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Polymer encapsulation of flexible top-emitting organic light-emitting devices with improved light extraction by integrating a microstructure

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ABSTRACT

An improved efficiency from an encapsulated flexible top-emitting organic light-emitting device (FTOLED) has been demonstrated by integrating a microstructure onto the polymer encapsulation film. Soft-nanoimprint lithography is employed to integrate the microstructure onto the polymer surface, which enables large area fabrication with high quality, low cost, and repeatable use of the poly(dimethylsiloxane) mold. The light extraction of the FTOLEDs has been improved by integrating the microstructure with two-dimensional tapered micropillars array on the polymer encapsulation film, which can suppress the reflection by enhancing the critical angle of total reflection owing to its gradually changed refractive index. Moreover, the microstructured surface exhibits a hydrophobic property owing to its high contact angle, which results in a self-cleaning ability to protect the FTOLEDs from being polluted by water droplets and dust particles in practical applications.

1. Introduction

Organic light-emitting devices (OLEDs) have been attracting increasing attention owing to their potential applications in full color flat panel displays and solid-state lighting [1–10]. Particularly, the flexibility of both small molecule and polymer further enables the use of these materials in flexible devices [1,4,11–13]. The flexible optoelectronic systems and their applications such as electronic newspapers, wearable displays and light collectors will be magnificent and exciting technology. However, encapsulation is necessary for the commercial use of the OLEDs due to the sensitivity of the organic molecules to the oxygen and water vapor. Although encapsulation of OLEDs between glass plates is straightforward, this solution obviously sacrifices flexibility and mechanical robustness of the final device. The development of flexible encapsulation technology has been recognized as key technology for the realization of the flexible OLEDs [11,14–18]. In recent years, there are several different strategies for the thin film encapsulation, such as multi-layered barriers consisting of inorganic thin films fabricated by plasma enhanced chemical vapor deposition (PECVD) [17] atomic layer deposition (ALD) [15,19], and polymer thin film encapsulation such as poly(dimethylsiloxane) (PDMS) film [16]. Unfortunately, these methods have inevitable drawbacks, such as high cost of PECVD and ALD and damage to the organic

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molecules during the heat curing process of the PDMS. Moreover, in case of encapsulation of flexible top-emitting OLEDs (FTOLEDs), light is transmitted from the encapsulation layer, which results in a power loss due to the total reflection at interface between encapsulation film and air. A simple and large-scale low-cost manufacturing method is required for the FTOLED encapsulation to overcome the power lost, and maintaining its good flexibility as well.

Integrating microstructure has been demonstrated an effective method to reduce reflection and enhance transmittance of thin films [20–27]. However, it is difficult to direct integrate microstructure onto the inorganic encapsulation layers, such as ALD or PECVD deposited AlO_x and SiN_x. In this letter, a highly flexible photopolymer encapsulation film with direct patterned microstructure has been employed in the FTOLEDs, and demonstrated its effect on the improved light extraction. Soft-nanoimprint lithography was employed to integrate the microstructure on the polymer surface, which enables large area fabrication with high quality, low cost, and repeatable use of the PDMS mold [28–30]. The microstructure on top of the encapsulation film is two dimensional (2D) tapered micropillars array. The micropillars array with tapered morphology showed a gradually changed refractive index, and the critical angle of total reflection would be enlarged. As a result, the light loss induced by the total reflection could be recovered effectively. The current efficiency of the FTOLEDs is increased from 9.26 to 12.36 cd/A after the microstructured photopolymer encapsulation was employed on top of the FTOLEDs. Moreover, the microstructured encapsulation film exhibit a high contact angle (CA) of 123.23°, while it is only 65.49° for that of the planar surface. The high CA permits a self-cleaning effect of the encapsulating film by reducing the probability of water condensation and dust accumulation, so it is beneficial to prevent the FTOLEDs from being polluted in practical applications.

