Research article

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Highly transparent and conductive metal oxide/metal/polymer composite electrodes for high-efficiency flexible organic light-emitting devices

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Abstract: Ultrathin metal films emerge as an innovative category of transparent electrodes in recent decades, holding great promises enabling the next-generation flexible organic light-emitting devices (OLEDs). Although metal thin films with polymer nucleation inducers have been extensively studied in OLEDs, satisfying the requirements of both superior optoelectrical and high optical outcoupling characteristics is still challenging. Here, we

demonstrate a metal oxide/ultrathin Ag/polymer (MAP) composite electrode with low sheet resistance of 15.1 Ω /sq, high transmittance of 87.4% at 550 nm, and smooth morphology with surface roughness of 0.768 nm. Besides, the composite electrodes significantly enhance the out-coupling of the light trapped in OLEDs due to the relatively high-refractive index polymer. Flexible OLEDs with the MAP anodes exhibit over 2.3 times enhancement in efficiency to that of indium tin oxide (ITO)-based OLEDs. The flexible OLEDs can survive 1000 bending cycles at a bending radius of 8 mm with negligible decrease in electroluminescent performance.

Keywords: flexible; high efficiency; MAP composite electrodes; organic light-emitting devices.

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1 Introduction

Organic light-emitting devices (OLEDs) have gained enormous interests driven by the desire of low-cost flat panel displays and light sources [1–6]. Although OLEDs have shown continuous success in consumer electronics, the absence of ideal transparent conductive electrodes (TCEs) is still the largest hurdle for realizing next-generation flexible and printable OLEDs [7–9]. Indium tin oxide (ITO) dominates the common use of TCEs in optoelectronic devices based on rigid glass substrates [10]. However, the use of ITO electrode in flexible devices is limited by its brittle nature [11, 12]. In addition, waveguide mode associated with the ITO anode due to its high refractive index is one of the major power losses in OLEDs [13–15]. Research efforts have devoted into the exploration of new TCE materials, boosting the development of metal nanowires [16–19], conducting polymers [20, 21], graphene [22–24], carbon nanotubes [25, 26], and hybrid electrodes [27, 28]. Despite the prominent progress made in the past decades, further enhancement in the performance of the TCEs is still needed for efficient optoelectronic devices.

The development of ultrathin metal electrodes has been considered as a practical technology to fulfill large-area and high-performance TCEs [29–31]. Ultrathin metal electrodes with thickness smaller than 10 nm allow for high visible light transmission, and mechanical flexibility [32, 33]. However, the thermal evaporation of the metals on substrates follows the metal-island growth process and further develops into discrete surface aggregations for the metal film with the thickness of less than 10 nm, which limits their conductivity [34]. Polymer-metal hybrid transparent electrodes have been reported as effective methods to circumvent the problem with polymer nucleation inducers [9, 35]. However, high-efficiency OLEDs need high-refractive-index optical coupling layers to reduce the total internal reflection in the device, while common polymer inducers unfortunately feature a low refractive index ($n \leq 1.6$). OLEDs efficiency built upon such ultrathin metal anode stacks is still limited by the light losses in devices [31]. Unlocking the full potential of OLEDs based on the ultrathin metal electrodes is still a challenge.

In this contribution, we demonstrate a multifunctional anode stacks, consisting of a relatively high-index polymer optical coupling layer, electrically conductive silver layer and hole-injection MoO_3 layer to collectively achieve high-efficiency OLEDs. The electrode stack consists of composite layers of metal oxide/ultrathin Ag/polymer (MAP), featuring low sheet resistance of $15.1 \text{ } \Omega/\text{sq}$, high transmittance of 87.4% at 550 nm and smooth morphology with surface roughness of 0.768 nm. Polyamide-imide (PAI) features the highest refractive index (~ 1.7) among the reported polymer wetting inducers for ultrathin Ag electrodes, which matches the refractive index value of organic materials in OLEDs and further improves the device performance as optical out-coupling layer with microcavity effect. The composite electrode has been applied to the OLEDs based on rigid glass or flexible industrial-grade polyethylene naphthalate (I-PEN) substrate, and enhanced electroluminescent (EL) performance has been obtained. The employment of such composite anode substituting for ITO eliminates the power loss to waveguide mode. The incorporation of flexible I-PEN substrate with built-in light scattering effect further extracts the trapped light from substrate mode. An optimized composite anode leads to flexible OLEDs with a current efficiency (CE) of 98.8 cd/A , which corresponds to 2.3 times enhancement in efficiency to that of the ITO-based OLEDs. The flexible OLEDs can survive 1000 bending cycles at a bending radius of 8 mm with negligible decrease in EL performance. Furthermore, the I-PEN based OLEDs exhibit improved viewing characteristics, which is desirable for practical applications.

