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Highly transparent and flexible fabric-based organic light emitting devices for unnoticeable wearable displays

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ABSTRACT

Fabric displays integrated into clothing are important members of wearable electronics. Highly transparent and flexible organic light emitting devices (OLEDs) fabricated on fabrics can make clothing keep the original appearance and wearing comfort and so are under focused research. Indium tin oxide is the main transparent electrodes due to its high transparency. However, its brittleness makes OLEDs lack of flexibility. Here ultrathin metal films are used as anodes and cathodes in OLEDs for both high transparency and flexibility. The anode and cathode are 7 nm Au film and 9 nm Ag film respectively. As a result, the total transmittance of the OLEDs is 74% at 550 nm wavelength when combining with a capping layer. Fabric displays based on the transparent and flexible OLEDs are realized. The patterns of the fabric with different colors can be seen clearly through the OLEDs. At the same time, the fabric display can withstand a 1 mm bending radius. This is the first reported transparent and flexible fabric displays, which will be useful for smart IT devices that are comfortable and fashionable to wear.

1. Introduction

Fabric-based light emitting devices have great potential in applications of wearable displays and lie in an important position among the next generation optoelectronic devices. High transparency and flexibility can make fabric displays unnoticeable visually and compliant in deformation, which are important for clothing to keep its original appearance and wearing comfort. Among various types of light emitting devices, such as inorganic light emitting diodes, alternating current electroluminescent devices and light-emitting electrochemical cell, organic light-emitting devices (OLEDs) are promising candidates for high-performance flexible and transparent fabric displays due to their high efficiency [1–4], tunable emission color [5–7], excellent flexibility [8–11] and the inherent transparency of vacuum-deposited organic functional films [12–16]. However, transparent and flexible fabric displays based on OLEDs have not been realized in previous reports. This is mainly restricted by the transparent and flexible electrodes.

Metal oxide semiconductors, such as indium tin oxide (ITO), are the most commonly used transparent electrodes due to their high transparency and conductivity. Transparent OLEDs (TrOLEDs) based on dual metal oxide semiconductors electrodes have been reported [13–15, 17–21]. Some of these devices exhibit high transparency with the average transmittance larger than 80% [14,20]. However, high-quality ITO films are usually fabricated by plasma-assisted sputtering deposition. The high-energy particles would significantly damage the underlying organic films during the sputtering process and degrade the devices performance. A buffer layer could protect the organic films against particle bombardment during depositing the top ITO electrode, while it increases the complexity of device structure. Thin metal films are promising candidates to replace ITO as the semitransparent top electrodes. TrOLEDs based on ITO bottom electrodes and thin metal films top electrodes have been extensively researched [16,22–30]. By using new electron injection materials as surface modification layer for Ag deposition, the cathode structure and the TrOLEDs exhibit extremely

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high transmittance of 91.2% and 87% at 550 nm respectively [27], which are comparable to the dual-ITO electrodes devices. However, the brittle ITO severely limits the devices flexibility and their applications in wearable displays.

Functional conductive materials, such as PEDOT:PSS, graphene and metal grids are suitable as flexible electrodes and have been studied to replace ITO in TrOLEDs [31–33]. For example, Xiang and coworkers have reported flexible and extremely efficient TrOLEDs by using metal–dielectric composite electrodes and Ag grid as cathode and anode respectively. The device exhibited a high transparency over 70% and a record external quantum efficiency of 72.4% [33]. Unfortunately, these functional conductive materials usually involve solution process which is not favorable for commercial manufacturing based on vacuum deposition and increases the complexity of device preparation process.

On the other hand, ultrathin metallic films with thickness smaller than 10 nm have been reported as electrodes and optical windows in organic optoelectronic devices [34–36]. By introducing seed layers and surface modification to control their nucleation and the growth kinetics, the ultrathin metal films exhibit uniform continuity, high conductivity, flexibility and transparency [37]. At the same time, the vacuum deposition mode is compatible well with other organic and inorganic charge injection, transport and light-emission materials, which greatly simplifies the OLED preparation process and makes ultrathin metal films show great potential as transparent and flexible electrodes. OLEDs based on ultrathin Ag films as both anode and cathode show high transmittance of 74.22% around 550 nm and flexibility when fabricated on a polyethylene terephthalate (PET) substrate [38].

