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Enhancement of a top emission organic light-emitting diode with a double buffer layer

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ABSTRACT

We report on luminance characteristics of top emission organic light-emitting diode (TEOLED) containing transparent cathode with various structures of buffer layers. Especially, we have focused on the buffer layers preventing OLED degradation from plasma damages which occur during deposition of inorganic passivation layer. The TEOLED with a double buffer layer showed much higher luminance and lower leakage current than that with single buffer layer of organic layer or inorganic layer.

1. Introduction

Organic light-emitting diodes (OLEDs) are ideal for flat panel display (FPD) applications due to their full color gamut and thin form factor [1]. Advantages of OLEDs among FPDs include their high emission efficiency in the visible spectrum, ease of processing, and seemingly infinite possibilities for modification [2]. Top emission organic light-emitting diode (TEOLED) have generated considerable interest in recent years, primarily owing to their use in active matrix displays [3]. The TEOLED is of great importance for achieving monolithic integration of OLEDs on a silicon chip, where the electrical drivers and pixel switching elements are located on opaque substrates, thus necessitating a transparent top electrode structure [4]. Top-emitter encapsulation requires full transparency and a moisture blocking barrier layer, preferably with no desiccant. Various schemes for thin-film encapsulation of TEOLEDs have been reported [5–7]. However, most of these schemes are based on physical vapor deposition (PVD) processes and low-temperature plasma-enhanced chemical vapor deposition (CVD) processes. These methods are not sufficiently robust to undergo accelerated environmental tests needed for product development [6,7]. Ghost et al. reported that thin-film encapsulation of OLED devices should meet the following criteria: (i) formation of a highly transparent layer (>90% transmission in the visible region); (ii) utilization of a low-stress material so that sufficiently thick layers can be developed; (iii) utilization of low processing temperature (preferably below 100 °C, maximum limit about 130 °C); (iv) application of a densely packed, continuous, and highly conformal coating, so that all small particles are well encapsulated; (v) realization of a pinhole-free coating; (vi) minimal UV light or sputtering damage [5]. These requirements should be more stringent in the case of flexible display units, which have huge applicability in consumer electronics [6–9].

Most reported deposition techniques for inorganic films are based on a plasma-enhanced process that is injurious to cathode metal materials or organic materials. Moreover, a single organic layer or inorganic thin-film passivation layer is not sufficiently dense to protect the OLEDs from permeation by moisture and oxygen [10]. A low-temperature thin-film deposition process is desirable, because high process temperatures are incompatible with OLED fabrication process. SiN_x, SiO_x, SIO_xN_y, AlO_x, and Al_2O_3:N films are currently employed as inorganic passivation layers for OLEDs [11]. In order to resolve this disadvantage, an indium zinc oxide (IZO) thin-film with buffer layers (B.L.) was applied. Amorphous IZO (a-IZO), an indium oxide-based transparent conductor, offers several advantages. For instance, unlike either crystalline or a-ITO, the lowest a-IZO resistivity ($3–6 \times 10^{-4}$ Ωcm) is found when no reaction oxygen is added to the sputter chamber, thus greatly simplifying the deposition process [12].

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In this work, we report on the enhancement of luminance characteristics in TEOLEDs with double B.L. to prevent OLED degradation from plasma damage occurring during deposition of the inorganic layer. We compared the lifetime of TEOLEDs with and without B.L. Improvement in the characteristics of the TEOLED is demonstrated.

2. Experimental

The devices have been fabricated on ITO coated glass substrates. The ITO-coated substrates were cleaned ultrasonically in chromerge, acetone, and methanol, respectively. Prior to vacuum deposition of the organic materials, the substrates were treated with oxygen plasma for 3 min. The TEOLEDs were deposited by conventional vacuum thermal evaporation, in a vacuum of less than 3×10^{-7} Torr. The substrate was then transferred into a separate sputtering chamber without breaking the vacuum. Finally, an IZO film was deposited by rf magnetron sputtering as a passivation layer, in a vacuum of 5 mTorr. We prepared four kinds of devices: device 1: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/IZO; device 2: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/1stB.L./IZO; device 3: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/2ndB.L./IZO; device 4: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/1stB.L./2ndB.L./IZO. The IZO film was deposited by low rf magnetron sputtering power at room temperature. The sputtering gas was a mixture of 99.9999% pure argon and 99.99% pure oxygen. The TEOLED structure is Glass/ITO/NPB/Alq_3/LiF/Al/Ag/1stB.L.(50 nm)/2ndB.L. (10 nm)/IZO(50 nm). A 60 nm *N*,*N*′-bis-(1-naphth1)-dipheny1-1, 1′-bipheny1-4,4′-diamine (NPB) film was used as a hole-transporting layer, and a 60 nm tris-(-8-hydroxyquinoline) aluminum (Alq_3) film was used as a light-emitting and electron-transporting layer. A 1 nm lithium fluoride (LiF)/2 nm Al/20 nm Ag structure was used as a cathode. A 50 nm NPB and a 10 nm LiF were used as a 1st B.L. and a 2nd B.L. deposited by thermal evaporation, respectively. The thickness optimization result of the B.L. was used in this experiment. The passivation layer of IZO film was deposited with a 20 cm target to substrate distance by rf magnetron sputtering. In this study, the light from both sides of the transparent OLED was measured from top cathode. Electroluminescence (EL) spectra were obtained by a Minolta CS-1000. The current/voltage and luminescence/voltage characteristics were assessed with a current/voltage source/measure unit (Keithley 238) and a Minolta LS-100. Luminance was measured in the surface direction.

