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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 for being polluted by water droplets and dust particles in practical applications.

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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

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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. MoO₃ 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 (Alq₃) was used as emitting layer. The structure of the TOLEDs is Ag (80 nm)/MoO₃



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

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