2. Experimental details

2.1. Fabrication of flexible substrate with ultrasmooth Ag anode

A photopolymer (NOA63, Norland) film was used as both flexible substrate and encapsulating film for the FTOLEDs. An ultra-smooth Ag film was deposited onto the flexible substrate as the anode by employing a template stripping technique [13,31–35]. A cleaned silicon (Si) template was loaded into a thermal evaporation chamber and a 80 nm Ag film was deposited on the template at a rate of 1 Å/s at a base pressure of 5×10^{-4} Pa. Then, photopolymer film was spun coated onto the Si template predeposited with the Ag film for 20 s at 1000 rpm and exposed to an ultraviolet light source for 5 min. The power of the light source is 125 W. Then, the cured photopolymer film can be peeled off from the Si template. Owing to the cured photopolymer has better adhesion with the Ag film than that with the Si template, Ag film would remain on the photopolymer after the cured photopolymer was peeled off and a flexible substrate with Ag film was obtained. Although the evaporated metal film has a rough surface after deposition, the smoothness of the opposite interface is near that of the Si templates. This method exhibits particular advantage in flexible optoelectronic devices, because not only the smoothness of the Ag anode can be improved, but the backing layer itself is flexible and can be used as the substrate.

2.2. Fabrication of microstructured PDMS mold

Si substrate was cleaned with acetone, alcohol, and deionized water. Then the photoresist (S1805, Rohm and Hass Electronic Materials K.K.) was spin coated on the substrate at 2000 rpm speed for 30 s. The lithography experiments was performed by using a frequency-tripled Nd:YAG laser (Spectra-physics Company) with 3 nm pulse width, 10 ns pulse length, 10 Hz repetition rate and 355 nm wavelength. The sample was exposed for 1 pulse at 0.8 W by two laser beams which were split from the UV laser. The microstructure recorded on the S1805 film with different period and depth can be obtained by adjusting the writing angle and the exposure pulses. Array of micropillar with tapered side wall and with the height and period of about 200 and 350 nm, respectively, was obtained. The patterned photoresist was used as the master. Then the PDMS mold was prepared as follows. Silicone elastomer base and curing agent (Dow Corning Co.) with a ratio of 10:1 were well mixed, degassed by centrifugal process for 5 min at 6000 rpm, spin cast onto the master molds, and then baked at 95 °C for 1 h for solidification. At last, the solidified PDMS film was peeled off from the master and the PDMS mold with the corresponding relief of the master was obtained.

2.3. Fabrication and evaluation of the FTOLEDs with the microstructured encapsulation

Fig. 1(a) presents a schematic diagram of the fabrication process of the FTOLEDs with the microstructured encapsulation by the soft-nanoimprint lithography. Prepared flexible substrates with ultrasmooth Ag films were loaded into a thermal evaporation chamber. The organic layers, LiF, thin Al and Ag cathode were deposited. MoO_3 is used as anodic buffer in the TOLEDs. 4,4′,4″-tris(3-methylphenylphe-nylamino) triphenylamine (m-MTDATA) and *N,N′*-diphenyl-*N, N′*-bis(1-naphthyl)-(1,1′-biphenyl)-4,4′-diamine (NPB) are hole-injection and hole-transport layers, respectively. Tris-(8-hydroxyquinoline) aluminum (Alq_3) was used as emitting layer. The structure of the TOLEDs is Ag (80 nm)/MoO_3

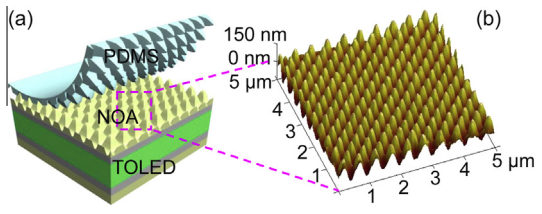

Fig. 1. (a) Schematic process of TOLEDs with microstructured encapsulation. (b) AFM figure of surface morphologies for the microstructured encapsulation film.

(4 nm)/m-MTDATA (30 nm)/NPB (20 nm)/Alq_3 (50 nm)/LiF (1 nm)/Al (1 nm)/Ag (20 nm). The active area of the device is 2×2 mm^2. A NOA63 film was spun coated on top of the FTOLEDs for 20 s at 1000 rpm. Placing the prepared PDMS mold into contact with the NOA63 film and then exposing it to ultraviolet light through the transparent mold cured the NOA63 into a solid form. The resulting NOA63 surface presented a relief structure in the geometry of the PDMS mold. At last, the PDMS mold was peeled off and it could be used repeatedly. The three dimensional atomic force microscopy (AFM) image of the reproductive microstructure on the surface of the encapsulation film was shown in Fig. 1(b). Two dimensional tapered micro pillars array are formed on the encapsulation film, and its period and the height are about 350 nm and 200 nm, respectively The voltage–luminance and voltage–current density characteristics of the FTOLEDs were measured by Keithley 2400 programmable voltage–current source and Photo Research PR-655 spectrophotometer. All of the measurements were conducted in air at room temperature.