In our previous work, ultrathin gold films have been deeply researched and employed as transparent electrodes for OLEDs and perovskite solar cells [39–41]. They exhibited comparable performance to ITO at the electrodes and devices level. On this basis, here we report fabric displays based on transparent and flexible OLEDs with ultrathin Au film and Ag film as anode and cathode respectively. By optimizing the thickness of the Ag film, the OLEDs show a total transmittance of 74% at 550 nm wavelength. The patterns of the fabric with different colors under the OLEDs can be seen clearly. At the same time, the fabric device can withstand a 1 mm bending radius. This is the first report of transparent and flexible fabric displays which promotes the development of wearable electronics.

2. Experiments

2.1. Materials

The photopolymer is NOA 63 (Norland Products Inc. (USA)). MoO_3, NPB (N,N′-diphenyl-N,N′-bis (1,1′-biphenyl)-4,4′-diamine), Ir(ppy)3 (tris(2-phenylpyridine)iridium(III)), mCP (N, N′-dicarbazolyl-3,5-benzene), TPBi (1,3,5-Tris(1-phenyl-1H-benzimidazol-2-yl)benzene) were offered by Xi'an Polymer Light Technology Corp. Fabrics were purchased from shops.

2.2. Transparent OLEDs fabrication

A polymer film was first spin coated on a clean glass substrate and cured by ultraviolet (UV) light. Its thickness was about 5 μm by tuning the spin speed and time. Then 5 nm MoO_3 and 7 nm Au were deposited on the polymer film as the ultrathin anode. The deposition rate of Au film was 0.5 Å s^{-1}. The following structure were MoO_3 (3 nm), NPB (N, N′-diphenyl-N,N′-bis (1,1′-biphenyl)-4,4′-diamine) (40 nm), $Ir(ppy)_3$ (tris(2-phenylpyridine)iridium(III)) doped mCP (N, N′-dicarbazolyl-3,5-benzene) (6%, 20 nm), TPBi (1,3,5-Tris(1-phenyl-1H-benzimidazol-2-yl)benzene) (35 nm) and Ca (3 nm). An ultrathin Ag (x nm) film with a deposition rate of 1 Å s^{-1} was used as the cathode. Finally, a capping layer of NPB with different thickness was deposited on the top side of the device. All materials were coated by vacuum thermal evaporation at a base pressure of 5×10^{-4} Pa.

2.3. Ultrathin Ag film fabrication

A polymer film was first spin coated on a clean glass substrate and cured by UV light. Then 30 nm TPBi and 3 nm Ca were deposited on the polymer film in series. Finally, Ag films with thickness of 11 nm, 10 nm, 9 nm and 8 nm were deposited on the TPBi/Ca layer, respectively. The TPBi/Ca layer was used as the modification layer for Ag films deposition.

2.4. Fabric display fabrication

Nylon was cut into desired size and taped to a glass sheet. A polymer film was spin coated on a clean glass substrate and cured by UV light. Then the same polymer was spin coated once again on the first cured layer. The glass/double layer polymer film was coated on the nylon surface with the polymer film facing down. The second polymer film was used as glue to bond the first cured layer and nylon together by UV light exposure. Finally, the polymer film/nylon composite was peeled off from the rigid and flat glass substrate. The polymer film was smooth and used as the planarization layer for nylon surface. Fabric displays with 7 nm Au anode, 9 nm Ag cathode and 50 nm NPB capping layer were fabricated on the planarization layer.

2.5. Characterizations

Sheet resistance values of the ultrathin Ag films were investigated by Four Point Probe instrument. Transmittance was measured by UV 2550 spectrometer. SEM images were taken by a JEOL JSM-7500F. AFM images were taken by a Dimension Icon AFM (Bruker Corporation). The electroluminescent characteristics of all OLEDs were measured by a source meter (Keithley 2400) and a spectrophotometer (Photoresearch 655). All OLEDs were measured in the air without encapsulation.

