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# Ultrathin and ultrasmooth Au films as transparent electrodes in ITO-free organic light-emitting devices†

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An ultrathin, ultrasmooth and flexible Au film as an alternative of the indium-tin oxide (ITO) electrode in organic light-emitting devices (OLEDs) has been reported. The 7 nm Au film shows excellent surface morphology, optical and electronic characteristics including a root-mean-square roughness of 0.35 nm, a high transparency of 72% at 550 nm, and a sheet resistance of 23.75  $\Omega$  sq<sup>-1</sup>. These features arise from the surface modification of the glass substrate by using a SU-8 film, which fixes metal atoms *via* chemical bond interactions between Au and SU-8 film to suppress the island growth mode. A 17% enhancement in current efficiency has been obtained from the OLEDs based on the ultrathin Au electrodes compared to that of the devices with the ITO electrodes. The OLEDs with the ultrathin Au/SU-8 anodes exhibit high flexibility and mechanical robustness.

# 1 Introduction

Organic light-emitting devices (OLEDs) are being broadly investigated owing to their advantages of light weight, low cost, material abundance, low power consumption and flexibility, as well as their potential applications for highly efficient large-area solid-state lighting and full-color flat-panel display.<sup>1–5</sup> Currently, indium–tin oxide (ITO) is the commonly used transparent electrode material in OLEDs.<sup>6,7</sup> However, the ITO electrode faces several challenges, particularly, for the fast and low-cost roll-to-roll process of light-weight and flexible devices on plastic substrates.<sup>8,9</sup> The ITO film is brittle and tends to form cracks in flexible devices, and the sputtering technology with high annealing temperature of ITO is incompatible with the plastic substrates.<sup>10–12</sup> Indium atoms in the ITO electrode may migrate into the organic active layer causing degradation of the device performance.<sup>13</sup> Furthermore, the waveguide mode in the ITO anode/organic active layer arising from the high refractive index of ITO is one of the main power losses in OLEDs.<sup>14,15</sup> To meet this challenge, new kinds of transparent electrodes have been developed to replace the ITO film, such as carbon nanotubes,<sup>16,17</sup> graphene,<sup>18–20</sup> new transparent conducting oxides,<sup>21,22</sup> metal nanowires,<sup>23,24</sup> conducting polymers,<sup>25,26</sup> patterned metal grids,<sup>27,28</sup> and ultrathin metallic films.<sup>29,30</sup>

An ultrathin and continuous metallic film with mechanical robustness, high electrical conductivity and high optical transparency has been considered as the ideal alternative of the ITO electrode.<sup>31,32</sup> Although a high optical transparency can be obtained for an ultrathin metallic film with a thickness of less than 10 nm, the thermally evaporated metallic layer follows the Volmer-Weber growth mode, and results in a non-continuous film in the long-region with a rough surface morphology.30,33 The poor continuity and rough morphology directly deteriorate its electrical conductivity. Employing composite structures is a common method to overcome this problem. A "nucleation-inducing seed layer" between the substrate and the metallic film, such as metal seeds,<sup>8</sup> metal oxide<sup>6,34</sup> and organic small molecules,<sup>35,36</sup> has been employed to realize the ultrathin and ultrasmooth metallic electrode.37 Co-depositing a small amount of Al into Ag without a seed layer has also been reported to realize an ultrathin electrode.<sup>30</sup> So far, developing a simple and effective method to obtain a single-material-based ultrathin metallic film with a continuous and smooth morphology to guarantee its high conductivity is still a challenge.

In this paper, we report a simple and low-cost method to realize an ultrathin Au film with an ultrasmooth and continuous morphology. As a replacement for ITO in OLEDs, higher device efficiency and nearly Lambertian emission characteristics have been obtained. A modification layer of SU-8 was spin-coated onto the glass substrate prior to Au thermal deposition, which fixes metal droplets *via* chemical bond interactions between Au atoms and the SU-8 film. The 7 nm Au film shows an excellent surface morphology with a root-mean-



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square (RMS) roughness value of 0.35 nm, favourable optical properties with a transparency of 72% at the wavelength of 550 nm, and good conductivity with a sheet resistance of 23.75  $\Omega$  sq<sup>-1</sup>. Application of the ultrathin Au anode in OLEDs leads to an enhancement of 17% in current efficiency compared to the OLEDs with the traditional ITO anode. A bending test has been conducted on the flexible OLED with the ultrathin anode engaging a SU-8 film as the substrate, demonstrating its high mechanical robustness and flexibility.

