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The feasible application of low-cost Al/Cu bimetal semitransparent cathode in top-emitting organic light-emitting diode

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Efficient top-emitting organic light-emitting diode (TOLED) using low-cost Al/Cu semitransparent cathode was demonstrated. With tris(8-hydroquinoline) aluminum as emitting and electron-transport layer, the maximum luminous efficiency reached 5.1 cd/A which is almost equal to that (5.5 cd/A) of TOLED with conventional Al/Ag semitransparent cathode. Moreover, the TOLED with Al/Cu semitransparent cathode exhibited weak microcavity effect, owing to the low reflectance of Al/Cu and therein reduced multiple-beam interference within the device. In comparison with Al/Ag bimetal semitransparent cathode, Al/Cu showed rather flat transmittance spectrum and caused less leakage current, which would further improve device performance.

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1. Introduction

Organic light-emitting diodes (OLEDs) have been attracted increasing research interest due to their superior opto-electrical characteristics such as high luminance, low driving voltage, wide viewing angle and good contrast. Top-emitting OLED (TOLED) in which the light is squeezed out of the top electrode (mostly cathode) is preferable to high-resolution full-color displays in comparison with the conventional bottom-emitting OLED (BOLED), because the fabrication process is in good compatibility with modern Si technology and the aperture ratio is not affected by the backplanes with complicated circuits. It is well established that the properties of semitransparent cathode play a key role in TOLED. Many attempts have been made to develop proper semitransparent cathode [1-17]. A direct and simple method of constructing semitransparent cathode is thinning the conventional metal electrode. However, this leads to instability and high sheet resistance. Covering a highly transparent indium-tin-oxide (ITO) such as Ca/ITO [1], Al/ITO [2] and Mg:Ag/ITO [1,3] can circumvent this problem. Similar attempts of using metal-doped organic conducting buffer layer covered with ITO (e.g., [BPhen:Li]/ITO [4] and [BCP:Li]/ITO [5]) as semitransparent cathode are also demonstrated. Unfortunately, the intense radiation energy created by sputtering of ITO would damage the underlying organic stacks even with protective buffer layer, which deteriorates device performance. Another semitransparent cathode structure is cermet such as Al:SiO [6]. However, the transmittance and conductivity of Al:SiO film are very sensitive to Al content. The fabrication process is difficult to control in the practical application. Recently, the intensive investigation has been focused on multimetal semitransparent cathode systems such as Ca/Ag [7,8], Ca/Mg [9], Sm/Ag [10,11], Sm/Au [12], Yb/Au [13], Ba/Au [14], Al/Ag [15,16] and Cs/Al/Au [17], owing to their superior conductivity and no damage to the underlying organic layers during the thermal evaporation. In this article, we report an effective and low-cost Al/Cu bimetal semitransparent cathode. A comparative study of TOLEDs using Al/Cu and Al/Ag semitransparent cathodes is also investigated, since Al/Ag is the most widely used semitransparent cathode in TOLED.

2. Experimental details

TOLEDs with structure (Fig. 1) of Glass/ Ag (80 nm)/ MoO_x (2 nm)/ NPB (58 nm)/ Alq_3 (50 nm)/ LiF (0.3 nm)/ Al (3 nm)/ Cu [or Ag] (18 nm) were fabricated in a multi-source organic deposition system under a vacuum of 3×10^{-4} Pa. Here, MoO_x deposited onto thick Ag anode was served as a hole-injection layer. NPB denotes *N*,*N'*-bis(naphthalen-1-yl)-*N*,*N'*-bis(phenyl) benzidine as a hole-transport layer, and Alq_3 denotes tris(8-hydroquinoline) aluminum as an emitting and electron-transport layer. The light was outcoupled through a semitransparent cathode of LiF/Al/Cu (Device **A**) [or LiF/Al/Ag (Device **B**)].

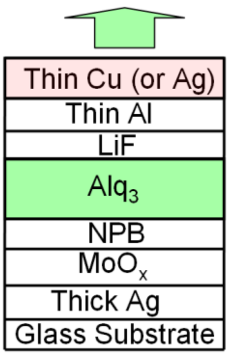

Fig. 1 Schematic structure of TOLED with Al/Cu (or Al/Ag) semitransparent cathode.