3. Results and discussion

We use ultrathin metal films as transparent electrodes in TrOLEDs. A 7 nm Au film is used as the anode at the bottom side which has been demonstrated as an efficient transparent electrode to replace ITO for high-performance organic optoelectronic devices [39,41]. The Au film has a smooth surface with root mean square (RMS) roughness of 0.41 nm (Fig. 1a). Its sheet resistance is about 12 Ω/□. Additionally, it has a transmittance value of 78% at 550 nm wavelength (Fig. 1b).

On the other hand, an ultrathin Ag film is selected as the cathode at the top side of the device. The thickness of the top Ag electrodes in top emission OLEDs is usually about 20 nm in previous reports [42–44]. Their transmittance at 550 nm is about 30%, which is not suitable for highly transparent OLEDs. Here, the properties of the Ag film is optimized by varying its thickness from 11 nm to 10, 9 and 8 nm respectively. The electrical and optical properties of ultrathin metal films are very sensitive to their thickness (Fig. 2). The sheet resistance increases while the transmittance decreases with increased thickness. Accordingly, they will influence the electroluminescence (EL) performance of TrOLEDs as shown in Fig. 3a and b. The devices are composed of a 7 nm Au anode and different Ag cathodes. EL performance are measured only from the Ag cathode side. The current density of the devices increases rapidly when the Ag cathode changes from 8 nm to 11 nm. The luminance increases from 8 nm Ag to 10 nm Ag, which corresponds well with the increased current density. However, it becomes stable when the Ag cathode changes from 10 nm to 11 nm. This is mainly due to the decreased transmittance from 10 nm Ag to 11 nm Ag although the device with 11 nm Ag cathode has larger current density.

The maximum current efficiency of the TrOLEDs decreases with increased thickness of Ag film. This means that the increasing rate of the device luminance is less than that of the current density at the same silver film thickness. For example, the current density of the device with 11 nm Ag cathode is 76 times more than that of the device with 8 nm Ag

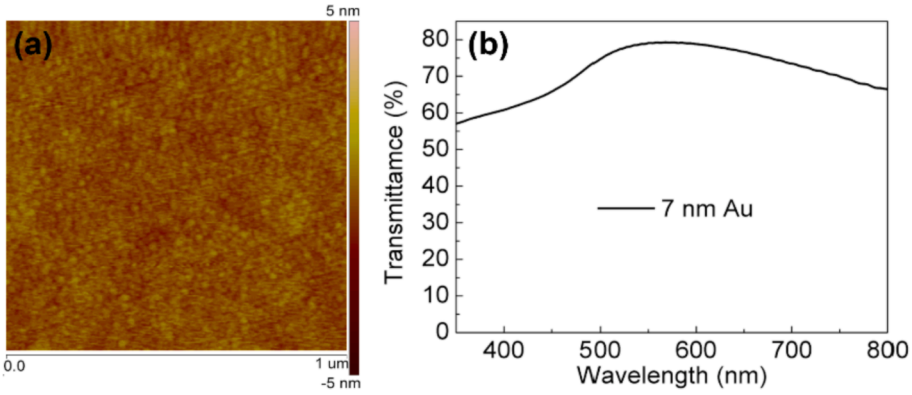

Fig. 1. (a) Atomic force microscope (AFM) image and (b) transmittance curve of the 7 nm Au film.

