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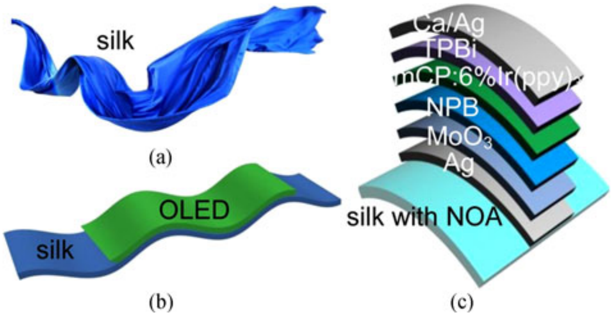

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Flexible Efficient Top-Emitting Organic Light-Emitting Devices on a Silk Substrate

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Abstract: A flexible efficient top-emitting organic light-emitting device (TOLED) on an off-the-shelf silk substrate has been demonstrated by planarizing the silk substrate with photopolymer NOA63. The flexibility of the bare silk substrates was retained in the planarized silk substrates due to ductile characteristics of cured NOA63. The planarized silk substrate has shown superiority on surface morphology, which is beneficial to the performances of OLEDs. Their maximum luminance and current efficiency are 45545 cd/m^2 and 37.7 cd/A, respectively. Moreover, our devices show not only high luminance and efficiency but also high flexibility and mechanical robustness. Emission of operating devices is uniform and free of defects under a very small bending radius and the luminance and efficiency do not deteriorate obviously after repeated bending. TOLEDs on silk substrate are a potential alternative to wearable displays.

Index Terms: Organic light-emitting devices, silk substrate, flexible, wearable.

1. Introduction

Organic optoelectronic devices are unique and attractive owing to their advantages in terms of flexibility. Flexible organic optoelectronic devices and their applications such as electronic newspapers, wearable displays and light collectors will be magnificent in the emerging technology [1]–[8]. As a key component, flexible organic light-emitting devices (OLEDs)can be applied as wearable displays, and therefore attracting much more attention for its technological and practical significance [1]–[4], [6]–[8]. Wearable displays not only realize the information display itself but also can be coupled with other flexible electronic devices such as sensors, thin film transistors, energy generating and storage devices to constitute a visual and wearable system [9]–[11]. This system has potential and valuable applications in human health monitors. In order to realize these fantastic devices, appropriate flexible substrates are crucial and imperatively necessary. Substantial efforts have been directed towards developing flexible OLEDs with different substrates, such as ultrathin glass sheet [12], metal foil [13] and plastic film [1]–[4], [6]–[8]. Unfortunately, ultrathin glass sheet is still very brittle so as to restrict its flexibility. Metal foil has shortages of heavy weight, difficulty in mass production, hard to endure in repeated bending and an additional planarization step is usually required to prevent electrical shorting. As a result, plastic film as an instance of polyethylene terephthalate (PET) has been used as a flexible substrate of OLED most commonly. However, plastic film substrates also present an important challenge to maintain essential fabric clothes properties such as comfort level and sewability. In comparison to flexible display applications that are just required to flexibility and mechanical stability, wearable displays must furtherly have better comfort level and sewability, for people are obviously inseparable from the clothes in daily life. Therefore, integrating flexible display devices into the everyday clothes plays a key role for human-friendly wearable displays.

To date, however, clothes based display devices research is still in its infancy and there have been only limited reports on these devices [14], [15]. OLEDs with high efficient and mechanical stability fabricated on actual fabric clothes substrate is still a crucial problem for the development of wearable OLED displays. It is well-known that the thickness of OLEDs is just hundreds of nanometers including electrode but a single fiber of fabric clothes is almost tens of micrometers and surface roughness originating from fabric construction is even larger. Therefore, the surface morphologies of fabric clothes have to be planarized to prevent shorting and defects such cracks and pinholes. In this letter, we have explored the fabric construction of different off-the-shelf fabric clothes and furtherly demonstrated a flexible efficient top-emitting OLED on silk substrate by planarizing the silk with photopolymer NOA63. The planarized silk substrate has shown superiority on both flexibility and surface morphology, which are beneficial to the performances of OLEDs. Their maximum luminance and current efficiency are 45545 $\mathrm{cd/m^2}$ and 37.7 cd/A respectively. Moreover, our devices show not only high luminance and efficiency but also high flexibility and mechanical robustness. The emission of devices is uniform and free of defects under a very small bending radius and the luminance and efficiency do not deteriorate obviously after repeated bending.