# 2 Experimental details

#### 2.1 Preparation of the ultrathin Au anode

The glass substrates were cleaned using acetone, alcohol and ultrapure water in sequence. A photoresist film (SU-8 2025, MicroChem Corp.) was used as the surface modification layer. The photoresist was diluted with cyclopentanone to 40 mg mL<sup>-1</sup> and then was spin-coated onto the substrate for 30 s at 4000 rpm. The thickness of the SU-8 film is about 100 nm. The glass substrates modified with the SU-8 layer were transferred into a thermal evaporation chamber. A pre-cleaned glass substrate without the SU-8 layer was also transferred into the chamber as the reference sample. An ultrathin Au film was then deposited on the substrates at a base pressure of  $5 \times 10^{-4}$ Pa with a rate of 0.5 Å s<sup>-1</sup>. The deposition rate and the film thickness of the deposited material were monitored by a quartz crystal oscillator. The surface morphologies of the Au film have been detected by using an atomic force microscope (AFM, Dimension Icon, Bruker) in tapping mode and a scanning electron microscope (SEM, JSM-7500F, JEOL). A fourpoint probe system was used to measure the sheet resistance of the Au film. The transmittance spectra were recorded by using a UV-Vis spectrophotometer (UV-2550, SHIMADZU).

#### 2.2 Fabrication and characterization of OLEDs

The OLEDs with the ITO anode or ultrathin Au anode were both fabricated by the physical vapor deposition process. The glass substrates covered with ITO electrodes were cleaned using standard procedure. A 5 nm MoO<sub>3</sub> film was used as an anodic buffer layer. N,N'-Diphenyl-N,N'-bis(1,1'-biphenyl)-4,4'diamine (NPB) and 2,2',2"(1,3,5-benzenetriyl)tris-[1-phenyl-1Hbenzimidazole] (TPBi) were the hole-transport and hole-blocking layers, respectively. The green phosphorescent emitter tris-(2-phenylpyridine)iridium  $(Ir(ppy)_3)$  with the ratio of 6 wt% was co-deposited with the host material m-bis(N-carbazolyl) benzene (mCP) as the emitting layer. Ca/Ag was the bilayer cathode. Each layer of the devices was deposited by thermal evaporation with a shadow mask in the chamber under the base pressure of  $5 \times 10^{-4}$  Pa. The stack structure of the OLED is glass substrate/anode (ultrathin Au or ITO)/MoO<sub>3</sub> (5 nm)/ NPB (40 nm)/mCP:Ir(ppy)<sub>3</sub> (6 wt%, 20 nm)/TPBi (35 nm)/ Ca (2 nm)/Ag (100 nm). The intersection of the anode and cathode decided an active device area of 4 mm<sup>2</sup>. The electroluminescence (EL) performance of the devices, such as luminance, current density and EL spectra, was measured by a

Photo Research PR-655 spectrophotometer and a Keithley 2400 programmable voltage–current source. All of the measurements were performed in air at room temperature.

#### 2.3 Fabrication of the flexible OLEDs

Octadecyltrichlorosilane (OTS) modification was carried out on the glass substrate for about 4 h by a vapor-deposition method to realize a hydrophobic surface, so that the SU-8 film can be peeled off from the substrate completely. A droplet of SU-8 2025 without dilution was spin-coated on the glass substrate for 60 s at 2000 rpm. Then the substrate covered SU-8 film was exposed to a UV light source for solidification at power of 125 W for 4 min. After depositing the ultrathin Au anode, organic layers and the cathode of the OLEDs on the solidified SU-8 film, the flexible OLED was obtained by peeling off the solidified SU-8 film with the pre-deposited OLEDs from the glass substrate. The thickness of the SU-8 flexible substrate is about 50 µm.

# 3 Results and discussion

As a choice of the transparent electrode in organic optoelectronic devices, the surface morphology of a metallic film is one of the key factors which affect the performance of the devices seriously. Firstly, we compared the surface morphology of the glass substrate and the SU-8 film by AFM as shown in Fig. S1.<sup>†</sup> The glass and SU-8 film show exceedingly a smooth surface and homogeneous roughness. We measured film morphologies of Au films on glass (Au/glass) and on SU-8 (Au/ SU-8) with various Au thicknesses using SEM and they are summarized in Fig. S2.† Depositing the Au film directly on the glass substrate exhibits obvious three-dimensional island growth mode (Volmer-Weber growth mode). Owing to the surface energy mismatch between Au and glass, thermally evaporated Au atoms slip easily and aggregate on the substrate surface, and the nucleation process leads to a rough and unconnected film in the long-region.<sup>23</sup> The AFM and SEM characterization studies in Fig. 1 show significantly different film morphologies between 7 nm Au on the SU-8 and on the glass. The 7 nm Au/glass film has a relatively high RMS roughness of 2.04 nm detected by AFM (Fig. S3<sup>†</sup>). When we engaged a SU-8 film to modify the glass surface prior to Au thermal deposition, the ultrathin Au film shows a uniform, continuous and smooth surface morphology. The roughness of the Au film on SU-8 is about 0.35 nm, which is nearly one order of magnitude lower compared with the Au film on the glass. The crosssection profiles of the Au films in Fig. S4<sup>†</sup> also demonstrate that the roughness and continuity have been improved by the insertion of the SU-8 film. Using the ultrathin Au film with a ultrasmooth and continuous surface morphology as the electrode of OLEDs would result in improvements in both conductivity and contact between the electrode and the organic layer and is benefit to the carrier injection.<sup>38</sup>