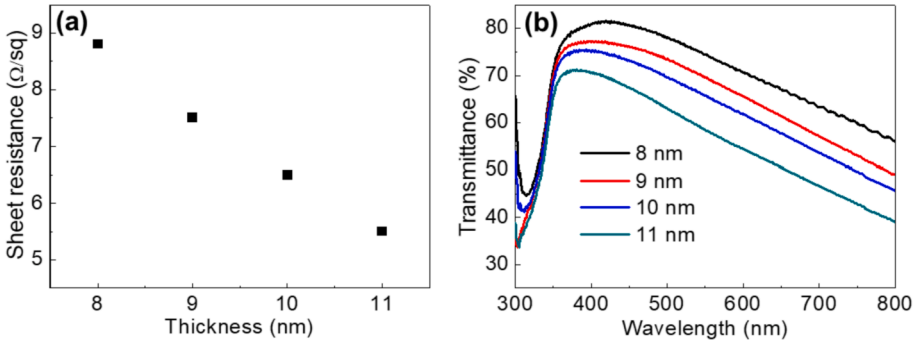

Fig. 2. Properties of Ag films. (a) Sheet resistance values and (b) transmittance curves of Ag films with thickness of 8, 9, 10 and 11 nm, respectively.

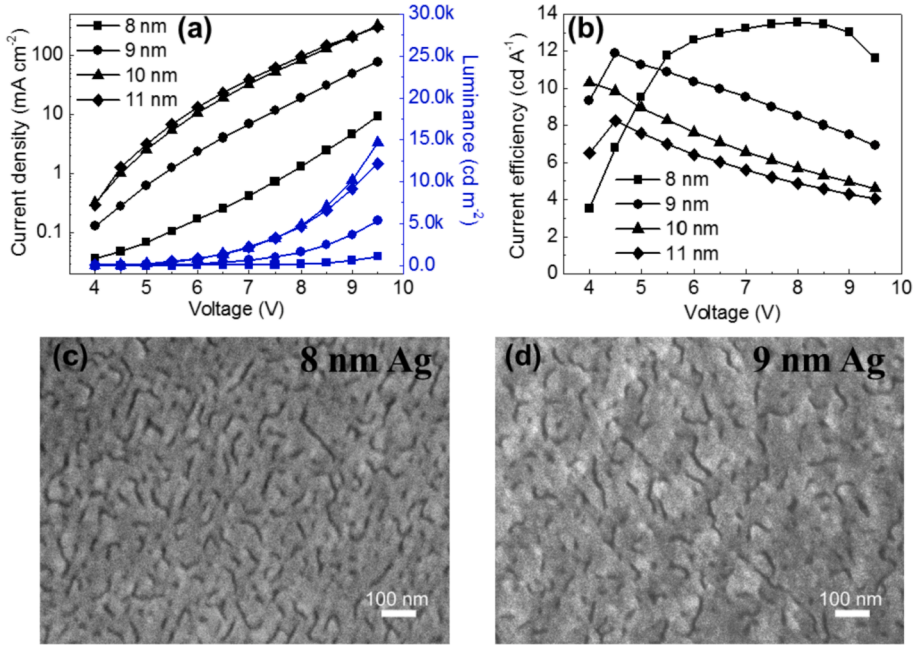

Fig. 3. Thickness selection of Ag films. (a–b) The electroluminescence performance of TrOLEDs with the thickness of cathode varying from 8 nm to 11 nm. (c–d) SEM images of 8 nm and 9 nm Ag cathode, respectively.

cathode at 6 V driving voltage. However, the luminance is only 39 times larger. The 8 nm device exhibits the largest current efficiency in all devices. However, the efficiency curve changes with voltage irregularly. This may be due to its poor film quality. We compare the surface morphology of 8 nm and 9 nm Ag films by scanning electron microscope (SEM) image as seen in Fig. 3c and d. The 8 nm film has smaller grains and more grain boundaries. This means that the integrality, continuity and surface coverage of the 8 nm film are worse than the 9 nm film. Therefore unstable and less charges would be injected from the 8 nm Ag film to the electron transport layer (ETL), which results in a big drop in

the current density, brightness and abnormal device performance. On the other hand, the sheet resistance difference of the two films is small due to the conductive path within the films and the excellent conductivity of Ag. So, we choose 9 nm Ag film as the transparent top electrode for fabric displays.