The layer thickness of deposited materials was monitored *in situ* using an oscillating quartz thickness monitor. The deposition rate for organic materials was ~1 Å/s. The active area of the devices was 0.25 cm^2. The current-voltage-luminance (*I-V-L*) characteristics were measured using a Keithley 2400 Source Meter and a Minolta LS-110 Luminance Meter. The electroluminescence (EL) spectra were measured using a PR-650 Spectra Scan.

3. Results and discussion

Fig. 2 shows the luminous efficiency-current density (η_J-J) and power efficiency-current density (η_P-J) (assuming Lambertian distribution) characteristics of Device **A** and **B**. It can be seen that the luminous efficiencies of Device **A** and **B** are almost equally high, the corresponding maximum luminous efficiencies reach 5.1 cd/A and 5.5 cd/A, respectively. This indicates that the bilayer Al/Cu semitransparent cathode is highly effective for squeezing light out of TOLED. On the other hand, the maximum power efficiency (2.5 lm/W) of Device **A** is a little lower than that (2.8 lm/W) of Device **B**. This *may* be resulted from the fact that the electrical resistivity (16.78 nΩ·m) of Cu is a little higher than that (15.87 nΩ·m) of Ag [18], which leads to higher sheet resistance of Al/Cu semitransparent cathode and thus reduced power efficiency in Device **A**. The *J-V-L* characteristics (Fig. 3) also elucidates that Device **A** shows higher driving voltage at the same current density, and at the same voltage it exhibits lower luminance.

