

The effects of ultra-thin cerium fluoride film as the anode buffer layer on the electrical characteristics of organic light emitting diodes



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ABSTRACT

In this study, the efficiency of organic light-emitting diodes (OLEDs) was enhanced by depositing a CeF₃ film as an ultra-thin buffer layer between the indium tin oxide (ITO) electrode and α -naphthylphenylbiphenyldiamine (NPB) hole transport layer, with the structure configuration ITO/CeF₃ (0.5, 1, and 1.5 nm)/ α -naphthylphenylbiphenyl diamine (NPB) (40 nm)/tris(8-hydroxyquinoline) aluminum (Alq₃) (60 nm)/lithium fluoride (LiF) (1 nm)/Al (150 nm). The enhancement mechanism was systematically investigated via several approaches. The X-ray photoelectron spectroscopy and ultraviolet photoelectron spectroscopy results revealed the formation of the UV-ozone treated CeF₃ film. The work function increased from 4.8 eV (standard ITO electrode) to 5.22 eV (0.5-nm-thick UV-ozone treated CeF₃ film deposited on the ITO electrode). The surface roughness of the UV-ozone treated CeF₃ film was smoother than that of the standard ITO electrode. Further, the UV-ozone treated CeF₃ film increased both the surface energy and polarity, as determined from contact angle measurements. In addition, admittance spectroscopy measurements showed an increased capacitance and conductance of the OLEDs. Accordingly, the turn-on voltage decreased from 4.2 V to 3.6 V at 1 mA/cm², the luminance increased from 7588 cd/m² to 24760 cd/m², and the current efficiency increased from 3.2 cd/A to 3.8 cd/A when the 0.5-nm-thick UV-ozone treated CeF₃ film was inserted into the OLEDs.

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1. Introduction

Organic light-emitting diodes (OLEDs) have attracted much attention for application in solid-state lighting and flat-panel displays due to their low-voltage operation, wide viewing angles, high contrast, flexibility, low cost, low weight, fast response, and full color reproduction when compared to liquid-crystal displays (LCDs) and light-emitting diodes (LEDs) [1–5]. Tang and VanSlyke reported the first efficient OLEDs, which had a structure consisting of double-layer organic materials sandwiched between two electrodes [1]. Indium tin oxide (ITO) is the most commonly used anode material due to its high transparency (~90% at 550 nm), low resistivity (~2–4 × 10⁻⁴ Ω cm), and high work function (~4.8 eV) in

OLEDs [6,7]. However, the ITO work function is insufficiently high to reduce the hole injection barrier, and so an energy barrier still exists at the ITO/organic interface despite it having been subjected to O₂-plasma or ultraviolet (UV)-ozone treatment [7–9]. Therefore, improving the charge balance is necessary for increasing the efficiency of OLEDs. Because the mobility of holes is much higher than that of electrons in organic materials, hole/electron injection and transport ability must be improved to obtain a better charge balance in an OLED. In order to enhance the charge injection at the interface and ultimately reduce the driving voltage and improve the power efficiency of the device, one solution is to insert an anode buffer layer between the ITO and the hole transport layer (HTL), thereby reducing the energy barrier.

Recently, the insertion of metal-oxides with high work functions such as molybdenum oxide (MoO₃) [10,11], tungsten oxide (WO₃) [12,13], vanadium pentoxide (V₂O₅) [14,15], and tantalum pentoxide (Ta₂O₅) [16], have been used as anode buffer layers for

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the injection of holes to improve both the charge injection at the interface and power efficiency. To this end, formations of various metal-doped ITO layers have been studied [3–5,10–16]. However, very few studies have reported metal-fluorides, such as NaF [17], CuF_2 [18], and AgF [19], as anode buffer layers to improve the charge injection at the interface and power efficiency of OLEDs. Moreover, to our knowledge no studies have attempted to improve the performance of OLEDs by using an ultra-thin CeF_3 buffer layer (rare earth fluorides), which is the motivation of the present work. The rare earth fluorides are being extensively researched because of their mechanical and chemical stability. CeF_3 is one of the potent rare earth fluorides attracting more attention because of its technological importance and superior properties. In our previous study, we used NaF, CuF_2 , and AgF layers as anode buffer layers (ABL) to improve the hole injection and power efficiency of OLEDs [17–19].

In this study, an ultra-thin CeF_3 layer was used as an ABL to improve the hole injection and power efficiency of OLEDs. CeF_3 layers of various thicknesses were grown via vacuum vapor evaporation on ITO/glass substrates, followed by UV-ozone treatment. The effects of the CeF_3 layer on the electrical and optical properties of the OLEDs were investigated using contact angle measurements, X-ray photoelectron spectroscopy (XPS), ultraviolet photoelectron spectroscopy (UPS), atomic force microscopy (AFM), and admittance spectroscopy analyses [3–5,17–27].

2. Experimental procedures

Prior to film deposition, the glass/ITO substrates with a sheet resistance of $15 \Omega/\text{sq}$, were ultrasonically cleaned using a neutral detergent/deionized (DI) water (1:3 volume) mixture, DI water, isopropanol, and ethanol in sequence, and then subjected to UV-ozone treatment in a Jelight UVO-42 system for 20 min. The structure of the devices is ITO/ CeF_3 (X nm)/ α -naphthylphenylbiphenyl diamine (NPB) (40 nm)/tris(8-hydroxyquinoline) aluminum (Alq_3) (60 nm)/lithium fluoride (LiF) (1 nm)/Al (150 nm). High purity cerium fluoride, purchased from Merck (purity, 99.9%), was used as the anode buffer layer material. Further, NPB was used as the hole-transport layer, Alq_3 was used as the electron-transport and emission layers, and the LiF/Al layers were used as the composite cathode to enhance electron injection. All layers were deposited in a high-vacuum thermal deposition system with a base pressure of 5×10^{-6} torr. The film thickness and deposition rate were controlled by a calibrated quartz-crystal oscillator, the deposition rates of which were $0.1 \text{ \AA}/\text{s}$ for the inorganic layers, about $1 \text{ \AA}/\text{s}$ for the organic layer, and $5 \text{ \AA}/\text{s}$ for the Al metal cathode. The active area of the devices was $2.5 \text{ mm} \times 2.5 \text{ mm}$ [3–5].