2 Results and discussion

It is known that 3D Volmer–Weber growth mode dominates the film formation process for the evaporated Ag film, and it tends to form unconnected islands and thus be electrically insulating under an ultrathin thickness of a few nanometers [35, 36]. In addition, the isolated Ag islands scatter incident light and appear opaque in certain degree (Figure 1a). We introduce a polymer, polyamide-imide [poly (trimellitic anhydride chloride-co-4,4'-methylenedianiline)] (PAI, Figure S1) between the glass substrate and the Ag film as a surface modification layer to change the growth mode of the Ag on the glass substrate. A 9 nm Ag film evaporated onto the PAI-coated glass substrate shows a continuous and smooth surface as illustrated in Figure 1b. The amino groups within the PAI polymer chain play important roles in the formation of continuous Ag films [9, 35]. In particular, the evaporated Ag atoms form coordinated bonds with the functional groups along the PAI polymer chains (inset in Figure 1b), which sufficiently suppresses the random migration of silver atoms to form isolated aggregates and results in the growth of high-quality continuous ultrathin Ag film. The surface morphologies of the 9 nm Ag on the PAI-coated glass or bare glass are investigated and shown in Figure 1d–g to evaluate the function of the PAI on the growth of the Ag film. The Ag on bare glass shows a discontinuous granular morphology with a surface roughness of $R_a = 3.06 \text{ nm}$, while the Ag on the PAI-coated glass electrode represents a continuous and smooth morphology with roughness value of $R_a = 0.768 \text{ nm}$. The introduction of the PAI enables a conductive Ag film with thickness as low as 5 nm (Figure 1h). Especially, the 9 nm Ag deposited on the PAI-coated glass exhibits a low sheet resistance of $15.1 \text{ } \Omega/\text{sq}$, in contrast to non-conductivity for the 9 nm Ag on bare glass (Figure 1h). The transmittance of the Ag/PAI electrode ($T = 73.6\%$ at 550 nm, measured by substrate base) over the entire visible range is much higher than Ag electrode with an opaque feature ($T = 44.3\%$ at 550 nm, measured by substrate base) (Figures 1i and S3). Besides, the 9-nm Ag-PAI film also features superior sheet resistance and optical transmittance than the conductive 11-nm Ag film ($211.32 \text{ } \Omega/\text{sq}$, 64.93% @550 nm, measured by substrate base). To further improve the transmittance of the Ag/PAI electrode, an anti-reflection layer (MoO_3) with a refractive index of 2.06 (Figure S2) was then introduced onto the top surface of the silver film to form the more transparent MAP composite electrode with the structure of $\text{MoO}_3/\text{ultrathin Ag/PAI}$. Figure 1c shows that incident light phase shift happens at the interfaces which further improves light transmission by weakening the reflectance of Ag film. As a result,

the transmittance is improved to 87.4% at 550 nm measured by substrate base (Figure 1i).

