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Corrugated structure through a spin-coating process for enhanced light extraction from organic light-emitting diodes

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ABSTRACT

We have demonstrated a simple fabrication method for an out-coupling structure to enhance light extraction from organic light-emitting diodes (OLEDs). Spin-coating of SiO_2 and TiO_x sol mixture solution develops corrugated film. The structural evolution of the corrugation was explained by the localization of surface tension during the solvent evaporation. The structural parameters of the corrugated structure were characterized by varying the spin-coating speed and the mixing ratio of the solution. Compared to conventional devices, OLEDs with a corrugated structure at the backside of the glass substrate showed increased external quantum efficiency without change in the electroluminescence spectrum. The light extraction enhancement is attributed to the decreased incidence angle at the interface of glass substrate and air.

1. Introduction

Organic light-emitting diodes (OLEDs) have attracted a lot of interest for use in displays and interior lighting due to their promising advantages of low power consumption, high power efficiency, high contrast ratio, and high speed of operation [1,2]. Recently, light extraction from OLEDs has been considered as one of the most important issues for research since less than 20% of the light generated from the emissive layer of a conventionally structured OLED can escape from the device as useful radiation [3,4]. However, most of the generated light is trapped at the interface between the organics/indium tin oxide (ITO) layers ($n_{ITO,organic}$ = 1.7–2) and the glass substrate ($n_{glass} \approx 1.5$) or the glass substrate and air (n_{air} = 1) by total internal reflection due to the multiple layered structures with different refractive indices. Therefore, out-coupling of the trapped light is in great demand for high efficiency OLEDs. In order to extract the light confined inside the devices for high external quantum efficiency, defined as the ratio of the number of emitted photons into air to the number of injected electrons, several out-coupling structures have been studied by modifying the shape inside [5–9] and outside the device [10,11]. Some research groups modified the emitting layer to have a light scattering medium inside the film [12] or to have the orientation of the transition dipole moments of organic emitters [13].

Rapid progress has been made in fabrication of out-coupling structures for light extraction from OLEDs; however, those methods require complex and expensive equipment

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with a vacuum system or complicated lithographic process. Considering practical applications, it is essential that fabrication techniques are applicable to a large area and are cost effective, so it is desirable to develop a simple fabrication technique for out-coupling structures. For this purpose, several groups have utilized imprint lithography to fabricate out-coupling structures for enhancing light extraction from OLEDs [14–16]. Although imprint lithography has the advantage of being a vacuum-free process and is easy to fabricate, additional complex steps are still needed to prepare a template for the patterned mold.

In this study, we developed a simple fabrication technique for out-coupling structures without using a mold or vacuum process to enhance the light extraction efficiency of OLEDs. A corrugated structure, as a result of the spatial distribution of the surface tension, can be prepared quickly by a spin-coating process with a small amount of a mixture of SiO_2 and TiO_x sol solution. The width and the height of the corrugation were controlled by the variation of the spin-coating speed and the mixing ratio between two sol solutions. Using standard small molecular emitters, OLEDs with and without the corrugated structure were fabricated, and their device performances were compared. With the introduction of the corrugated structure at the backside glass substrate of the devices, the light trapped from total internal reflection at the interface between the glass substrate and air can be extracted and the out-coupling efficiency is increased.

2. Experimental

2.1. Preparation of SiO_2 and TiO_x sol solutions and their mixture

To prepare the SiO_2 sol solution [17], 0.77 ml of tetraethyl orthosilicate (TEOS), 3.5 ml of (3-glycidoxypropyl) trimethoxysilane (GPTMS), 1.2 ml of methanol, 0.065 ml of acetic acid, and 1 ml of deionized water were mixed by stirring at room temperature for 3 h. Then, 2.6 ml of methanol and 1.9 ml of 2-methoxy ethanol were added to the solution. To prepare the TiO_x sol solution [18], 4.3 ml of titanium tetra isopropoxide (TTIP) was dropped into 5.1 ml of methanol and 0.76 ml of glacial acetic acid was then added to the solution at room temperature. After 30 min, 0.2 ml of deionized water was dropped into the solution and the reaction proceeded for 24 h. At the wavelength of 500 nm, the refractive indices of the SiO_2 layer and TiO_x layer formed with pure sol solutions (annealed at 70 °C) are 1.53 and 1.77, respectively. The SiO_2 and TiO_x sol solutions were mixed with the volume ratios of 35:65, 40:60, and 45:55 and kept in a vortex mixer for 1 h.