3. Results and discussion

The period and height of the 2D array of micropillar are crucial for the light transmission of the encapsulation film. The period should be less than λ/n to suppress orders diffracted inside the materials and the height should be larger than 0.4 λ [36–38]. The λ is wavelength (532 nm for Alq_3) and n is refractive index (around 1.5 for the NOA63). For the planar encapsulation film of the NOA63, light is trapped light in the NOA63 film due to the total internal reflection at the NOA63/air interface. While in case of the microstructured NOA63 surface, the micropillars array with tapered morphology results in a gradual change of the refractive index. This transition of effective refractive index is realized by changing material index from NOA63 to air and it is crucial for the film transmission properties. Owing to the gradual change refractive index, the critical angle would be enlarged. Therefore, the light loss of the total reflection could be lowered effectively by modifying the critical angle.

The electroluminescent (EL) performances of the FTOLEDs are investigated. In order to verify the effect of the structured encapsulation film on light extraction of the FTOLEDs, the FTOLEDs without any encapsulation and with a planar encapsulation film are fabricated for comparison. Their schematic structures are shown in Fig. 2(a), and their EL spectra, current density–voltage (J–V) and luminance-J-efficiency characteristics are summarized in Fig. 2(b) and (c) respectively. They exhibit almost identical EL spectra and J–V curves, due to their same device structure. While the luminance and current efficiency show significant difference. The FTOLEDs with the microstructured encapsulating layer show highest luminance and current efficiency. The maximum current efficiency is 12.36 cd/A, which corresponds to an enhancement factor of 16% to that of the FTOLEDs with the planar encapsulating layer (10.64 cd/A) and 33.5% to that of the FTOLEDs without the encapsulation (9.26 cd/A), respectively. This enhancement is originated from the improved light extraction induced by the microstructure. In case of the planar polymer film encapsulation on top of the FTOLEDs, the capping layer acts as a refractive index-matching layer and shows an effect in improving the light transmittance, which results in a higher efficiency compared to that of the unencapsulated FTOLEDs. However, the total reflection at the interface between the encapsulation film and air results in a power lost. The efficiency can be further improved by recovering this reflection lost for the FTOLEDs with

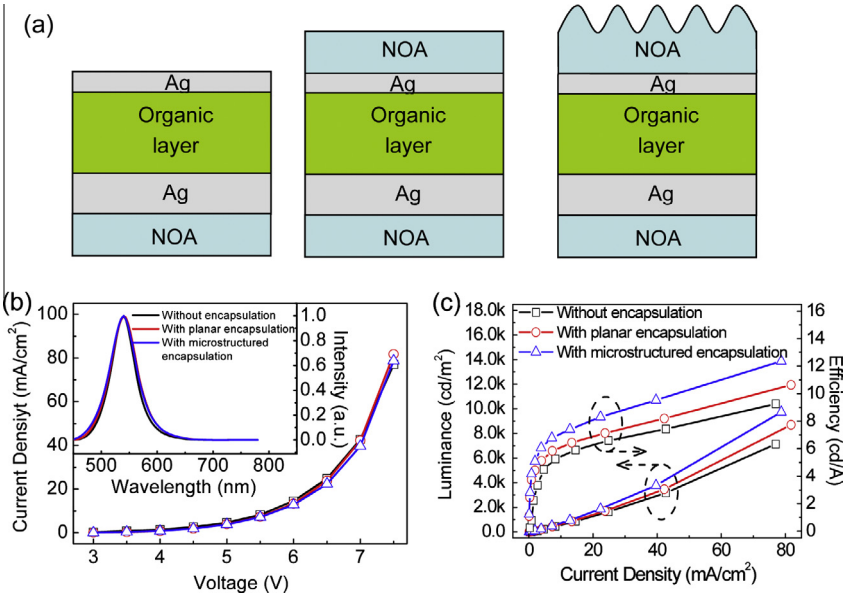

Fig. 2. The structure of the TOLEDs with and without the encapsulation (a), and their current density–voltage (b), and luminance–current density–efficiency (c) curves. Inset in (b): EL spectra of the TOLEDs with and without the encapsulation.