To investigate the performance of the MAP electrodes as effective alternatives of ITO electrodes, MAP-based OLEDs with green emissive layers were fabricated and shown in Figure 2a. PAI layers underneath the ultrathin Ag layer in the MAP stacks plays important roles in not only surface modification of the substrates but also providing fine adjustment to the microcavity effect. The light out-coupling efficiency can be enhanced without altering the device structure and the charge carrier balance within the device. The outcoupling enhancement of the device structure with the MAP electrodes was calculated based on the dipole emission model and the transfer matrix method by the Setfos software [4, 15]. Figure 2b presents the enhancement ratios of the outcoupled luminance as a function of the PAI thickness, which indicates a good agreement with the experimental measurements. The optimized thickness of the PAI is around 110 nm. For comparison, we also fabricated OLEDs with MoO_3 -modified ITO anodes as reference devices. Figures 2c and d demonstrates the current density–voltage–luminance (J–V–L) characteristics and CE of the optimized MAP-based device (MoO_3 (5 nm)/Ag (9 nm)/PAI (110 nm) on glass) in comparison with the ITO-based device. The OLED with MAP electrode shows a maximum luminance as high as $115,500 \text{ cd/cm}^2$ at 10 V and a CE of 82.3 cd/A , which corresponds to an enhancement factor of 1.9 in the efficiency to the ITO-based device (42.8 cd/A). The emission pattern

evolutions of the MAP-based OLEDs and ITO-based OLEDs over the observation angles were further investigated and shown in Figures 2e, f and S5. It was found that the optical microcavity had formed between the silver electrodes. Figure S6 shows the calculated optical density at normal emission angle with different PAI thickness, which can represent the optical microcavity effect in the device structures [37]. The microcavity effect due to the employment of metallic electrodes results in the emission enhancement influenced by the thickness of PAI. When the resonant cavity length of the device matches the emission peak wavelength, the enhancement effect will reach the maximum. In our contribution, the devices with 110 nm–130 nm PAI show the relatively high peak intensity and narrow full width at half maximum (FWHM) determined by the microcavity resonance. Although the optical microcavity is beneficial to improve the efficiency of OLEDs, it has a relatively narrow bandwidth and exhibits a blue shift when the emission angle increases. The angle-dependent EL emission is a shortage for the practical applications of the OLEDs.

I-PEN substrates (refractive index, ca. 1.7) combined with built-in scattering particles (diameter 300 nm, Figure S7) has been employed as cost-effective flexible substrates for the OLEDs. The I-PEN substrates can extract substrate-confined modes and improve the viewing characteristics in OLEDs [38]. Figures 3a and b show the schematic flexible electrode and corresponding surface morphology based on the I-PEN substrates. The PAI

Figure 1: (a)–(b) Schematic illustration of the ultrathin Ag film without and with a polyamide-imide (PAI) layer on glass substrates. Inset of b: Coordination bond between PAI and evaporated Ag. (c) Conceptual diagram of the incident light phase shift in the Ag film with the anti-reflective layer of MoO_3 . (d)–(g) Surface morphology images of the Ag/glass and Ag/PAI/glass taken using scanning electron microscopy (SEM) and atomic force microscopy (AFM), respectively. (h) Evolution in sheet resistance of the Ag, Ag/PAI electrodes with different Ag thickness. (i) Optical transmittance and corresponding photographs of the Ag, Ag/PAI and metal oxide/ultrathin Ag/polymer (MAP) electrodes.

Figure 2: (a) Device structure of the Ir(ppy)₃-based organic light-emitting devices (OLEDs). (b) Calculated and experimental values of luminance normalized to its peak value as a function of polyamide-imide (PAI) thickness. Theoretical simulation results were obtained from the Setfos software. (c)–(d) Comparison of current density–voltage–luminance (J–V–L) and current efficiency (CE)–L characteristics of OLEDs with MoO₃/indium tin oxide (ITO) and metal oxide/ultrathin Ag/polymer (MAP) anodes. (e)–(f) Angular dependence of electroluminescence (EL) intensity.

coating supplies a smooth morphology and reduces the roughness of I-PEN as shown in Figure S8. Figure 3c presents the total transmittance spectra of I-PEN in comparison with glass substrate. Actually, I-PEN shows optical haze with ca. 20–30% measured for 130- μ m-thick I-PEN substrate (Figure S9). Besides, the MAP electrode on I-PEN exhibits superior mechanical robustness under bending strains as shown in Figures 3d–f. The MAP electrode can be bent to a radius curvature of 1 mm without resistance increase (Figure 3e). Furthermore, Figure 3f demonstrates the resistance evolution of our MAP electrode during

continuous 1000 cycles with bending radius of 1 mm, which maintains a value between 14 and 14.5 Ω /sq.