3. Results and discussion

The total pressure during the sputtering process was 5×10^{-3} Torr in argon mixed with 1% oxygen flow regulated by mass flow controllers at room temperature. It is very well known that at sputtering power levels the resistivity increases sharply at a certain percentage of oxygen, while the transmission remains fairly constant [12]. It has been speculated that the cathode may be damaged by ion bombardment during rf magnetron sputtering deposition [13]. The ion bombardment energy increases with increased rf sputtering power, and overly intensive bombarding can damage the organic layer [14,15]. Therefore, IZO films with a double B.L. are used to reduce bombardment damage to the organic layer. Fig. 1 shows current density (*I*)–voltage (*V*)–luminance (*L*) characteristics of the TEOLED devices. The *I*–*V*–*L* curves reveal degraded TEOLED performance resulting from sputtering plasma damage. Device 1 shows a high leakage current density at a reverse bias. This leakage current density is much higher than that of the TEOLED with buffer layers. The turn-on voltage of device 1, device 2, device 3, and device 4 was 5.5 V, 4.5 V, 4.5 V, and 2 V, respectively. Hung and Madathil demonstrated a new transparent top

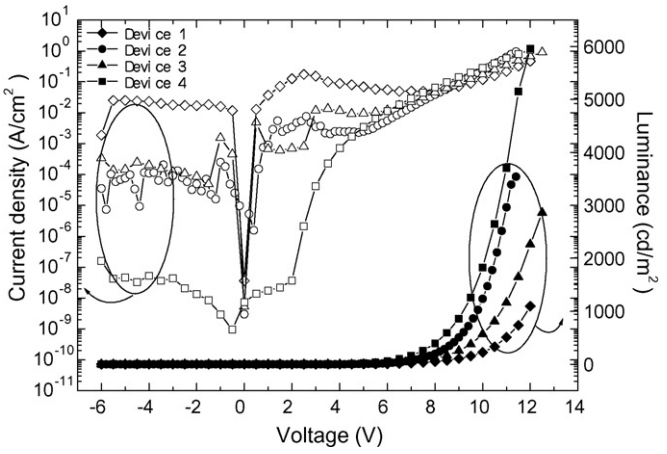

Fig. 1. Current density (*I*)–voltage (*V*)–luminance (*L*) characteristics of the TEOLED devices with buffer layers. Device 1: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/IZO; device 2: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/1st B.L./IZO; device 3: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/2ndB.L./IZO; device 4: Glass/ITO/NPB/Alq_3/LiF/Al/Ag/1stB.L./2ndB.L./IZO.

electrode employing a thin organic film of copper phthalocyanine (CuPc) as a B.L. overlaid by a sputter-deposited ITO film. The CuPc B.L. effectively minimized the radiation damage inflicted on the OLED organic layer stack during the ITO deposition. The reasons are uncertain, as a metal layer normally would provide better protection against irradiation than an organic layer [16].

The luminance was roughly 6000 cd/m^2 at 12 V in the case of device 4, which is almost two times higher than that of device 2 and device 3, respectively. Devices 2 and 3 show a higher leakage current density at a reverse bias in comparison with device 4. The higher leakage current density of the TEOLED deposited by rf magnetron sputtering is attributed to the bombardment of energetic particles during the sputtering process [17]. Therefore, the double B.L. could be used between the metal cathode and IZO film by the evaporation as a damage-free process. The *I*–*V* characteristics of device 4 were dramatically improved by the double B.L. composed of organic and inorganic layers. Kim et al. reported that OLEDs exhibited a high leakage current density at a forward bias before the turn on of OLEDs due to a large shunt resistance, which was indicative of leaky interfaces between Al and LiF/Alq_3 layers [17,18]. The high leakage current density is believed to be caused by the bombardment of energetic particles during the sputtering process, which results in damage to the underlying organic layers. *I*–*V*–*L* results of device 4 indicate that plasma damage during the IZO sputtering process was minimized as a result of double B.L. protection.