2. Experimental Details

2.1 Fabrication and Characterization of Substrates

Firstly, the surface morphologies of a number of fabric clothes with different materials and weaving process were investigated by the scanning electron microscope (SEM, JEOL JSM-7500F). These off-the-shelf fabric clothes involving cotton, linen, silk, nylon, acrylic and terylene were all commercially available. Then we have chosen the silk as a substrate for OLEDs because of its neat and exquisite fabric construction. The thickness of the silk is about 150 μm. A photopolymer (NOA63, Norland) film was used as planarization layer for the silk substrate via spin coating. Then the silk substrate with NOA63 was exposed to an ultraviolet light source for 5 min. The power of the light source is 125 W. The thickness of cured NOA63 films was less than 200 μm. Although coming slightly thicker, the flexibility of bare silk was almost maintained due to their excellent ductile characteristics. Moreover, many other characteristics of the bare silk substrates were retained in the planarized silk substrates, such as comfort level and sewability. The surface morphologies of planarized silk substrates were also investigated by SEM and atomic force microscopy (AFM, iCON, Veeko).

2.2 Fabrication and Characterization of OLEDs

Fig. 1(a) and (b) presents a schematic diagram of silk substrates without and with OLEDs. The top-emitting OLEDs (TOLEDs), a stack structure designed by considering the Fabry-Perot optical resonant microcavity effects [16], [17], were fabricated on the planarized silk substrate by thermal evaporation. The organic layers and electrodes were deposited layer by layer at a rate of 1 Å s^{-1} with a shadow mask in the vacuum chamber under the base pressure of 5×10^{-4} Pa. Both anode and cathode were Ag film. A classic green-emitting phosphorescent tris- (2-phenylpyridine) iridium ($\mathrm{Ir(ppy)_3}$) with the ratio of 6 wt% doped into a suitable host material m-bis(N-carbazolyl) benzene (mCP) was used as emitting 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 used as hole-transport layer and electron-transport layer [18], [19]. $\mathrm{MoO_3}$ was used as anodic buffer and Ca was inserted into the cathode and organic layers to enhance electron injection. The detailed structure is Ag (80 nm)/ $\mathrm{MoO_3}$ (4 nm)/ NPB (40 nm)/ mCP: $\mathrm{Ir(ppy)_3}$ (6% wt, 20 nm)/ TPBi (35 nm)/ Ca (3 nm)/

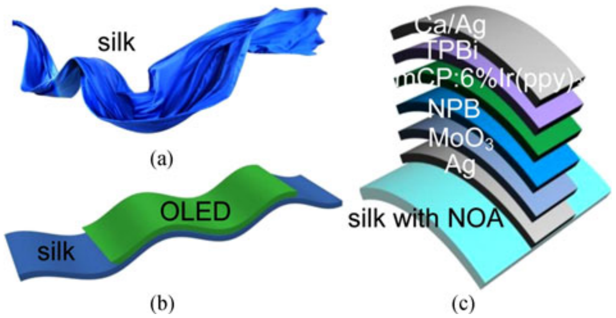


Fig. 1. Schematic images of (a) silk, (b) OLED on silk substrate and (c) stacked structure of TOLED.

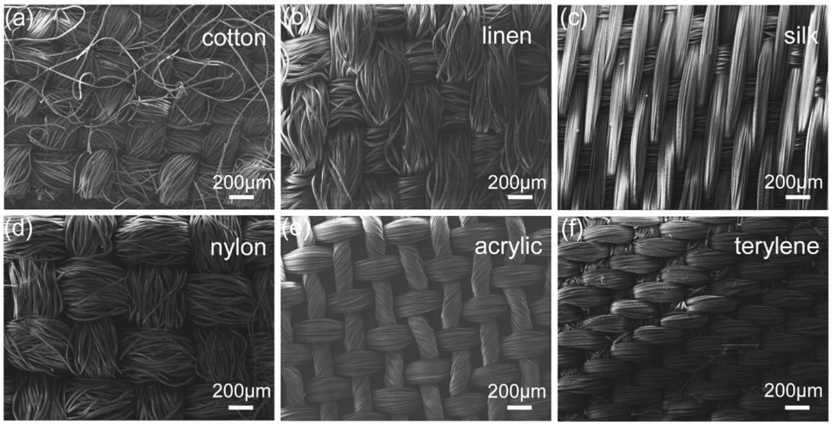


Fig. 2. SEM images of different off-the-shelf clothes fabrics: (a) cotton, (b) linen, (c) silk, (d) nylon, (e) acrylic and (f) terylene.

Ag (20 nm), as shown in Fig. 1(c). The active area of the device is $2 \times 2\,\text{mm}^2$. The current density-voltage (J-V), luminance-current density-efficiency (L-J-E) and spectra electroluminescence (EL) characteristics of the devices were measured by Keithley 2400 programmable voltage–current source and Photo Research PR-655 spectrophotometer. In addition, we have also measured EL characteristics of devices after multiple bending to evaluate their flexibility and mechanical stability. All of the measurements were conducted in air at room temperature.