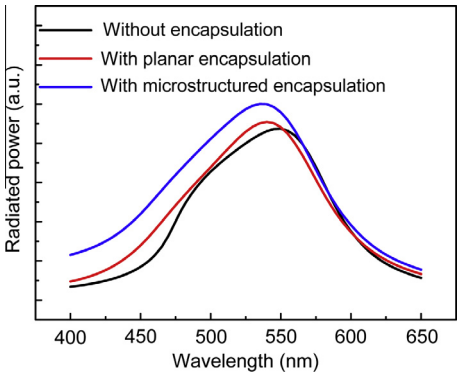

Fig. 3. Simulated radiated power of TOLEDs with and without the encapsulation.

the microstructured encapsulating layer. The gradient in refractive index between air and the surface of the encapsulation film with the micro pillars array will dramatically suppress the reflection loss and results in the highest efficiency for the microstructured FTOLEDs.

To gain a supporting evidences of the effect of the microstructured encapsulating polymer film on the optical characteristics of TOLEDs, the radiated power of above-mentioned three devices was calculated by exploiting the well established classical optical model treating the emissive dipoles embedded in the active layer as forced damped harmonic oscillators [39]. The simulated radiated powers for the three devices show agreement with that of the experimentally measured efficiency as can be seen in Fig. 3. The enhancement of radiated power for the FTOLEDs with the planar encapsulation to that of the devices without any encapsulation indicate that the power loss at semi-transparent top Ag contact has been successfully recovered as emission light in far field for the TOLEDs. While the further enhancement of the radiated power for the FTOLED with the microstructured encapsulation is attributed to the recover of the power lost to the total reflection at the interface of the NOA63/air.

The photographs of the FTOLEDs with the microstructured encapsulation are shown in Fig. 4. The structured surface shows orange color due to the scattering and diffraction of the microstructure, which are clearly observed by the naked human eyes as shown in Fig. 4(a). Fig. 4(b) and (c) show the operating devices under a driving voltage of 6 V. The emitting area is uniform and there is no obvious color variation for the emission from the FTOLEDs with the planar or structured encapsulation. The flexibility of devices was demonstrated in Fig. 4(c). The operating device is free of cracks and dark spots even under a small bending radius. The substrate and encapsulation layer

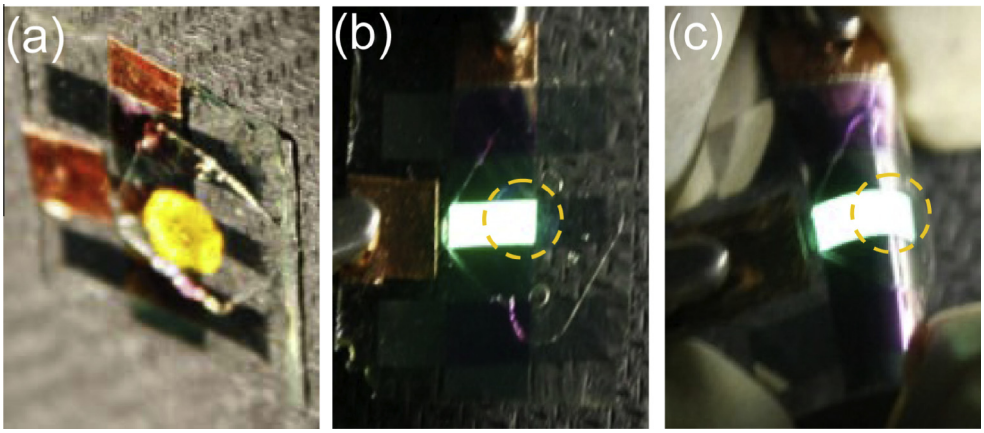

Fig. 4. The photographs of TOLEDs with the microstructured encapsulation at planar and bending situation.