Fig. 2 shows the external EL quantum efficiency versus applied voltage for the TEOLEDs. The efficiency could be further improved in device 1. The external EL quantum efficiency increases with increasing voltage up to 10–12 V. The low turn-on voltage of the device with the B.L. might be attributed to plasma damage, the bombardment of particles, and/or sputtering of atoms during the sputtering process. The insert of Fig. 2 shows the EL spectra of TEOLEDs with B.L. (at 20 mA/cm^2). Devices 1, 2, 3, and 4 show maximum emission at about 530, 536, 531, and 540 nm, respectively. The EL peak appeared at 530 nm in a bottom emission OLED with Alq_3 based on ITO/glass (not shown). The emission spectra are similar to those obtained for Alq_3-based bottom emission OLEDs. The optical transmittance of glass/1st B.L./2nd B.L./IZO was greater than 90% at 515 nm, and was measured for all samples. The samples showed similar transmittance, but difference thickness and materials becomes absorptive at short wavelengths ($\lambda < 400$ nm). In order

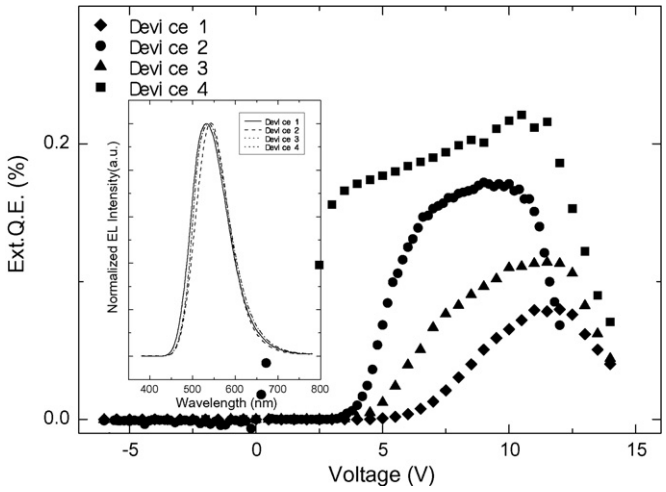

Fig. 2. External electroluminescence quantum efficiency vs. voltage for the TEOLEDs. The insert of Fig. 2: electroluminescence spectra of TEOLEDs with buffer layers (at 20 mA/cm^2).

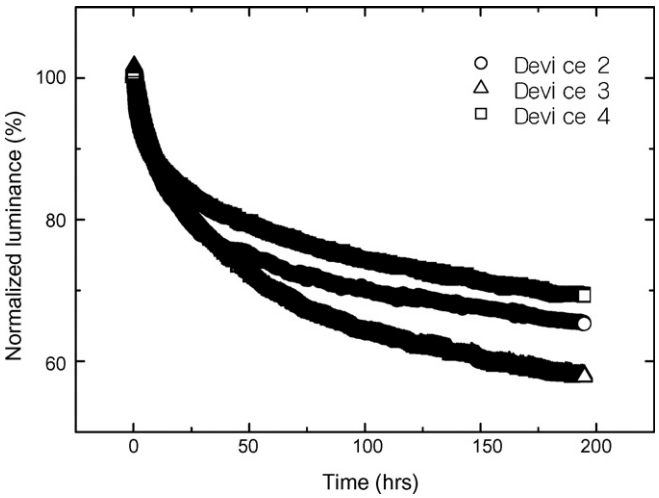

Fig. 4. Life time of normalized luminance vs. operating time for IZO thin-film encapsulated TEOLEDs with buffer layer.

to apply the B.L. to TEOLEDs, the deposited B.L. should have high transmittance, because most of the emitted light must be extracted through this double layer.

Fig. 3 shows the emission images of devices 1, 2, 3, and 4. The dark spots in the images were caused by bombardment of energetic particles during the sputtering process (Fig. 3(a)). This verifies that a single organic layer (Fig. 3(b)) or inorganic layer (Fig. 3(c)) cannot adequately protect the device from plasma damage during the sputtering process. Ke et al. reported that electrical shorts and metal diffusion into organics lead to the formation of dark spots and device failure [19]. These short circuit points also lead to the formation of pinholes and local fusion of the organics and the metallic cathode [18]. It is clear that there was negligible degradation in device 4 (Fig. 3(d)).

