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Optical and electrical effects of nickel oxide interlayer for anode-recessed organic light-emitting diodes

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ABSTRACT

Forming a nickel oxide (NiO_x) layer on recessed indium tin oxide (ITO) anodes has successfully enhanced the current efficiency of an organic light emitting diode (OLED) by up to 46%. The recesses largely increased the haze of ITO film and led to a 32.8% current efficiency enhancement of a planar OLED. The introduced NiO_x interlayer allowed another 12.9% efficiency enhancement mainly due to lowered potential barrier at the ITO/organic interface and further elevated haze that led to a more efficient light extraction.

1. Introduction

It is well known that the external quantum efficiency of a conventional organic light-emitting diode (OLED) generally remains below 20% mainly due to the internal total reflections in its refractive-index-mismatched structures and the optical absorption by surface plasmon poraritons near metallic electrodes [1–5]. A variety of light extraction approaches have been proposed to eliminate such light trappings in stacked OLED structures. Approaches employing structures such as micro-lens [6–8], photonic crystals [9,10], optical grating [11,12], scattering media [13,14] have demonstrated to be able to minimize the total reflections and enhance light out-coupling to some extends. Among them, the use of a micro-lens or scattering film outside a glass substrate allows the glass-mode total reflection to reduce and has been commercially available. In contrast, the media used to reduce the wave-guided mode total reflection such as photonic crystals and optical gratings are not yet practically applicable. This is because these media are built in the stacked OLED structure and would potentially deteriorate device reliability for their poor contacts with the organics. Besides, the processing for yielding such media is usually complicated. Minimizing wave-guided mode total reflection with no additional complicated media in the existing OLED structure is then still an important topic to address.

Koh *etc.* [15] have reported that a micro-structured indium tin oxide (ITO) could be used to enhance optical out-coupling of an OLED without extra media. However, they had to use low-reflective-index PEDOT:PSS polymer to replace part of ITO and that limited OLED performances. Aziz etc. [16] recently reported a light extraction enhancement approach which simply roughed ITO surfaces (Ra 3.5 nm → 8.3 nm) to yield a light scattering effect at the ITO/organic interface. This approach employed no additional media and has been shown to improve not only the light out-coupling efficiency but also the lifetime of OLEDs. In our earlier reports [17], we have also demonstrated that the power efficiency of OLEDs could be enhanced by up to 28% using recessed ITO anodes. The recessed ITO was directly in contact with organics without any additional medium in between. The power efficiency enhancement was found to be attributed to the optical effects at the recessed ITO/organic interfaces. These approaches were all proved to enhance light extraction efficiency without extra media and are expected to have higher potential for practical OLED applications.

Based on the recessed ITO approach developed previously, in this study we added a nickel oxide (NiO_x) thin layer on top of a recessed ITO anode and investigated its effect on the performances of OLEDs. The introduction of this NiO_x layer aimed to enhance

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OLED performances by lowering the potential barrier between ITO and organics [18,19]. This is because NiO_x has a higher work function (~5.1 eV) than ITO (~4.6 eV), therefore the carrier injection at the ITO/organic interface can be promoted. However, the introduced NiO_x layer may also generate additional optical effects to the existing recessed ITO film. It is then interesting to understand how both optical effect and electrical effect of this NiO_x layer influence the characteristics of OLEDs. The dominating factors for the performances of OLEDs were examined and will be discussed.

2. Experiments

Commercially available ITO films with a thickness of 250 nm on glass were recessed using traditional photolithography and wet etching processes. The recesses were designed to have four types of circular patterns individually arrayed in four areas. Each pattern has physical dimensions of 4 μm or 6 μm in diameter and 10 μm or 12.5 μm pitched. To fabricate these recesses, ITO/glass substrates were first cleaned using acetone, alcohol and distilled water sequentially in an ultrasonic bath for 15 min in each step. The substrates were nitrogen brown and baked at 100 °C for 10 min before a standard photolithography process was conducted for recess pattern define. After the photolithography process, ITO films were masked with photoresist of designed circular holes. Etching of ITO films through the photoresist masked circular holes to a depth of 150 nm using hydrochloride acid was followed. Recesses were formed after the photoresist was removed from ITO surfaces. The percentage of recessed area, or recess coverage ratio, in each pattern area was measured to be 7.9%, 14.1%, 20.9% and 27.7%. A NiO_x layer was then formed on top of the recessed ITO by coating a 5-nm-thick Ni using e-beam evaporation at room temperature and thermally annealed in vacuum at 300 °C for 2 h.

The fabrication of OLED devices was conducted on both recessed ITO and NiO_x/recessed ITO substrates. The OLED device consists of a glass/ITO/N,N″-di-phenyl-N,N″-di-[4-(N,N-di-phenyl-amino) phenyl]benzidine (NPNPB, 75 nm)/n-Propyl bromide (NPB, 10 nm)/ 4,4′,4″-Tri(9-carbazoyl)triphenylamine (TCTA, 5 nm)/WHP401 (Wanhsiang Corp., 37.5 nm) doped 7% Tris(2-phenylpyridinato) iridium(III) (Ir(ppy)3)/(10-Hydroxybenzo[h]quinolinate) beryllium complex (BeBQ2, 30 nm)/LiF(0.5 nm)/Al(150 nm) structure. Fabrication of OLED devices commenced with the ITO surface treatment in a 100 W oxygen plasma ambient for 10 min to guarantee a clean surface in contact with the organics. The organics were then sequentially coated on top of the ITO anode using thermal evaporation at room temperature, and this was followed by the thermal evaporation of LiF and Al to complete the device. Figure 1 schematically shows the cross section of the complete OLED device with a NiO_x/recessed ITO anode. All devices were finally packaged in a glove box before being characterized.