To investigate the effect of the SU-8 film on the growth process of the Au film, X-ray photoelectron spectroscopy (XPS) spectra of the Au/glass, SU-8/glass, and Au/SU-8 samples have



**Fig. 1** AFM image (a) and SEM image (b) of surface morphologies of the 7 nm ultrathin Au film deposited directly on the glass substrate (Au/ glass). AFM image (c) and SEM image (d) of the 7 nm ultrathin Au film on the SU-8 modified glass substrate (Au/SU-8).

been recorded as shown in Fig. S5.† The main components of SU-8 2025 are a bisphenol A novolak epoxy oligomer and a triarvlsulfonium salt as the photoacid generator (Fig. S6<sup>†</sup>).<sup>39</sup> The XPS spectra of the Au/SU-8 sample exhibit a positive chemical shift at the Au 4f core level and a negative chemical shift at the S 2p core level compared with those of the Au/glass and SU-8/ glass samples, while the XPS spectra of C 1s core level and O 1s core level remain unchanged. The chemical shift indicates the formation of the coordinate covalent bonds between the triarylsulfonium salt in the SU-8 layer and Au atoms.<sup>23</sup> The chemical bond is stronger than the Au-Au interaction during the film deposition. The deposited Au atoms are fixed on the surface of the SU-8 film through the chemical bond, providing dense nucleation centres for the following Au atoms. The chemical bonding between the SU-8 and Au suppresses the formation of large Au islands in the deposition process, resulting in the ultrasmooth and continuous ultrathin Au film.

Another important indicator of the continuity of the ultrathin Au film is sheet resistance which is also a key parameter for the electrode of OLEDs. The sheet resistance is strongly dependent on the thickness of the Au film. Fig. 2a shows the sheet resistance variation as the function of thickness. The Au/ SU-8 film exhibits obviously lower sheet resistance due to the SU-8-induced uniform and continuous Au film deposition. The sheet resistance of the 4 nm Au/SU-8 film is about  $8.3 \times 10^4$  $\Omega$  sq<sup>-1</sup>, while the Au/glass is not conductive. The average sheet resistance of 6 nm Au/SU-8 films is  $87.6 \Omega$  sq<sup>-1</sup>, and it is two orders of magnitude lower than that of the Au/glass films with an average sheet resistance  $4703.3 \Omega$  sq<sup>-1</sup>. The relatively low sheet resistance of the 7 nm Au/SU-8 film (23.75  $\Omega$  sq<sup>-1</sup>) is comparable to that of the ITO electrode (15  $\Omega$  sq<sup>-1</sup>).



**Fig. 2** (a) Sheet resistance (Rs) of the Au/glass and Au/SU-8 film as the function of the Au film thickness. (b) Transmittance of the Au/glass and Au/SU-8 film as the function of the Au film thickness at the wavelength of 550 nm.

The transmission spectra of the Au/glass and Au/SU-8 film with various Au film thicknesses have been recorded as shown in Fig. S7.† The transmittance decreases by increasing the thickness of the Au, and the 7 nm Au/SU-8 films still have a high transmittance of about 72% at 550 nm. The film-thickness-dependent transmittance under the normal incident light at the wavelength of 550 nm extracted from the transmission spectra is shown in Fig. 2b. They exhibit comparable transmittance at this wavelength. While in the longer wavelength region (Fig. S8<sup>†</sup>), the transparency of the Au/SU-8 is obviously higher than that of the Au/glass film, which is probably originated from the light scattering and the excitation of surface plasmon resonances of the Au/glass film due to its rough surface.<sup>30,31</sup> On the other hand, the SU-8 layer beneath the Au film acts as a refractive index-matching layer (n = 1.6), which may contribute to the enhanced light transmittance at a longer wavelength.40 The impact of the SU-8 thickness on the surface morphology, optical and electronic characteristics of the ultrathin Au/SU-8 film has also been investigated as shown in Fig. S9-S11 of the ESI.†