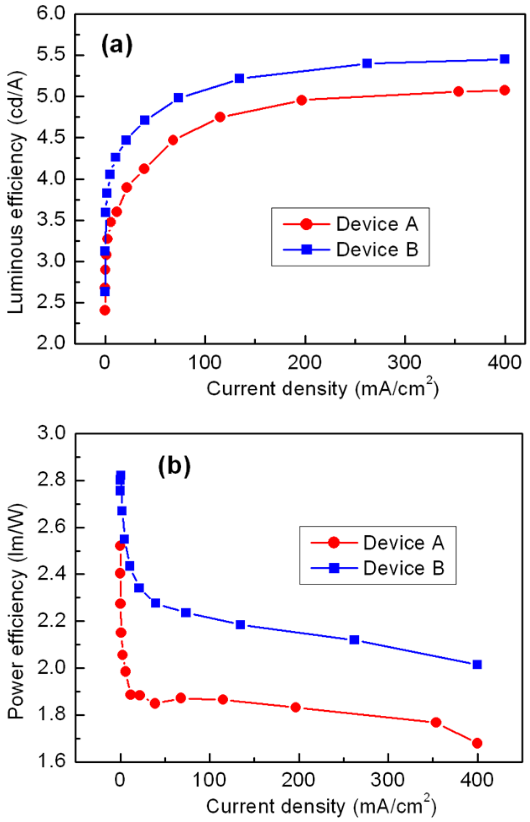

*Fig.2 (a)η_J-J and (b) η_P-J characteristics of Device **A** and **B***

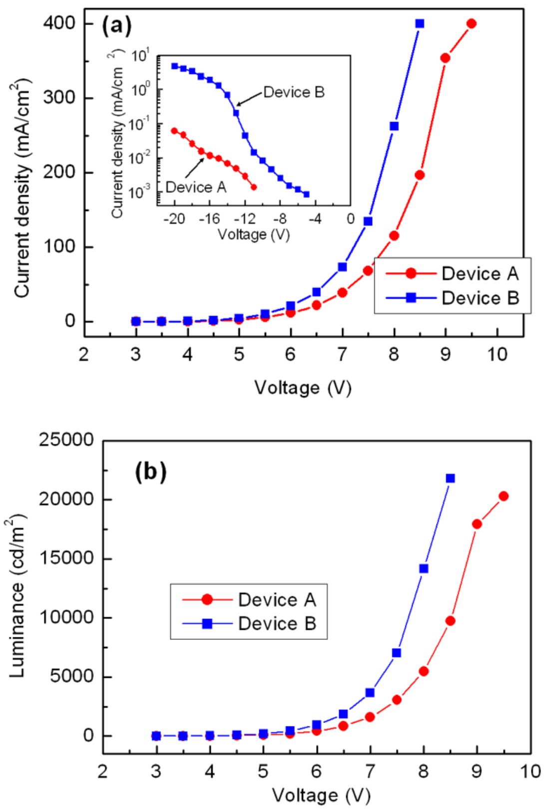

*Fig.3 (a) J-V and (b) L-V characteristics of Device **A** and **B**. Inset: Reverse J-V characteristics*

It is widely acknowledged that the transmittance and reflectance of the semitransparent cathode play a crucial role in TOLED. Taking into account that the semitransparent cathode of Al/Cu (or Al/Ag) is adjacent to Alq_3, the transmittance measurement is performed using Glass/Alq_3/Al/Cu (or Ag) multi-layer structure and only the normal incident is under consideration. The transmittance results are calibrated by eliminating the effect of the glass substrate. Fig. 4 shows the measured and calculated transmittances of the semitransparent cathodes of Al (3 nm)/Cu (18 nm) and Al (3 nm)/Ag (18 nm). It is obvious that Al/Cu and Al/Ag show approximate transmittance in the visible region. However, the transmittance of Al/Ag decreases monotonically with the increase of wavelength due to the increasing extinction coefficient of Ag [19], while the transmittance of Al/Cu is rather flat in the visible region. The flat transmittance spectrum of semitransparent cathode is of great importance for practical application, because the RGB (red, green and blue) primary colors can be equally squeezed. This is one characteristic of Al/Cu takes advantage over Al/Ag as semitransparent cathode. On the other hand, from the comparison of the calculated transmittance to the measured ones, the measured data are systematically higher than the calculated values. Such difference is perhaps due to the difference between the refractive indices used in the calculations and the real values for our experiments. Here, the calculations are carried out by using a transfer matrix method [11], and the complex refractive indices for the calculations are derived from Ref. [19].

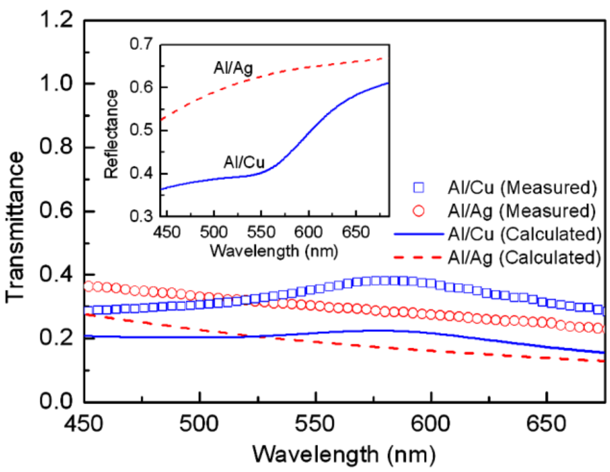

Fig. 4 Measured (symbols) and calculated (lines) transmittances of the semitransparent cathodes of Al (3 nm)/Cu (18 nm) and Al (3 nm)/Ag (18 nm). Inset: Calculated reflectances of semitransparent cathodes of Al/Cu and Al/Ag.