Figures 4a and b present the J–V–L and CE–L characteristics of MAP-based flexible OLED prepared on the I-PEN substrate. The flexible OLED on the I-PEN substrate can reach CE as high as 98.8 cd/A and maintain nearly 90 cd/A at 1000 cd/cm², exhibiting a 20% enhancement compared to the MAP-based OLED on the glass substrate. To clarify the origin of the improvement, we simulated an optical analysis by the method introduced [37]. Figure 4c presents the power dissipation spectra conducted without scattering effect;

Figure 3: (a) Schematic illustration of the metal oxide/ultrathin Ag/polymer (MAP) electrode fabricated on flexible industrial-grade polyethylene naphthalate (I-PEN). (b) Surface morphology image of the MAP/I-PEN electrodes with 9 nm Ag. (c) Measured total transmittance spectra and photographs of the I-PEN substrates compared with glass substrates. (d)–(e) Optical images of the MAP/I-PEN electrode. The bending radius in (e) is 1 mm. (f) Mechanical robustness measurement of the flexible MAP/I-PEN electrode with 1000 times bending cycles.

Figure 4: (a)–(b) Device performance of the organic light-emitting devices (OLEDs) prepared on the metal oxide/ultrathin Ag/polymer (MAP)/industrial-grade polyethylene naphthalate (I-PEN) electrode. Inset: Photographs of lit OLEDs with MAP/I-PEN electrodes before and after bending. (c) Power dissipation spectra without consideration of scattering effect versus normalized in-plane wavevector u and wavelength of the OLEDs with MAP/I-PEN electrodes. The dipole emitter orientation was treated as isotropic. Regions 1–4 represent air mode, substrate mode, waveguide mode, and surface plasmon polaritons (SPPs) mode, respectively. A combination of region

1–2 corresponds to the total light emitted to a substrate when it is regarded as infinitely thick. (d)–(e) Electroluminescent (EL) intensity as a function of emission angle of the MAP/I-PEN based devices. (f) Normalized luminance and current efficiency versus bending cycles with 8 mm bending radius for the flexible OLEDs.

meanwhile, the thickness of the substrate was treated as infinite. The optical power distributed in the OLEDs can be divided into four regions, corresponding to outcoupled (1), substrate-confined (2), waveguided (3), and surface plasmon polariton (4) modes, respectively [38]. It should be noticed that the built-in scattering particles can effectively extract the substrate-confined mode in region 2, which contributes largely to the total fractions of outcoupled optical power. There are almost no perceived shifts in the EL spectra along with the observation angles, and the angular emission pattern is even widened than that of the Lambertian emission for the OLED fabricated on I-PEN as shown in Figures 4d and e. Moreover, bending test has been carried out to evaluate the flexibility of the I-PEN device. As shown in the insets of Figure 4b, there is no visually EL degradation even with the bending curvature radius of 2 mm. The mechanical durability of such flexible OLED was further investigated with EL performance after repeated bending cycles at a bending radius of 8 mm (Figures 4f and S10). After 1000 bending cycles, no obvious decrease occurred in luminance and efficiency of the flexible OLED based on MAP electrode, indicating the robustness of each component.

3 Conclusion

In conclusion, we have explored an ultrasmooth MAP electrode consisting of relatively high-refractive index polymer of PAI as nucleation inducer and optical out-coupling layer, ultrathin Ag as electrically conductive layer, and MoO_3 as an anti-reflective and hole-injection layer, which can function as a desirable alternative of the traditional transparent ITO electrode in OLED. The corresponding flexible MAP-based OLED on the I-PEN substrate with built-in scattering particles exhibits a

maximum CE of 98.8 cd/A, which is 2.3 times enhancement in efficiency compared to the ITO/glass-based OLED, manifesting the effectiveness of the MAP composite electrode. The flexible OLED is mechanically robust and maintain its high performance under the bending radius of 8 mm after 1000 continuous cycles. The MAP electrodes combined with the I-PEN substrates have great potential for the simple and cost-effective production of flexible OLEDs, paving the way for next-generation commercially flexible and printable displays and solid-state lighting.

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