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Vacuum-deposited MoO_3/Ag/WO_3 multilayered electrode for highly efficient transparent and inverted organic light-emitting diodes

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ABSTRACT

We report a highly efficient transparent and inverted organic light-emitting diode (OLED) with a vacuum-deposited MoO_3 (5 nm)/Ag (12 nm)/WO_3 (40 nm) (MAW) multilayered electrode having a low sheet resistance of 4.2 Ω/sq, an average transmittance of 88% at a wavelength of 500–550 nm, and a smooth morphology with a root-mean-square roughness of 0.339 nm. The inverted phosphorescent OLED with MAW anode exhibits a total (sum of the bottom and top emissions) maximum power efficiency of 62.1 lm/W and an external quantum efficiency of 20.1%. In addition, the transparent device can achieve almost equal bottom-to-top emission ratio of 1:1.1 and a total transmittance of > 80% in the visible-light range. The better device performance arises from the transparent MAW electrode acting as a capping layer to modulate the bottom/top emission direction and the optical properties of the device.

1. Introduction

Organic light-emitting diode (OLED) technology has been used in digital displays with great success since the beginning of the 2010s. In addition to its rapid adoption in smart-phones, the OLED has also emerged as a solution for solid-state lighting and flexible digital displays [1,2]. The OLED industry is now targeting another novel application: transparent devices [3,4]. Several transparent electronic devices are expected to enter the market shortly (e.g. head-mounted devices for audio-visual entertainment, automobile-windshield navigation devices, and real-time medical imaging devices).

Reducing the thickness of a typical metal cathode in an OLED for transparency is straightforward; however, the discontinuous thin film morphology of metal electrodes may lead to current leakage in devices and lower their stability through oxidation. Transparency may subsequently be limited by the reflectivity and surface plasma loss of semi-transparent metals [5,6]. Consequently, use of the correct transparent conducting electrode (TCE) is essential and requires thorough research to confirm potential TCE's transparency, conductivity, and mechanical durability. Gu et al. proposed a primary transparent cathode comprising a magnesium silver alloy and post-sputtered indium tin oxide (ITO) [7]. However, growing high quality ITO without damaging the underlying organic layers is challenging, and energetic plasma may bombard previously evaporated organic layers during sputtering. Recently, transparent conducting oxides created through sputtering technique have been used as the top electrode of OLEDs with various protective materials inserted afterward [8–10]. However, the relatively long sputtering process has led to other candidates for TCEs, such as graphene electrode [11,12], laminated films [13,14], and conductive multilayer electrodes (CMEs) [15–17]. In addition, metal nano-network electrodes have also been regarded as promising technique in photoelectric devices for not only because of its transparency but high operational durability [18–20]. Among all suggested TCEs, CMEs have received particular attention because of their free structural design and compatibility with the thermal evaporation process. Several dielectric/metal/dielectric (DMD) configurations [21,22] have been reported [e.g., ZnS/Ag/WO_3 (ZAW) [23], ZnS/Ag/MoO_3 (ZAM) [24], ZTO/Ag/ZTO (ZAZ) [25], and ITO/Au/V_2O_5 (IAV)] [26]. The transmittance of CMEs can be further improved by introducing thin film with nanoparticles [27]. However, few have been adopted as the top electrode in a transparent OLED with high device efficiency [28,29].

For full-color and large-size display units, active-matrix OLED (AMOLED) has been developed along with thin film transistors (TFTs). Based on an n-channel TFT, inverted OLED (IOLED) can easily match

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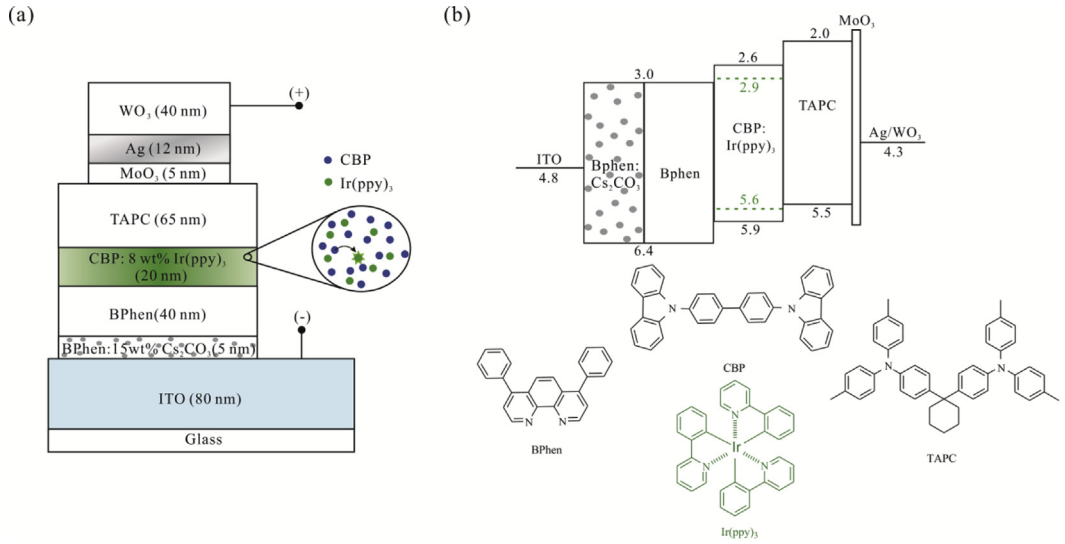

Fig. 1. (A) The schematic configuration of inverted OLED with transparent electrodes. (b) The energy band and chemical structure of organic compound used in this work.

the source and drain electrodes of a TFT at bottom. The primary challenge faced by IOLEDs is charge injection from the electrode. Approximately 20% of the maximum external quantum efficiency (EQE) of an IOLED can be achieved with perfect electron injection as suggested by Lee et al. [30]. In our previous work, we successfully presented an efficient IOLED structure utilizing a 4,7-diphenyl-l, 10-phenanthroline (BPhen): cesium carbonate (CS_2CO_3) co-deposited layer as an ideal electron-injection medium [31].