3. Results and Discussion

As a substrate for flexible OLEDs, surface morphology is crucial for the performances of devices. In terms of commercial off-the-shelf clothes, weaving process and fabric materials play a fundamental role in the surface morphology. Fig. 2 shows SEM images of different common clothes fabrics: (a) cotton, (b) linen, (c) silk, (d) nylon, (e) acrylic and (f) terylene. All weaving involves two perpendicular sets of yarns which are interlaced to form a fabric, but the fabric constructions and surface morphologies are obviously different due to different materials and weaving process. Specifically, cotton is rich in thread residue. Linen and nylon weave loosely and irregularly. Though acrylic and terylene are compact, there are many holes in the acrylic and some little thread residue in one of the perpendicular sets of yarns of terylene. Among them, silk has shown great potential as a substrate due to more sophisticated weaving and more neatly and tidily align of silk fiber and therefore is chosen to fabricate TOLEDs.

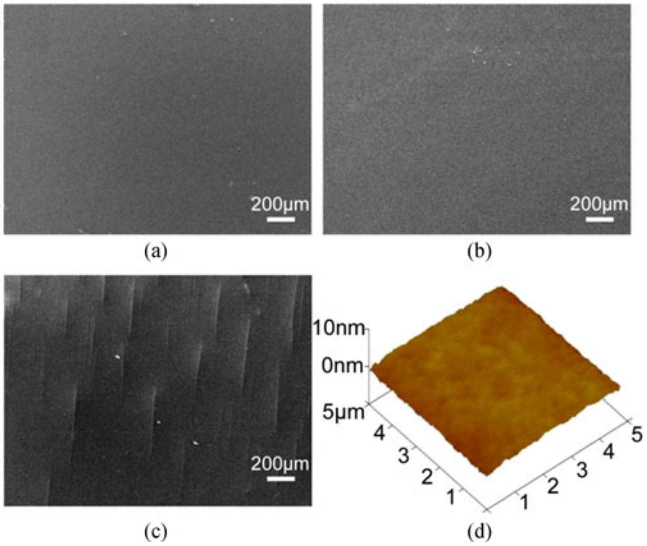
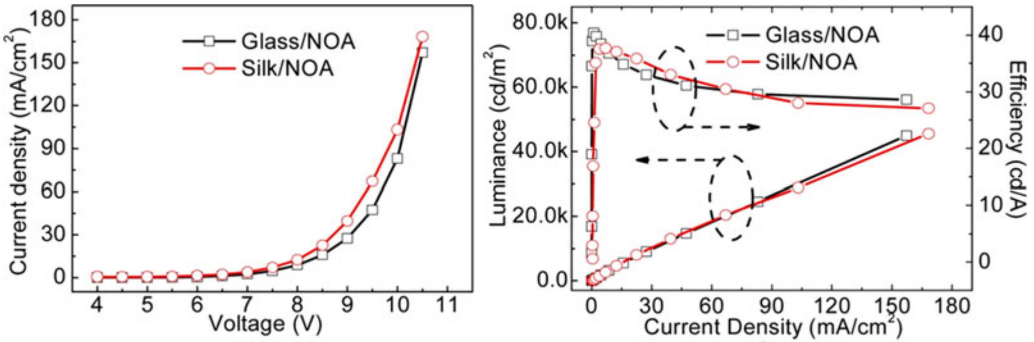


Fig. 3. SEM images of silk with photopolymer at different spin speed: (a) 1000 rpm, (b) 1500 rpm and (c) 2000 rpm. (d) AFM image of surface morphology for silk with photopolymer at 1500 rpm spin speed.