In order to improve the device transparency, a capping layer of organic material NPB (N,N′-diphenyl-N,N′-bis (1,1′-biphenyl)-4,4′-diamine) is coated on the top of the TrOLEDs [16,23,25]. The thickness of the capping layer increases from 20 nm to 80 nm and the corresponding device transmittance is measured and shown in Fig. 4a. The initial transmittance of the TrOLEDs with 9 nm Ag cathode at 550 nm is 52%. It can be seen from the curves that the total transmittance of the TrOLEDs increases when the thickness of the capping layer increases from 20 nm to about 50 nm and starts to decrease from 60 nm to 80 nm. The largest transmittance at 550 nm is 74% with the 50 nm capping layer. Fig. 4b shows the photograph of the TrOLEDs with the highest transparency. The building in the background can be seen through the device indistinguishably comparing to other parts of the picture. Fig. 4c and d shows the EL performance of the TrOLEDs measured from the anode and cathode side, respectively. The maximum luminance from the anode side and the cathode side is 17900 cd/m^2 and 15300 cd/m2, respectively. The maximum current efficiency from both sides is 12.4 cd/A and 12.8 cd/A. So, the total current efficiency of the TrOLEDs is 25.2 cd/A.

The angular dependence characteristics of the EL intensities of the TrOLEDs have been measured from the bottom and top side, respectively (Fig. 5a). It can be seen that the TrOLEDs follows the ideal Lambertian emission from both bottom and top sides. The emission characteristics is further investigated by the angular dependence EL spectra (Fig. 5b and c). No blue shift has been observed at different viewing angles in the EL spectra from both the anode and cathode sides. The Lambertian emission and the stable EL spectra are mainly attributed to the high transparency of the ultrathin metal electrodes which decrease the micro-cavity effect in the TrOLEDs. Such angle-independent EL behaviors are desirable for display applications.

The TrOLEDs with high transparency are integrated with fabrics by a simple film transferring process. Fabrics are usually composed of weaved thread which makes them porous and rough and not suitable for OLEDs fabrication. Here a polymer planarization layer is used to smooth its surface. First, a polymer film is fabricated on a glass substrate by spin coating. Then it is transferred onto the surface of the fabric (Fig. 6a). They bond together at the contact points by using a layer of photosensitive polymer as glue (Fig. 6b). Then the fabric together with the polymer film is peeled off from the rigid and flat glass sheet (Fig. 6c). Because the top side of the polymer film is contacted with the glass substrate, its surface is smooth. Nylon is neat and weaved tidily without unnecessary thread residues and so is selected as the fabric substrate (Fig. 6d). The surface root mean square roughness (RMS) of the polymer planarization layer on its surface is only 0.6 nm (Fig. 6e). The smooth surface is suitable for OLEDs fabrication.

Fig. 7a–c shows the fabric displays integrated with TrOLEDs. The color and patterns of the fabric can be seen clearly when the TrOLED is turned off and the fabric can maintain its visual aesthetic feeling (Fig. 7a). When the TrOLED is turned on, its brightness is able to be tuned by changing the driving voltage (Fig. 7b). Enough brightness is able to obtain to reduce the impact of the fabric for information display (Fig. 7c). Additionally, the thickness of the polymer planarization layer is about 5 μm. The thin film makes the fabric displays flexible. It can withstand a 1 mm bending radius (Fig. 7d–f). Cyclic bending test has been conducted, while the device degrades quickly after tens of bending cycles. The poor bending stability may be due to the breakdown of the ultrathin metal electrodes. During the cyclic bending test, the thread of the fabric may slide locally. The changes of the weaved structure can result in abnormal deformations of the planarization layer. Consequently, the ultrathin metal electrodes would be damaged, which leads to the quick degradation of the fabric OLEDs. Further research is needed to improve the bending stability of the fabric devices by optimizing materials and process. Fig. 7g and h are the EL performance of the fabric displays. Because the fabric is not transparent, the measurement is only conducted from the top side. The turn-on voltage is 3 V. The leakage current is small, which is attributed to the smooth surface of the planarization layer and the Au anode. The maximum luminance is larger than 10000 cd/m^2 which is large enough for indoor and outdoor display applications. At the same time, the maximum current efficiency is