The current density-luminance-voltage characteristics and the electroluminescence spectra of the devices were respectively measured with a source meter (Keithley2400) and a luminance meter (LS-100 and PR650). Admittance spectroscopy measurements were performed in the frequency range of 100 Hz to 10 MHz using an Agilent 4294A impedance analyzer. The surface morphology of the cerium fluoride films grown on the glass/ITO substrate were characterized using AFM in tapping mode under ambient conditions. The chemical changes of the CeF_3 /ITO surface and the work function of the CeF_3 thin films were examined using XPS and UPS observations. XPS measurements were performed with a VG ESCA210 XPS system with monochromatic Al K α (1486.6 eV) as the radiation source. For charge reference, the C 1s peak at 284.6 eV was used. UPS measurements were performed in air with a Riken Keiki AC-2 system. Further, the polarity and surface energy were determined through contact angle measurements using the sessile drop and contact angle goniometer (MagicDrop, USA) techniques. The test liquids drop on the surface of the solid, and measured the angles through contact angle goniometer. For contact angle mea-

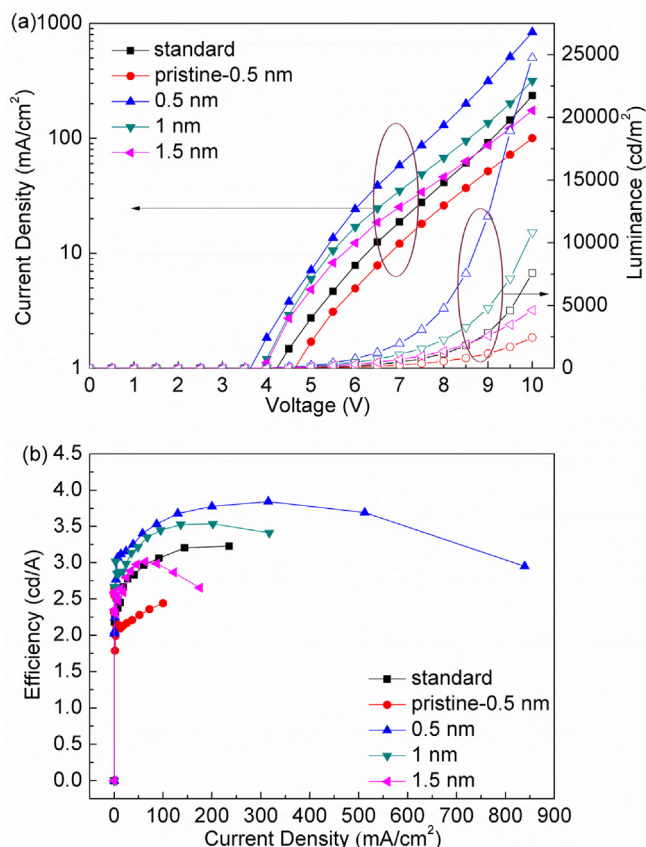


Fig. 1. (a) Current density-voltage-luminance and (b) current efficiency-current density characteristics of the OLEDs with standard ITO, pristine 0.5 nm CeF_3 anode buffer layer, and UV-ozone treated 0.5, 1, and 1.5 nm CeF_3 anode buffer layers deposited onto the ITO substrates.

surements, DI water and methylene iodide (CH_2I_2) were used as the test liquids, the surface energies from the measured contact angles (θ) of which were calculated via the following geometric mean expression:

$$\gamma_L \times (1 + \cos \theta) = 2(\gamma_s^p \times \gamma_L^p)^{\frac{1}{2}} + 2(\gamma_s^d \times \gamma_L^d)^{\frac{1}{2}} \quad (1)$$

where γ_L ($= \gamma_L^p + \gamma_L^d$) and γ_s ($= \gamma_s^p + \gamma_s^d$) are the surface tension of the test liquid and surface energy of the solid, respectively. These two values can be readily estimated from contact angles of different test liquids by solving two simultaneous equations. Surface tension data of H_2O and CH_2I_2 , which respectively represent polar and non-polar solvents, are given in the supplementary material shown in reference [21].

3. Results and discussion

3.1. Current density-voltage-luminance

Fig. 1(a) and (b) respectively shows the current density-voltage-luminance and current efficiency-current density characteristics of OLEDs with a pristine 0.5 nm CeF_3 anode buffer layer and UV-ozone treated 0.5, 1, and 1.5 nm CeF_3 anode buffer layers deposited onto the ITO substrate. Compared to the devices with the UV-ozone-treated CeF_3 layers, the pristine CeF_3 device had a lower current density, luminance and current efficiency. The optimal thickness for the UV-ozone treated CeF_3 layer was found to be 0.5 nm. When a 0.5 nm-thick UV-ozone-treated CeF_3 layer was inserted, the turn-on voltage decreased from 4.2 to 3.6 V (at $1 \text{ mA}/\text{cm}^2$) compared with the standard device, and then increased to 4.0 V when the layer

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