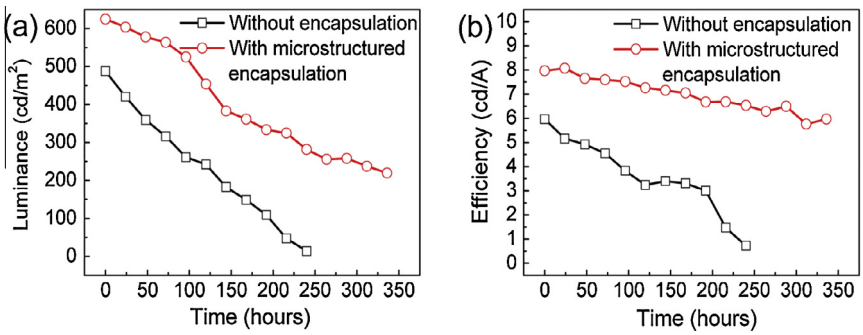

Fig. 5. The stability of the TOLEDs with and without the encapsulation.

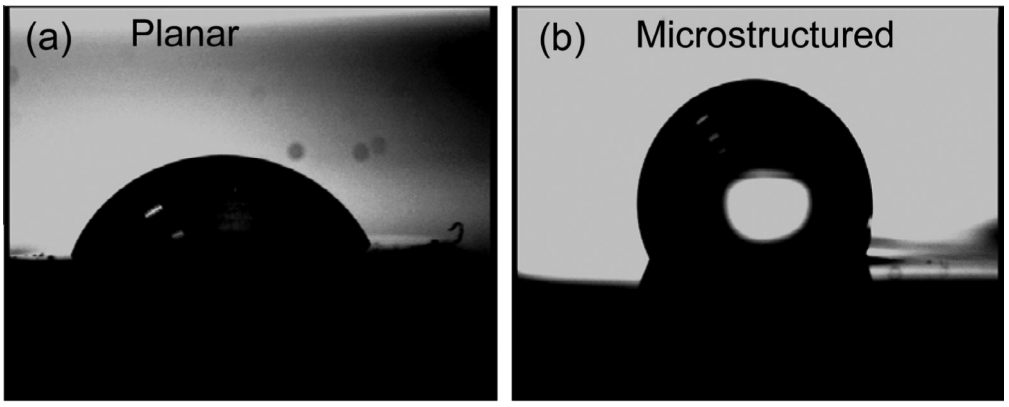

Fig. 6. Contact angle of the encapsulation film with and without the microstructure.

were both NOA63 film and they can adhere well with each other, which results in the high flexibility of the encapsulated FTOLEDs. Moreover, effective protection from water vapor and oxygen diffusion by the good adhesion between the substrate and the encapsulating films is expectable. Luminance-time and efficiency-time curves of the FTOLEDs with and without the encapsulation are shown in Fig. 5. The decay rate of devices with the encapsulation is obviously slowed down. These results show that the encapsulation by employing the NOA63 effectively prolong their operational lifetimes.

On the other hand, the microstructured encapsulation film exhibits hydrophobic and self-cleaning ability. Generally, the surface with a contact angle (CA) of larger than 90° is defined as a hydrophobic surface, which is determined by surface micro/nanostructure and surface energy. Fig. 6 presents the CA of the planar and microstructured NOA films. The microstructured polymer surface exhibited a high CA of 123.23°, while it is only 65.49° for that of the planar surface. The high CA corresponds to a hydrophobic property, which would reduce the probability of water condensation and dust accumulation.

4. Conclusions

In summary, we have demonstrated FTOLEDs with enhanced light extraction and self-cleaning ability by integrating a microstructure with 2D tapered micropillars array on the polymer encapsulation film. The simulated and experimental results both confirm the enhancement of the light extraction by recovering the light loss induced by the total reflection at the encapsulation film and air interface. The efficiency of the FTOLEDs is increased from 9.26 cd/A for the uncapsulated devices to 12.36 cd/A for the device with the microstructured encapsulation. The encapsulated FTOLEDs maintain excellent flexibility and improved lifetime by using the NOA63 polymer as both substrate and encapsulation layer and permitting a good adhesion to each other. Moreover, the hydrophobic property of the microstructured encapsulation film is beneficial to prevent the FTOLEDs from being polluted by water droplets and dust particles in practical applications. This encapsulation technology may provide a promising strategy for improving the efficiency of the FTOLEDs and maintaining its high flexibility.

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