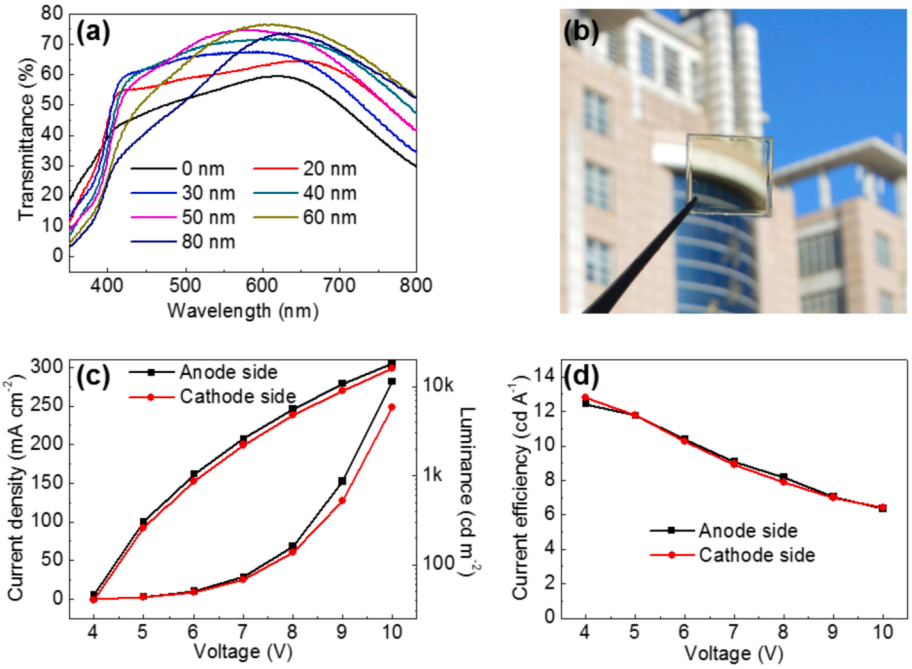

Fig. 4. Properties of the TrOLEDs. (a) Transmittance curves of TrOLEDs with the capping layer thickness of 0, 20, 30, 40, 50, 60 and 80 nm respectively. (b) A photograph of the TrOLEDs with 9 nm Ag cathode and 50 nm capping layer. (c) and (d) EL performance of the TrOLEDs measured from the anode and cathode side, respectively.