Fig. 4 shows a typical plot of normalized luminance versus operating time for IZO thin-film encapsulated TEOLEDs with the proposed B.L. The lifetime of an OLED is generally defined as the time to decay to half of the initial luminance at a constant current [20]. The devices were lifetime tested at room temperature and ambient pressure under dc driven at constant current conditions.

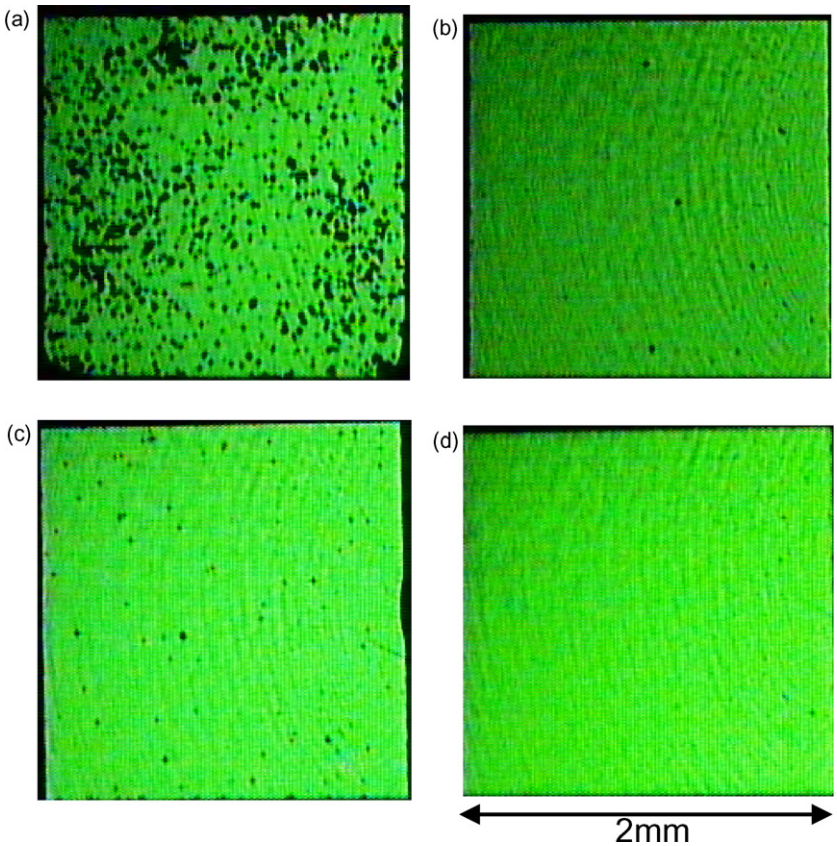

Fig. 3. The electro-luminescence images of TEOLEDs with B.L. (a) device 1, (b) device 2, (c) device 3, and (d) device 4.

To determine the rate of the degradation, a current density of 10 mA/cm^2 was applied to the devices, yielding an initial luminance of 1000 cd/m^2 for the devices. The lifetime of device 1 could not be measured because it failed immediately after emitting light at 1000 cd/m^2. The device having a double B.L. was observed to have much longer lifetime than either device with single buffer layer at a constant current density of 10 mA/cm^2. Normally, the luminance decay curves of an OLED show gradually decreasing behavior with operating time [19]. The OLED encapsulated by SiN_x film, showing a lifetime of 120 h, displayed a decay curve of approximately −30%/100 h at 500 cd/m^2 [18]. To the contrary, the rate of luminance decrease of the TEOLED with the double B.L. was approximately −26%/100 h at 1000 cd/m^2, slower than a previous report even at a higher luminance condition. The slow rate of decrease of luminance for the TEOLED with a double B.L. and an IZO passivation layer verifies that the B.L. and passivation layer provide good passivation. The *I–V–L* results and lifetime data of the TEOLED passivated with a double B.L. and an IZO film deposited by rf magnetron sputtering suggest that the double B.L. technique is promising for realizing plasma damage-free devices. The double B.L. on the TEOLEDs is very effective in terms of protecting against sputtering damage.

4. Conclusion

In summary, we reported on the enhancement of a top emission organic light-emitting diode with a double B.L. The TEOLED with double B.L. showed much higher luminance and lower leakage current than that with a single organic B.L. or inorganic B.L. A fabricated device having a double B.L. is observed to have much longer lifetime than other devices with single buffer layers, at a constant current density of 10 mA/cm^2. The double B.L. is suitable for protecting TEOLEDs from sputtering damage.

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