Here, we propose a highly efficient inverted OLED with an overall device transmittance of 80% in visible-light wavelengths (see Fig. 1a). The top anode comprises a thermally evaporated MoO_3/Ag/WO_3 (MAW) multilayer structure for both efficient charge injection and transparency. In contrast to the symmetrical structure of MAM (MoO_3/Ag/MoO_3) [32] or WAW (WO_3/Ag/WO_3) [33] multilayers, in MAW, MoO_3 serves as an inner hole injection layer, whereas WO_3 is employed for light extraction. The relatively lower melting point of MoO_3 (795 °C) compared with that of WO_3 (1473 °C) effectively prevents the possibility of the interfacial diffusion of a hot atom to the organic layers underneath [33]. The MAW multilayer demonstrated a low sheet resistance of 4.2 Ω/sq with an average transmittance of 81% in the visible light region. In our device, a sputtered ITO was used as the bottom cathode because direct-current sputtered ITO demonstrated a light out-coupling ability with a high refractive index and nano-pinhole properties, resulting in improvement of the device's bottom emission [34]. The OLED with a transparent MAW electrode exhibits a total (the sum of the bottom and top emissions) maximum EQE of 20.1%, the current efficiency of 67.7 cd/A, and power efficiency of 62 l m/W. In addition, the transparent device achieves an equal emission ratio for the bottom and top emission sides.

2. Experiments

All materials including CS_2CO_3 (Alfa Aesar, 99.994% metals basis, Puratronic), MoO3 (Alfa Aesar, 99.95% metals basis), and WO_3 (Alfa Aesar, 99.99% metals basis), and organic materials, BPhen, di-[4-(N,Nditolyl-amino)-phenyl]cyclohexane (TAPC), 4,4′-Bis(N -carbazolyl)-1,1′-biphenyl (CBP), and Tris [2-phenylpyridinato-C2,N]iridium (III) [Ir (ppy)$_3$] were purchased from UNI-Onward Corp., Taiwan. Before sample fabrication, all organic materials were purified twice through gradient sublimation under a high vacuum, and the thickness of each material was measured using a surface profiler (Dektak XT). The sputtered ITO substrate was fabricated following the standard procedure outlined previously [34]. Subsequently, the substrate was immersed in acetone and isopropyl alcohol for ultrasonic cleaning. After drying the remaining solution by using blown nitrogen under 5 N of pressure, the OLED device was sequentially deposited on the substrate at a base vacuum pressure of 5×10^{-6} Torr. Fig. 1 illustrates the device and material structures in this work. Our proposed device was fabricated as follows: ITO/BPhen:15 wt%CS_2CO_3 (5 nm)/BPhen (40 nm)/CBP:8 wt% Irppy$_3$ (20 nm)/TAPC (65 nm)/MoO_3 (5 nm)/Ag (12 nm)/WO_3 (40 nm).

The transmittance and absorbance of the MAW thin films were measured using an UV-visible spectrophotometer (Thermo Scientific Evolution 220). The refractive index and extinction coefficient of each thin film were measured through standard ellipsometry (Raditech SE-950). The surface morphology of the MAW thin film was investigated using an atomic force microscope (Park systems, XE-70) in non-contact mode. Device performance was simulated using the Setfos software (version 4.4; Fluxim). The current density-voltage-luminance characteristics and electroluminescent spectra of the OLED device were measured using a system that combined a Keithley 2400 source meter and a spectrum meter (Photoresearch PR655). All measurements were taken in an ambient environment.

3. Results and discussion

The DMD multilayered structures have been extensively reported to have high transparency (normally more than 70% in the visible light range), low sheet resistance (approximately 10 Ω/sq), and matching charge-injection ability [21,22]. In this study, we used MoO_3 as an efficient hole-injection layer with suitable thickness. The p-type MoO_3 injection layer not only introduced a hole into the organic layers but also acted as an antireflective layer for reflective silver with a high refractive index [35,36]. An Ag electrode is widely utilized in DMD structures because of its low extinction coefficient and high electrical conductivity compared with other common metals [37,38]. Because of light out-coupling and the prevention of Ag oxidation, WO_3 is an effective capping layer with a high refractive index [39,40]. The index-matching capping layer suppresses the radiative loss from the surface plasmon in the Ag layer [41]. We simulated the optical transmittance of the MAW thin film under various WO_3 thicknesses as shown in Fig. 2a. When WO_3 thickness was increased, the peak values of the spectra were shifted to a long wavelength and began to decrease when WO_3 thickness exceeded 40 nm. The transmittance of the MAW thin film at a long