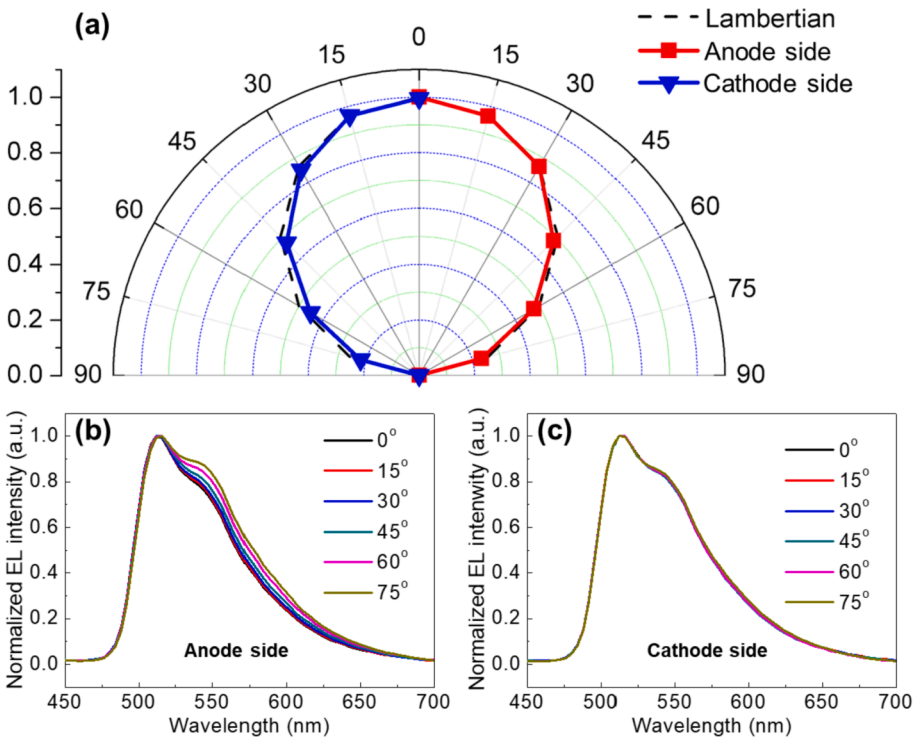

Fig. 5. Angular dependence characteristics of the TrOLEDs. (a) The angular dependence characteristics of the EL intensities of bottom and top emissions. EL spectra of the TrOLEDs at viewing angles of 0, 15, 30, 45, 60 and 75° from the anode side (b) and the cathode side (c), respectively.

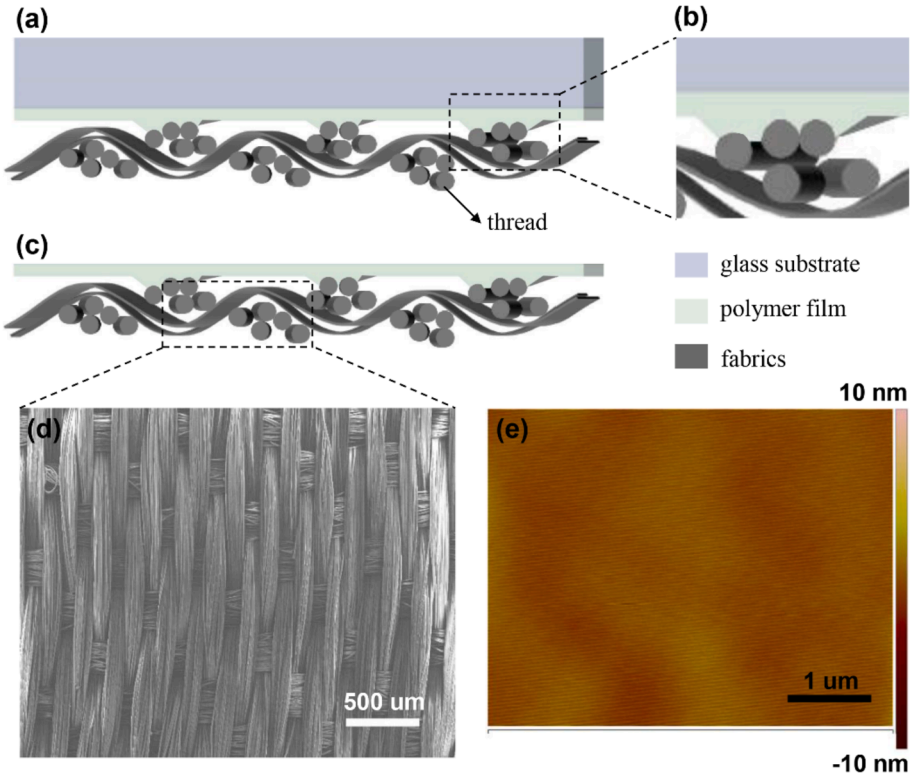

Fig. 6. Fabrication of the surface planarization layer on fabrics. (a) Transferring a polymer film from a glass substrate to the fabric surface. A layer of photosensitive polymer is used as glue to bond them together. (b) Enlarged image of the bonding point. The polymer film and the fabric bond together at the high spots of the fabric surface. (c) The fabric together with smooth planarization layer after peeling off from the glass substrate. (d) SEM image of Nylon. (e) Atomic force microscope (AFM) image of the polymer planarization layer on Nylon.