2.2. Fabrication of corrugated structure and characterization

The backsides of ITO-coated glass (1 in. × 1 in.) were cleaned and treated with oxygen plasma for 10 min. The mixture solution was dropped (about 100 μl) and spin-coated on the backside of the ITO-coated glass at various spin-coating speeds of 4000, 4500, and 5000 rpm for 30 s. Then, the samples were kept in a convection oven at 70 °C for 10 min. Morphology of the corrugated structure and surface profiles were analyzed with an optical microscope (Nikon, L150) and a Veeco Dektak-8 surface profiler (stylus force, 15 mg; stylus tip radius, 2.5 μm).

2.3. Fabrication of OLEDs and measurement

The ITO surface of substrates with and without the corrugated structure were treated with UV–ozone for 10 min and the following organic layers were deposited by thermal evaporation at a pressure of <10^{-6} Torr: N,N′-di(naphthalene-1-yl)-N,N′-diphenylbenzindine (NPB, 40 nm)/Tris (8-hydroxy quinolinato)aluminum (Alq_3, 50 nm)/lithium fluoride (LiF, 0.5 nm)/aluminum (Al, 100 nm). NPB, Alq_3, LiF, and Al were used as a hole transporting layer, emissive layer, electron injection layer, and a cathode, respectively. The emissive area of the devices is 2 × 2 mm^2. The current density–voltage–luminance characteristics and the electroluminescence (EL) spectra of the fabricated OLEDs were measured with a Keithley 2400 source measurement unit and a Minolta CS 2000 spectroradiometer.

3. Results and discussion

Fig. 1 contains optical microscope (OM) images of the corrugated structure fabricated by spin-coating a mixture of SiO_2 and TiO_x sol solutions at various spin-coating speeds and several blending ratios. The images show that one-dimensional corrugation is formed along the radial spreading direction of the mixed solution by spin-coating. Fig. 1a–c compare OM images of the corrugated structure fabricated at spin-coating speeds of 5000, 4500, and 4000 rpm and a fixed mixing ratio of 40:60 (SiO_2:TiO_x sol solution, vol./vol.). The OM images were taken at the same radial distance (1 cm) from the center of the glass substrate, where the vertical direction is the radial spreading direction of the mixture solution. Compared to the corrugated structure formed with a spin-coating speed of 5000 rpm, one fabricated with a spin-coating speed of 4500 rpm shows thicker corrugation, and the corrugation formed with a spin-coating speed of 4000 rpm shows even thicker corrugation. These observations indicate that the corrugated structure fabricated with a higher spin-coating speed shows finer corrugation than that formed with a lower spin-coating speed. Fig. 1a, d, and e compare OM images of the corrugated structure fabricated with mixing ratios of 40:60, 45:55, and 35:65 and a fixed spin-coating speed of 5000 rpm. Compared to the corrugated structure fabricated with a mixing ratio of 45:55, the sample formed with a smaller mixing ratio of 40:60 shows thinner corrugation. The corrugated structure formed with a mixing ratio of 35:65 appears almost two-dimensional. The results reveal that finer corrugation can be obtained by using a solution mixture with a smaller mixing ratio of the SiO_2 sol solution as well as spin-coating at a higher spin-coating speed.

The corrugated structure is thickness variation occurred during spin-coating process, which is called striation [19,20]. The thickness variation in spin-coated films has been observed in the sol–gel spin-coating technique