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Direct formation of random wrinkle on emission surface for improved light out coupling and stable angular spectrum of white organic light emitting diodes

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ABSTRACT

To enhance the light out coupling and stabilize the angular spectrum of white organic light emitting diodes (WOLEDs), we formed a randomly distributed corrugation directly on the external glass surface. Using a metallic master, which was prepared using laser ablation, we have imprinted the corrugation pattern directly on the glass surface. Our method yields 32% increase in the external quantum efficiency, widened luminance distribution and low angular spectra variation. Considering WOLED as a lighting source, this feature is particularly appealing. Besides the implications of the spectrally stable efficient WOLEDs through random corrugation, our work suggests a new structural approach for various light applications in which efficiency and spectral stability matter.

1. Introduction

Organic light emitting diodes (OLED) have been desired in displays and lighting applications because of low power consumption, high color quality, fast response time and flexibility. Especially white organic light emitting diodes (WOLED) have been regarded as promising candidates for next generation solid state lighting source. Despite of these excellent characteristics, the troublesome issues related to light extraction efficiency in devices have been still remained.

In conventional OLEDs, the differences of the refractive indices between their constituting layers cause poor light extraction efficiency. Even though the use of phosphor materials in white OLEDs allows 100% internal quantum efficiency, the out-coupling efficiency is approximately 20% [1]. Because some created photons in organic layers cannot pass through the device and they are trapped in indium tin oxide (ITO)/organic layer ($n_{ITO,\ organic}$ = 1.7–2) as a waveguide mode. In addition another fraction of the generated light is dissipated by total internal reflection at air (n_{air} = 1)/glass substrate interface. The surface plasmon-polaritons (SPP) mode taking place at the metal/organic interface is causing photon loss [2]. Finally the limited light out coupling of OLEDs not only demerits the power consumption but also shortens the device life time [3]. Therefore, many approaches have been researched to minimize loss of light out coupling efficiency such as modifying of substrate surface, inserting scattering medium and low index medium, employing micro cavity structure, etc [4–8]. Particularly, introduction of microlens array (MLA) on glass substrates has improved the out-coupling efficiency of WOLED. However, the distortion of white EL spectrum depending on viewing angle has not resolved [9,10].

In this study, we demonstrated improvement in light out coupling and EL spectral stability of bottom emissive WOLEDs with wrinkle polymeric light extraction film (WPF). External substrate modifications for light extraction have been proposed but most methods rely on rather complex processes. Here we suggest simple, cost-effective, reproducible and powerful imprinting technology by directly forming wrinkle structure on WOLED. We have formed directly wrinkle structure on emissive surface of WOLED using a corrugated stainless steel master and investigated of effect on the performance of the WOLED.

2. Experimental section

To prepare corrugated master, a polished stainless steel (SUS 304) substrate was ablated using a laser processing. The corrugated pattern was designed to have a height and a ridge width of 5 μm and 20 μm,

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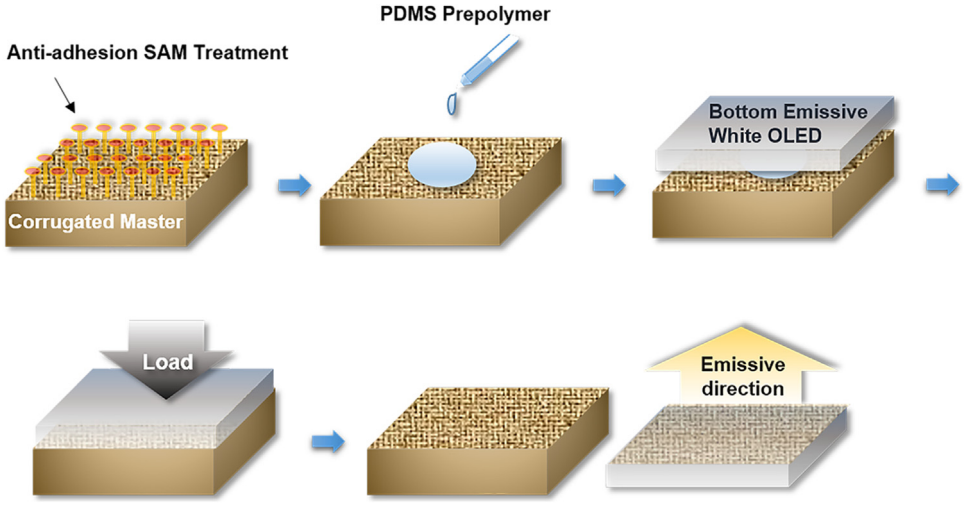

Fig. 1. Fabrication process of random wrinkle polymeric film (WPF) on emissive surface of WOLED.

respectively, using computer-aided design program. The distribution was intentionally designed to be random. The randomness may contribute to eliminate the detrimental angular dependency of emission spectra. These values are based on our previous work [11]. A Nd:YAG laser source was used with average power of 8 W and a repetition rate of 100 Hz. A substrate surface area of 20 × 20 mm^2 was scanned at a scanning speed of 200 mm/s. The wavelength of laser beam was 335 nm in ultraviolet range allowing higher energy absorption into metal and focusing onto the smaller beam spot. In order to control the laser power a half-wave plate and a polarizer were used and the horizontally polarized beam was focused by a telecentric scan lens which have a maximum scan area of 64 × 64 mm^2 (Sill Optics S4LFT4100/075). The beam was manipulated over the sample by a two-mirror galvoscanner (SCANLAB Hurry SCAN 14). The focused beam size was 16 μm and its working distance was 103 mm. The experimental procedure of laser ablation processing we used has been described elsewhere in detail [12].

