnO-based inverted device structure decreases the external quantum efficiency (EQE) from 17% to 10% as electron injection becomes more difficult. Electron injection can be improved with a polyethyleneimine interfacial layer, reducing the ZnO work function and improving the EQE to 23%. This facilitates a shift to low-cost solution processing with a simultaneous efficiency enhancement.

Keywords— OLED; efficiency; electron injection; inverted; ZnO; PEI

A simplified OLED with the following structure of indium tin oxide (ITO) [80nm] / MoO₃ [5nm] / 4,4'-Bis(N-carbazolyl)-1,1'-biphenyl (CBP) [30nm] / (95% CBP: (5%) Tris[2-phenylpyridinato-C₂,N]iridium(III) (Ir(ppy)₃) [15nm] / 2,2',2''-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) [45nm] / LiF [1nm] / Al [100nm] is depicted in Figure 1(a). In such devices, electron injection from the LiF/Al cathode into TPBi and hole injection from the ITO/MoO₃ anode toward the Ir(ppy)₃-doped emissive layer occur promptly due to the small energy barriers at the electron (ETL) and hole transport layer (HTL) interfaces as illustrated in Figure 1(b). However, when a solution processed ZnO nanoparticle film is used as the ETL this is no longer true as the conduction and valence bands are much deeper than the corresponding lowest unoccupied (LUMO) and highest occupied molecular orbitals (HOMO) of TPBi. This is evident when comparing the energy level diagram of the inverted ZnO OLED in Figure 1(c) to that of the control TPBi OLED in Figure 1(b).

A comparison of the external quantum efficiencies (EQE) between the control device and an inverted ZnO-based device in Figure 2(a) shows that while the control device is reasonably efficient with a maximum EQE of 17%, introducing ZnO leads to complete quenching of the device emission. The exciton recombination zone within the ZnO-based device is expected to occur at the ZnO-CBP interface as electrons accumulate on ZnO and holes on CBP, and exciton formation becomes injection-dependent. ZnO is a very effective exciton quencher, so excitons created at this interface undergo non-radiative recombination rather than light emission. Introducing a 4nm polyethyleneimine (PEI) layer improves the maximum EQE of such devices to 4% at high current density as the recombination zone shifts away from the ZnO quenching sites. Substituting electron transporting TPBi as the Ir(ppy)₃ host material in place of the bipolar CBP further shifts the recombination zone away from ZnO interface and essentially eliminates the quenching altogether. However, the maximum EQE of such devices decreases to 10% as seen in Figure 2(b) due to the difficulty of electron injection from ZnO to TPBi. The maximum EQE can be improved to 23% with increased electron injection into the emission layer via the incorporation of a PEI layer between ZnO and TPBi, greater than that of the thermally evaporated control devices. Thus, a solution processable electron transport layer can be substituted into an otherwise thermally evaporated device while improving performance and is a promising inorganic alternative in the shift toward large-scale fabrication of OLEDs.