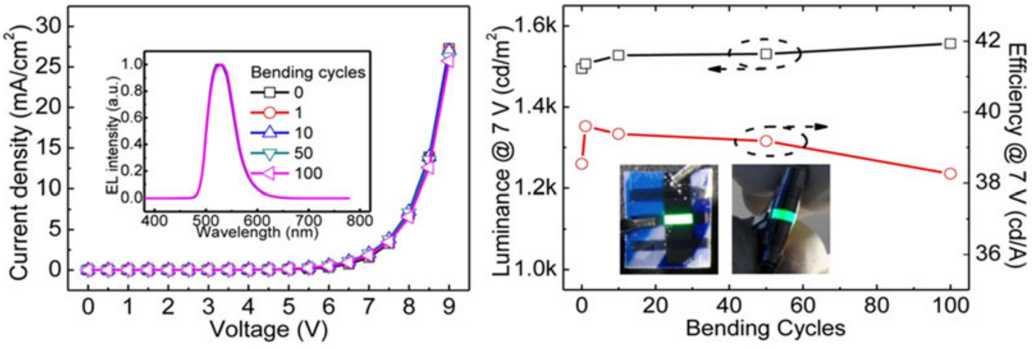
NOA63 has been used to planarize the silk substrate which has proved excellent flexibility and surface morphology by using as a substrate of organic optoelectronic devices in our previous work [20]–[22]. It offers relatively smooth surface morphology and retains flexibility of the substrate in this work, and not only can be deposited easily using spin-coating but also cured rapidly under the ultraviolet source. The NOA63 would disperse and permeate into fabric construction of silk substrate due to proper viscosity when spin-coating process. In addition, the short cured time of NOA63 also plays an important role. After spin-coating, fast curing would prevent fluidic NOA63 persistently permeating into the silk substrate up to down which obviously give rise to collapse of the planarized film and exposure of the fabric construction. The excessive NOA63 permeate across the silk substrate is also adverse to keep the flexibility of substrate because dispensable NOA63 surrounding the silk substrate would degrade its flexibility and comfort level. In order to optimize the condition of spin-coating, we have used different spin speed to planarize the substrate because excessive higher speed have little impact on planarizing the surface morphology, while excessive lower speed will lead to dramatically deteriorate in flexibility and comfort level of the substrate. As shown in Fig. 3(a)–(c), the surface morphologies of silk substrate are obviously different under various spin speed. The SEM images exhibit smooth surfaces at the spin speed of 1000 rpm and 1500 rpm as shown in Fig. 3(a) and (b), while observable embossments originating from fabric constructions are present at the spin speed of 2000 rpm as shown in Fig. 3(c). These SEM images prove the spin speed of 1500 rpm may be proper to planarize the silk substrate. To clarify whether the surface morphology is smooth enough to fabricate the OLEDs, we have also measured and shown the AFM image of the planarized silk substrate at the spin speed of 1500 rpm in Fig. 3(d). The root mean square (rms) roughness of the substrate is 0.682 nm in the area of 5 μm. It indicates that the planarized silk substrate is generally applicable in OLEDs which require a smooth surface.

Therefore, TOLEDs have been fabricated and characterized in this planarized silk substrate and TOLEDs are also fabricated on conventional glass substrate under identical process conditions as control. The J-V and L-J-E characteristics are summarized in Fig. 4. The current density, luminance and efficiency of TOLEDs on the planarized silk substrate are all very close and comparable to that of the glass substrate. The devices on the planarized silk substrate exhibit high EL performances and the maximum luminance and efficiency are 45545 cd/m^2 and 37.7 cd/A respectively. As above-mentioned results, by taking advantage of comfort level, sophisticated weaving process of the silk



(a) (b)

Fig. 4. EL performances of OLEDs on glass and silk with photopolymer substrates. (a) Current density-voltage and (b) luminance-current density-efficiency characteristics.



(a) (b)

Fig. 5. (a) Comparison of EL spectra and J-V characteristics before and after repeated bending. (b) Luminance and efficiency at 7 V as a function of the number of bending cycles. Inset: photographs of OLEDs on the silk substrate before and after bending.

and the flexibility, rapid cured speed and surface smoothness of the NOA63, we obtained a high efficient TOLED on off-the-shelf fabric clothes.

In order to evaluate the flexibility and mechanical stability of the TOLEDs on the planarized silk substrate, a bending test has been conducted. We bend the flexible substrate from plane to semicircle and convolve the device on a glass bar. As shown in Fig. 5(a), the EL spectra and J–V curves are almost coincident up to 100 bending cycles at the condition that the bending radius is 8 mm. In addition, no obvious deterioration in the luminance and efficiency can be observed after repeated bending as shown in Fig. 5(b). We have also shown the photographs of the flexible TOLEDs operating at 7 V before and after bending. The emission of TOLEDs is uniform and there are no defects observed on the operating devices. These results prove that the TOLEDs on the planarized silk substrate are not only highly flexible but also highly mechanically robust.

4. Conclusion

In conclusion, we have demonstrated a flexible efficient top-emitting organic light-emitting device (TOLED) on off-the-shelf silk substrate by planarizing the substrate with photopolymer NOA63. The planarized silk substrate has shown not only superiority on surface morphology but also high flexibility and mechanical robustness, which are beneficial to the performances of flexible OLEDs. The devices on the planarized silk substrate exhibit high luminance and efficiency. Moreover, the emission of operating devices is uniform and free of defects under a very small bending radius. The luminance and efficiency do not deteriorate obviously after repeated bending. We expect that our results may provide an alternative way to practical wearable displays.

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