16.7 cd/A which is a little larger than the single side value of the TrOLEDs on glass substrate. This may be attributed to the reflection of the fabric, which results in more light emission from the top side.

4. Conclusions

In conclusion, we have reported fabric displays based on organic light emitting devices with ultrathin metal films as both anode and cathode. By optimizing the thickness of the top Ag electrode and the

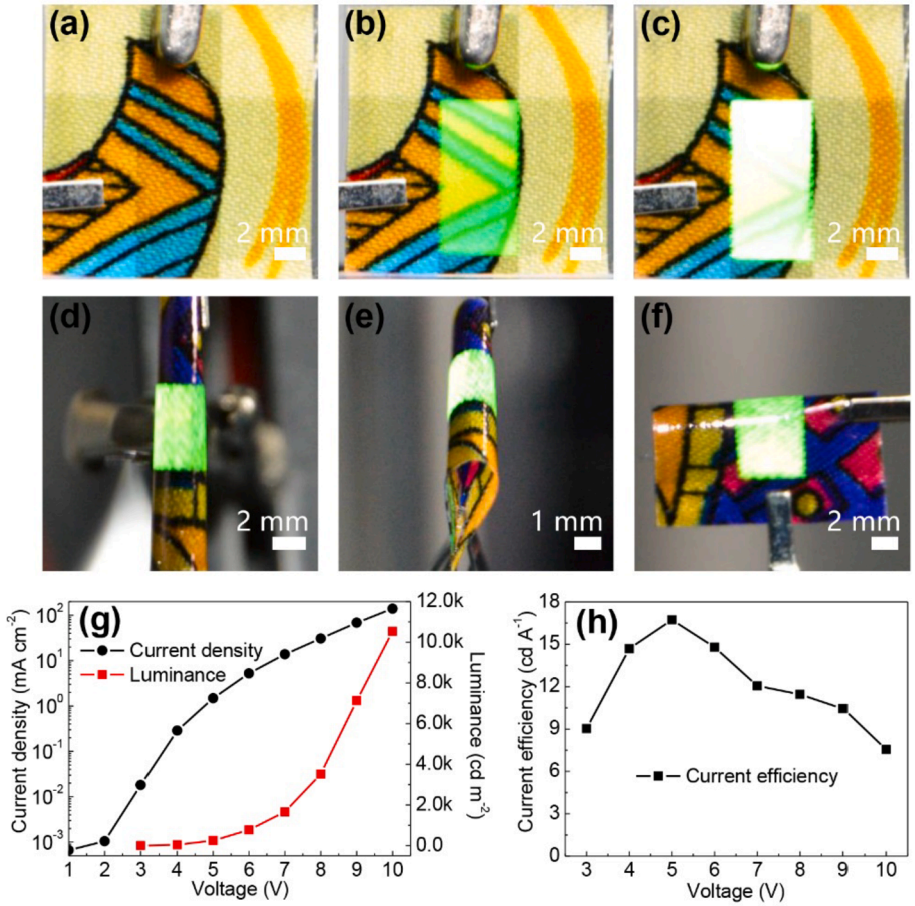

Fig. 7. Fabric displays. (a–c) Photographs of fabric displays at different driving voltage: 0 V, 5 V and 7 V. (d–f) Flexibility demonstrations of the fabric displays from different viewing angles. (g) and (h) EL performance of the fabric displays.

capping layer, the TrOLED exhibits high transparency with its total transmittance of 74% at 550 nm wavelength. Fabric displays based on the TrOLEDs have been fabricated by a simple film transferring process. The high transparency of the OLEDs makes the fabric displays nearly invisible when it is turned off, which is beneficial for clothing to maintain its visual aesthetic feeling. On the contrary, high brightness can be achieved by tuning the driving voltage to eliminate the negative effects of fabrics for information display. At the same time, the fabric displays are very flexible. They can withstand a 1 mm bending radius. The high flexibility of the fabric displays is important for clothing to keep wearing comfort. The high transparency and flexibility make fabric displays step forward to wearable applications.

Notes

The authors declare no competing financial interest.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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