Fig. 5(a) and (b) show the measured EL spectra (normalized) for Device **A** and **B** at viewing angles of 0°, 30° and 60° off the surface normal. It can be seen that both TOLEDs show blueshift with increasing viewing angles due to the microcavity effect. The polar plots of emission intensities (normalized to the 0° intensities) for Device **A** and **B** comparing with the Lambertian emitter are depicted in Fig. 5(c). It is clear that both TOLEDs exhibit sub-Lambertian distribution with enhanced intensity in the forward direction and suppressed intensity at large viewing angles. This phenomenon is more obvious in Device **B** compared with Device **A**, indicating a stronger microcavity effect occurred within Device **B**. This can be ascribed to the following two factors: (i) the reflectance (39% at 540 nm) of Al/Cu is much lower than that (62% at 540 nm) of Al/Ag [inset in Fig. 4], and (ii) the transmittance (21%) of Al/Cu is slightly higher than that (19%) of Al/Ag at 540 nm [Fig. 4]. These intrinsic properties of Al/Cu semitransparent cathode contribute to less multiple-beam interference within Ag||Al/Cu microcavity (Device **A**) as compared with Ag||Al/Ag microcavity (Device **B**). Consequently Device **A** exhibits weak microcavity effect and thus wide output EL spectra. The full-width at half-maximum (FWHM) of 0º EL spectrum of Device **A** is 93 nm (close to the conventional BOLED, not shown) [Fig. 5(a)] which is much wider than that (59 nm) of Device **B** [Fig. 5(b)]. The wide EL spectrum is very useful for white TOLEDs, because one of major problems of fabricating white TOLEDs is their narrow output EL spectra as a result of strong microcavity effect [10,16]. Al/Cu semitransparent cathode may provide an alternative route to circumvent this problem and would improve device performance.

To demonstrate the advantage of Al/Cu bimetal semitransparent cathode in constructing white TOLED. We fabricated a white TOLED (Device **C**) with structure of Glass/ Cu (100 nm)/ MoO_x (2 nm)/ [NPB:rubrene] (45 nm)/ BPhen (30 nm)/ Alq_3 (15 nm)/ LiF (0.3 nm)/ Al (3 nm)/ Cu (18 nm) and a BOLED (Device **D**) with structure of Glass/ ITO/ MoO_x (2 nm)/ [NPB:rubrene] (45 nm)/ BPhen (30 nm)/ Alq_3 (15 nm)/ LiF (0.3 nm)/ Al (2 nm)/ Cu (100 nm). Here, [NPB:rubrene] served as the white-emitting media [20], rubrene = 5, 6, 11, 12-tetraphenylnaphthacene, and BPhen = 4,7-diphyenyl-1, 10-phenanthroline. The normalized EL spectra of both devices are depicted in Fig. 6 which clearly shows that Device **C** exhibits almost the same EL characteristics as Device **D**. For instance, Device **C** and **D** show a bluish emission from NPB (~460 nm) and a reddish emission from rubrene which combine a white emission with Commission Internationale d'Eclairage (CIE) color coordinates of (0.29, 0.30) for Device **C** and (0.30, 0.30) for Device **D**. This sufficiently indicates that the Al/Cu bimetal semitransparent cathode can effectively squeeze the light out of the cavity and give a broad emission, suggesting a promising easy-to-do method to fabricate white TOLED. In addition, Cu is one of the cheapest metals, which is well suited for low-cost fabrication of OLED displays.