To demonstrate the application of the ultrathin Au transparent electrodes as the effective alternative of ITO, we fabricated OLEDs with a 7 nm Au film on SU-8 as the anode. The structure of OLED is shown in the inset of Fig. 3a. For comparison, an OLED with an ITO anode was also prepared using the same structure. The EL performance of the OLEDs with ITO and 7 nm Au/SU-8 anodes has been compared as shown in Fig. 4. The OLED with the 7 nm Au/SU-8 film as the anode shows favourable device performance as expected owing to the



**Fig. 3** EL performance of the OLEDs with the Au/SU-8 anode and traditional ITO anode. (a) Current density-voltage and (b) luminancecurrent density-efficiency of OLEDs. The inset in (a) shows the schematic structure of OLEDs based on the ultrathin Au/SU-8 anode.

good surface morphologies, optical and electrical characteristics of the ultrathin metallic film. The maximum luminance of the Au/SU-8 based OLED is 54 930 cd cm<sup>-2</sup>, and is much higher than that of the ITO based device with the luminance of 42 370 cd cm $^{-2}$  in spite of the high transmittance of ITO. Compared with the ITO based device, the current efficiency is increased from 34.5 cd  $A^{-1}$  to 40.6 cd  $A^{-1}$  by using the ultrathin Au anode, and it realizes a 17% enhancement. The improvement of EL performance arises from the excellent characteristics of the ultrathin Au film, as well as the suppression of the power loss induced by the waveguide modes associated with the ITO anode. The SU-8 layer under a Au electrode as a refractive index-matching layer to enhance the light extraction is also beneficial to improve the efficiency of OLEDs, which we can confirm from the cross-section views of the energy flux density in Fig. S13<sup>†</sup> simulated by the Finite-Difference-Time-Domain (FDTD) method.<sup>4</sup> The simulated energy flux density distributions of the Au/SU-8 and ITO based devices also demonstrate that the OLEDs based on Au/SU-8 electrodes have higher light extraction efficiency. Fig. 4a shows the EL spectra of the OLEDs with the Au/SU-8 and ITO anode, respectively. The Au/SU-8 based OLED exhibits an even broader EL spectrum, which implies a very weak microcavity effect. The weak microcavity effect can be further confirmed from polar plots of the EL





**Fig. 4** (a) EL spectra of the OLEDs with the ultrathin Au/SU-8 anode and traditional ITO anode. (b) EL intensity as a function of emission angle.

**Fig. 5** Flexibility and mechanical robustness of the flexible ultrathin Au/ SU-8 film and OLEDs. (a) The sheet resistances of the ultrathin 7 nm Au/ SU-8 film as a function of the number of bending times. (b) Luminance and efficiency of the OLEDs at 6 V as a function of the number of bending times. The insets of (a) and (b) show the photographs of the Au/ SU-8 film and the OLEDs operating at 6 V before and after bending, respectively.

intensities in Fig. 4b. The EL intensities of the Au/SU-8 based devices exhibit a slower decrease with the increased viewing angles compared to those of ITO based devices, and display a nearly Lambertian emission characteristic.

Bending tests have been conducted to evaluate the flexibility of the ultrathin Au/SU-8 film as well as OLEDs with the ultrathin anode on the peeled off SU-8 substrates. The ultrathin Au/SU-8 film exhibits favourable mechanical robustness, and its sheet resistance is maintained after repeated bending (Fig. 5a). The photographs of the Au/SU-8 film and the OLEDs operating at 6 V before and after bending are shown in the inset of Fig. 5a and b, respectively. There are no cracks or dark spots observed on the operating device after bending. The mechanical robustness of the flexible OLEDs is further investigated by measuring their EL performance after repeated bending in Fig. 5b. No obvious deterioration can be observed in the luminance and efficiency over up to 1000 bending cycles. The above results demonstrate that the OLEDs based on the ultrathin Au/SU-8 anode on the peeled off SU-8 substrates are highly flexible and mechanically robust.

### 4 Conclusions

In summary, we demonstrate an ultrathin and ultrasmooth Au film as an excellent alternative of the traditional transparent ITO electrode in OLEDs. The insertion of the SU-8 film modifies the glass substrate and fixes Au atoms via chemical bond interactions to form the ultrasmooth and continuous surface morphology of the deposited Au film. The 7 nm Au film shows excellent characteristics with a RMS roughness of 0.35 nm, a high transparency of 72% at the wavelength of 550 nm, and a sheet resistance of 23.75  $\Omega$  sq<sup>-1</sup>. By applying the ultrathin Au as the anode of the OLEDs, 17% enhanced current efficiency with Lambertian emission was realized. The high flexibility and the mechanical robustness of the ultrathin films and OLEDs have been demonstrated by conducting the bending tests. The ultrathin Au film satisfies the requirements of a desirable transparent electrode by its simple fabrication process, smooth film morphology, good transparency and conductivity, which offers an innovative and low-cost approach for the fabrication of large-area and flexible optoelectronic devices.

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