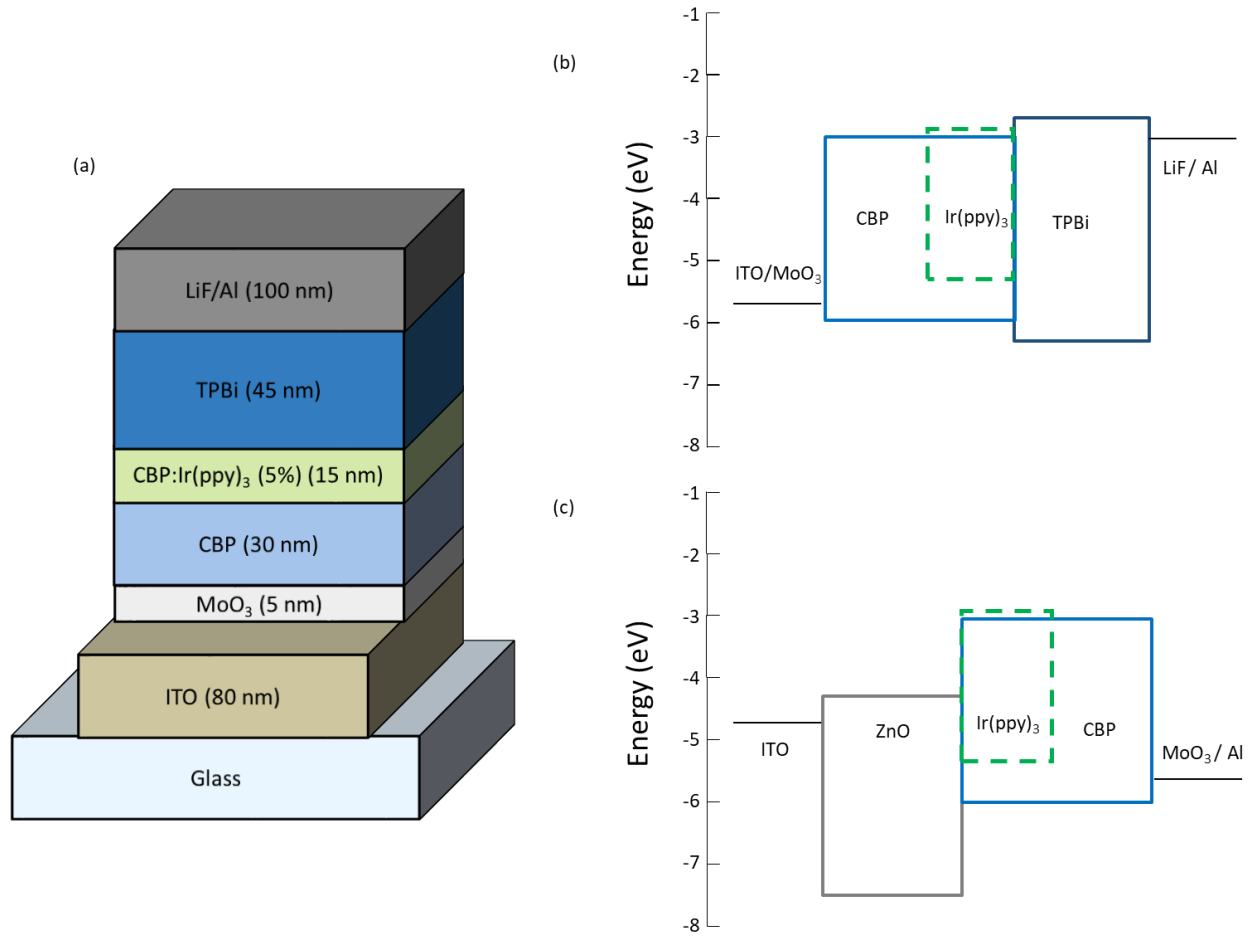


Figure 1 (a) Control OLED architecture and energy level diagrams for (b) control OLED and (c) inverted ZnO-based OLED.

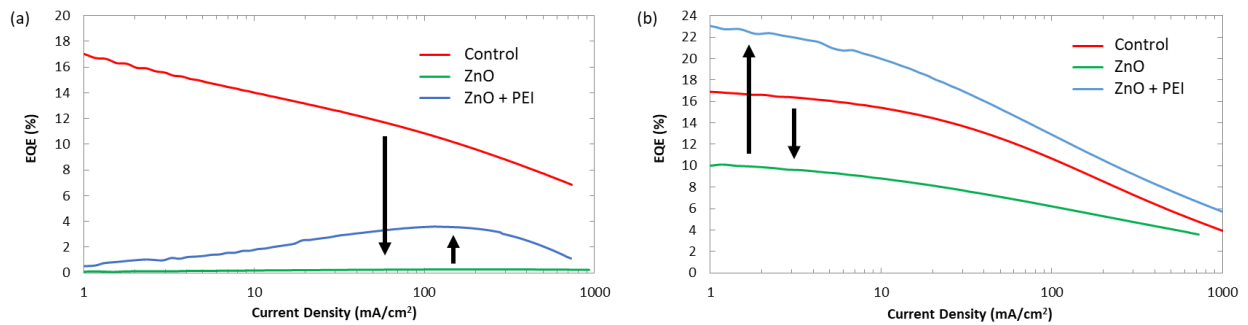


Figure 2 External quantum efficiency vs. current density characteristics for OLEDs where (a) CBP and (b) TPBi are used as the host for Ir(ppy)₃. The three device structures are the upright control OLED (red), an inverted OLED with ZnO ETL (green), and an inverted OLED with ZnO ETL and PEI EIL (blue).