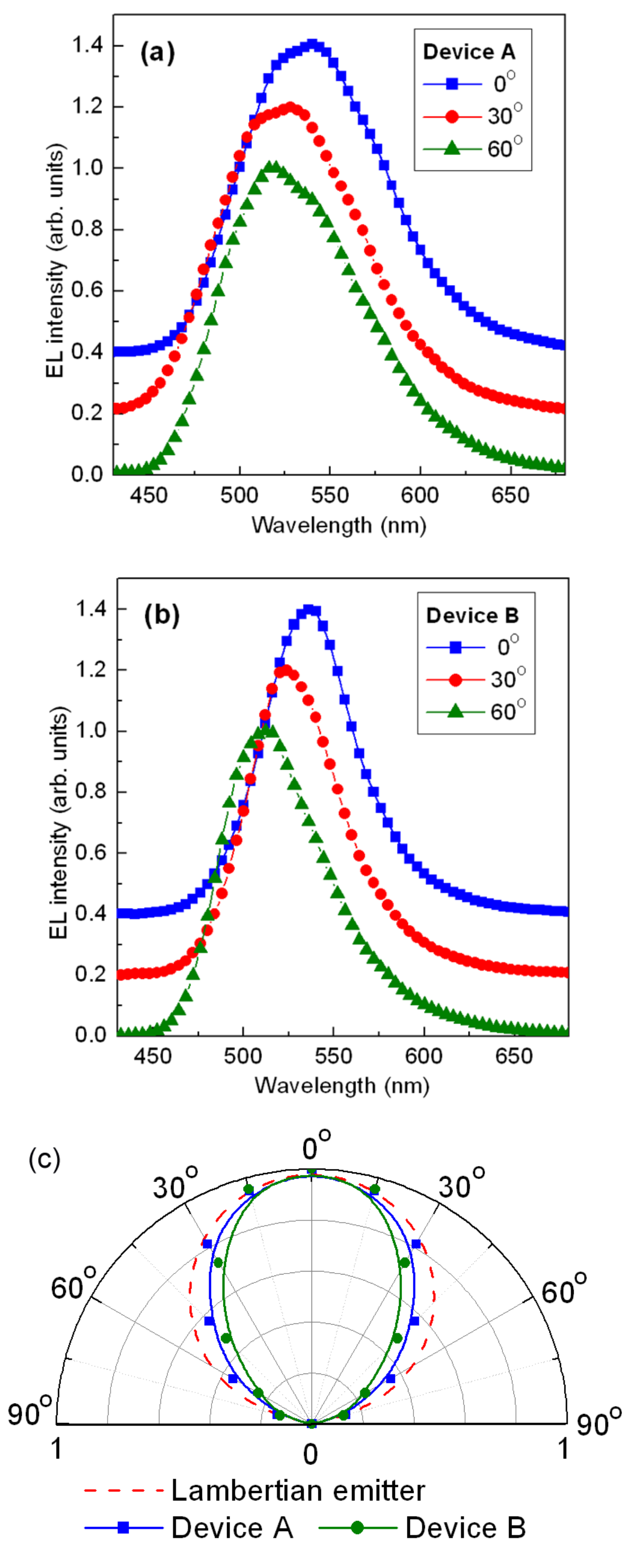

*Fig. 5 Normalized EL spectra at viewing angles of 0º, 30º and 60º off the surface normal of (a) Device **A** and (b) Device **B**; (c) Polar plots of EL intensities (normalized to the 0º intensities)*

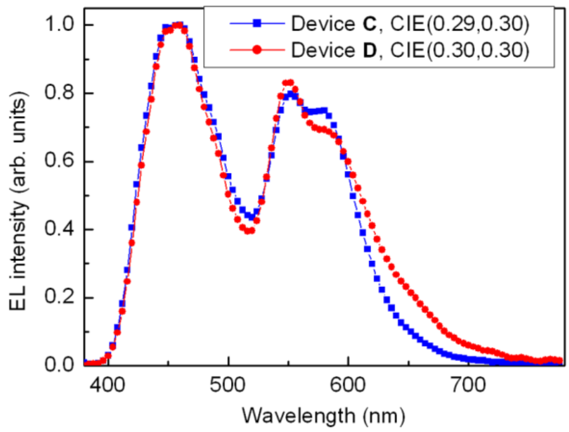

*Fig. 6 The normalized EL spectra and CIE color coordinates of Device **C** and **D**.*

The reverse *J-V* characteristics of Device **A** and **B** are depicted in the inset in Fig. 3(a) which clearly shows that the reverse current density of Device **B** is much higher than Device **A**, suggesting less leakage current in Device **A**. For example, at the reverse voltage of -20 V, the reverse current density (4.9 mA/cm^2) of Device **B** is about two orders of magnitude higher than that (0.061 mA/cm^2) of Device **A**. This maybe resulted from the diffusion of Ag into organic layers which results in higher leakage current [21]. Therefore we *would* like to make a conclusion that the TOLED with Al/Cu semitransparent cathode can improve device durability and thereby promoting operating life-time.

4. Conclusions

Al/Cu semitransparent cathode with flat transmittance spectrum and low-cost features was proven to be highly efficient for squeezing light out of TOLED and reducing leakage current. The maximum luminous efficiency of TOLED with Al/Cu semitransparent cathode reached 5.1 cd/A which is almost equal to that (5.5 cd/A) of device with Al/Ag semitransparent cathode. Moreover, the TOLED using Al/Cu semitransparent cathode showed wide EL spectra and weak microcavity effect as a result of low reflectance of Al/Cu, which is beneficial to the fabrication of white TOLED.

Acknowledgements

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