Fig. 1 shows the overall process of fabricating wrinkle polymeric film (WPF) for light extraction. In the first step, a polydimethylsiloxane (PDMS) as a WPF was prepared by mixing Sylgard 184 and its curing agent (Dow Corning, USA**).** The PDMS prepolymer was placed in vacuum desiccator for 30 min to remove the bubbles inside before use. To facilitate the detachment a self-assembled monolayer (SAM) was formed on the corrugated surface of master using trichloro(1*H*, 1*H*, 2*H*, 2*H*-perfluorooctyle)silane (FOTS) as a precursor [13]. Briefly, the corrugated stainless sample was treated with UV ozone and H_2O vapor to achieve a hydrophilic surface. Then FOTS was evaporated at 160 °C. This process gives a fluorine rich SAM terminal, which is highly hydrophobic.

Subsequently, prepared PDMS prepolymer whose bubbles were completely removed was dropped onto the master and the emissive surface of WOLED covered above. In order to transfer the pattern accurately with a good step coverage, a load of 3 kg was applied during the curing period. Samples were cured for 48 h at a room temperature to avoid thermal damage in the organic layers of device. And then the master was removed from completely cured PDMS film. Finally, the inverted wrinkled patterns were transferred onto the glass substrate of WOLED.

In order to evaluate the out coupling capacity of our film, we have fabricated bottom emissive WOLEDs. All organic layers and cathode were deposited by thermal evaporation method. The whole structure of our WOLED was as follows; Indium tin oxide (70 nm)/1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile (HAT-CN, 5 nm)/1,1-bis[(di-4-tolyamino)phenyl)]cyclohexane (TAPC, 50 nm)/HAT-CN (10 nm)/TAPC (50 nm)/HAT-CN (10 nm)/TAPC (45 nm) (total HTL thickness: 170 nm)/4,4 ´,4 ´´-tris(N-carbazolyl)-triphenylamine (TCTA): bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl)iridium(III) (FIrpic, 7%) (5 nm)/TCTA: bis(2-methyldibenzo[*f*,*h*]quinoxaline)(acetylacetonate)iridium (III) ($Ir(MDQ)_2(acac)$, 5%) (0.5 nm)/ TCTA: tris(2-phenyl-pyridine)iridium(III) ($Ir(ppy)_3$, 7%) (1 nm)/2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy): FIrpic (10%) (5 nm)/ 1,3-bis (3,5- dipyride-3-yl-phenyl) benzene (BmPyPB, 55 nm)/LiF (1 nm)/Al (100 nm). The base pressure was maintained below 1.3 × 10^{-6} Pa before deposition. The alternating layers of n-HAT-CN and TAPC organic materials were adopted for a HTL which has demonstrated in our previous report that improve the electrical properties and luminance without distortion of the EL spectrum [14]. Our WOLED was realized by three color-type of phosphorescent emitter. To generate white light, the green (TCTA-$Ir(ppy)_3$) and red (TCTA-$Ir(MDQ)_2(acac)$) emissive layers were sandwiched between blue emitting layers. This structure is effective to extend the lifetime of generated triplet excitons, which could result from the effective diffusion of recombination in HTL and ETL hosts and be subsequently redistributed between the blue, green and red dopants correspond to their concentrations in these hosts and their distance from the TCTA/26DCzPPy interface [14–16]. This energy level design of stacked layers could affect to enhance light extraction of OLED, therefore, it must be taken into account. Fig. 2(b) shows the energy alignment diagram of our WOLEDs. All WOLEDs were encapsulated in glass in nitrogen glove box to prevent degradation caused by oxygen and moisture in the air.

The morphologies of wrinkle structure were examined using scanning electron spectroscopy (FE-SEM, Sirion 400). The optical properties of WPF were measured via UV–visible spectroscopy (Lambda 750 Spectrometer, Perkin Elmer) and a haze meter (Haze-gard plus, BYK). In order to investigate the influence of directly formed WPF, we performed the current density-voltage-luminance (JVL) characteristics which were collected from with a current-voltage source unit (Keithley 238) and spectroradiometer (CS-2000, Minolta). The angular distribution and electroluminescence (EL) spectra were obtained via spectroradiometer (CS-2000, Minolta) equipped on a goniometer at a constant current density of 2 mA/cm^2.

3. Results & discussion

Fig. 3(a) to (b) show SEM images of the wrinkle structure on the surface of corrugated master made by laser machining. The wrinkle structure was randomly textured and has approximately 20 μm-spacing between their ridges. The presences of spots between ridges are due to the non-continuous laser ablation process. Fig. 3(c) to (d) show SEM images of the WPF directly formed onto the glass substrate of WOLED.