Surface morphologies of recessed ITO films with and without the NiO_x top layer were observed using a JEOL/JSM-6701F scanning electron microscope (SEM) and a Veeco CP-R II atomic force microscopy (AFM). Optical haze, transmittance, and reflectance were examined using a NIPPON DENSHOKU INDUSTRIES NHD-5000 haze meter and a Labguide TRF2006 UV–Vis spectrometry. The electrical resistance was measured by an Ecopia HMS-3000 Hall effect measurement unit. Current-voltage (*J*-V) and luminance-voltage (*L*-V) characteristics of OLED devices were determined using a Keithly Instruments model 237 source-measure unit and a PhotoResearch PR-650 optical spectrometer.

3. Results and discussions

The SEM image in Fig. 1 reveals the morphology of a recessed ITO film. It can be clearly seen that the recesses generated using

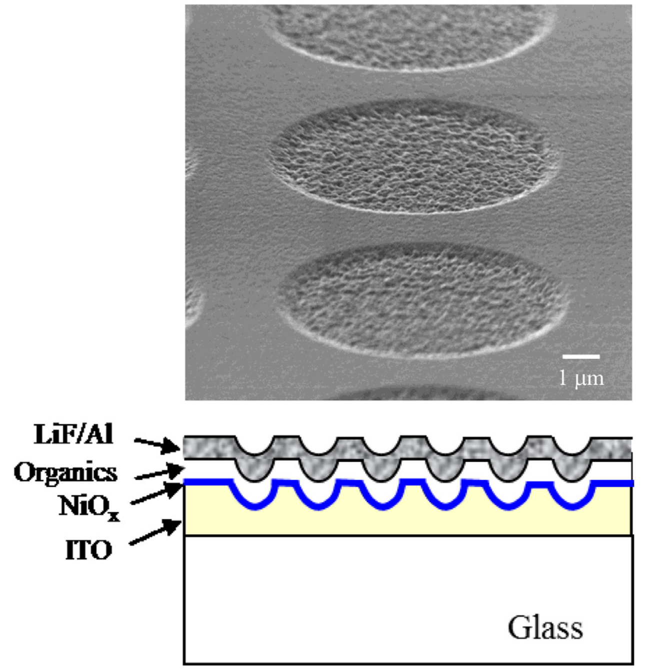

Fig. 1. Schematic of OLED device structure with a NiO_x/recessed ITO anode and the SEM image of a 150 nm recessed ITO film.

chemical wet etching appear to have a flat bottom and a tapered wall, but their surfaces have been slightly roughed. The depths of the recesses in all four areas, measured by an α-step surface profilimeter, vary indistinctly between 146.2 and 152.8 nm. This suggests that the micro-loading effect during chemical etching was not prominent and all recesses at various coverage ratios were assumed to have the same depth. Figures 2 (a) - (c) show the AFM images of initial, recessed, and NiO_x/recessed ITO films, respectively. These AFM images again identifies that both recessed and NiO_x/recessed surfaces are flat and free of apparent spikes. The averaged recess depth measured from AFM profiles is 150.7 nm, which is nearly the same as those obtained by the α-step surface profilimeter. The images also give clear evidence that the recesses have surfaces rougher than the initial ITO surface. The surface roughness (Ra) of initial ITO was 0.4 nm, and it increased to 3.3 nm when recessed and decreased to 2.9 nm when the recessed ITO was coated with a NiO_x layer. It seemed that the presence of a NiO_x top layer could slightly smooth the surface of the recess.

To realize how this NiO_x layer influences the performance of a recessed OLED, *J*-V and *L*-V characteristics of both recessed and NiO_x/recessed OLEDs were examined. Figure 3 (a) compares the *J*-V characteristics of OLEDs with a planar anode and recessed ITO anodes. It can be seen that all recessed OLEDs performed improved *J*-V characteristics with lower current leakages. At an applied voltage of 4 V, the current density of the planar device was measured to be 1.43 mA/cm^2, and it noticeably increased to 2.33 mA/cm^2 as ITO was 7.9% recessed. But, the current density gradually decreased to 2.17, 1.94, and 1.59 mA/cm^2 as the recess coverage ratio increased to 14.1%, 20.9%, and 27.7%, respectively. This observation suggests (1) carriers could be injected into the device more effectively through the recessed ITO surfaces; (2) the current density tends to decrease when more ITO is removed away from its surface, and this is believed to be the consequence of increasing electrical resistance at high recess coverage ratios. Indeed, the planar ITO had an electrical resistance of 5.4 Ω/□, and it increased to 6.2 Ω/□ and 8.4 Ω/□ when ITO was 7.9% and 27.7%