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# Organic Light Emitting Diode with Uniform Luminance Distribution and Enhanced Efficiency via Random Embossing Structure

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Using the lateral phases separation of a binary immiscible polymer blend, we have fabricated a random embossing structure (RES). The RES was investigated as a potential structure for organic light-emitting diode (OLED) light extraction. Our RES yields uniform light extraction in a broad angle (0°∼70°) range with spectral stability. Blue phosphorescence OLEDs equipped with the RES showed an enhancement of 20% in external quantum efficiency (EQE)and spectral stability within $\Delta x,y < 0.01$ in a viewing angle range of 0°∼70°. Our RES provides a new and practical approach for achieving enhanced efficiency and spectral stability of OLEDs.
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For both energy saving and maximization of device life-time, it is important to enhance the external quantum efficiency (EQE) of organic light emitting diodes (OLEDs). Thanks to recent advances and optimization of organic materials, stack structures and electrode designs, the internal quantum efficiency of OLEDs is near 100%.[1–4] However, due to the presence of waveguide modes and surface plasmon polariton losses, the total out coupling efficiency of planar OLEDs is limited to ∼20%.[5]

Methods for recovering light are called light extraction techniques. Various light extraction methods have been suggested. Examples include scattering wave-guided mode,[6,7] removal of surface plasmon between organic and cathode through internal nanostructures,[8,9] and refracting substrate mode using microlens arrays (MLAs).[10–12] In this work, we suggest a method for extracting the light confined at the substrate/air interface. For OLED lighting applications, it is desired to have a structure which cannot only enhance luminance over a broad range but also induce spectral stability. Conventionally, MLAs have been used to extract this light. While effective in reducing the substrate mode, MLAs have a drawback of enhancing the luminance mainly in the normal direction.[13] Here, we suggest and demonstrate a structure which is achieves efficiency enhancement and spectral stability. Our structure is based on the lateral phase separation of two immiscible polymers. Due to the spontaneous nature of the phase separation, our method does not require patterning or photolithography processes.

Binary immiscible polymers can be dissolved in a common solvent. Using spin-coating techniques, it is possible to form thin polymer films which contain immiscible components. During the solvent removal process immiscible polymers undergo demixing or phase separation.[14–16] The phase separation may be guided using templates to control the separation alignment.[17] In the demixing of binary polymers, one component is observed to be finely dispersed in another. This phase separation state is reminiscent of a surfactant-stabilized two-phase solution. Heriot and Jones have used small-angle light-scattering methods to investigate the underlying principles involved in the formation of such structures.[18] Based on the spectrum data, they draw the conclusion that the lateral phase separation is essentially due to an amplified capillary wave, which is responsible for the instability or phase separation. From processing perspectives, the distributions of separated components may be changed by modifying the relative concentration of polymers, spin-coating conditions and thermal processes. Among the phase separation systems, we selected polystyrene (PS) and polyethylene glycol (PEG) blended in toluene. By selective removing laterally separated PEG components and transferring the patterns, an external light extraction structure which can enhance the out coupling efficiency throughout the whole angle range has been obtained.

## Experimental

PS of $M_w = 280{,}000$ g/mol and PEG of $M_w = 1{,}000$ g/mol were used in this work. Both polymers were purchased from Sigma-Aldrich and used without additional purification. The PS and PEG were dissolved in a common solvent of toluene. The PS: PEG ratio was 73:27, and the total polymer concentration in the solvent was approximately 8%. The mixture was stirred for 24 hours at room temperature, after which, this solution was spin-coated on glass substrates. The samples were annealed for 2 hours at 110°C in an air atmosphere to remove the solvent component and obtain polymer thin films. The aforementioned process yielded a polymer thin film in which the phase-separated PEG is finely dispersed in PS. The PEG component was selectively removed by rinsing the samples in de-ionized water. The film pattern was transferred on a glass index matching adhesive plastic film by using UV curable polymer (Ormocomp, Micro Resist Technology). In the pattern transfer process, a uniform pressure of 12 kPa was applied at room temperature. Fig. 1 illustrates the fabrication of the phase-separation film and transfer of the pattern to adhesive plastic film. To evaluate the light extraction capacity of the transferred phase-separation film, we fabricated blue phosphorescence OLEDs. All organic layers were deposited in a high vacuum chamber below $6.67 \times 10^{-5}$ Pa by a thermal evaporation method. Our OLEDs has a stack structure of indium tin oxide [ITO] (70nm)/1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile [HAT-CN] (5nm)/4,4′-cyclohexylidenebis[*N*,*N*-bis(4-methylphenyl)benzenamine] [TAPC]

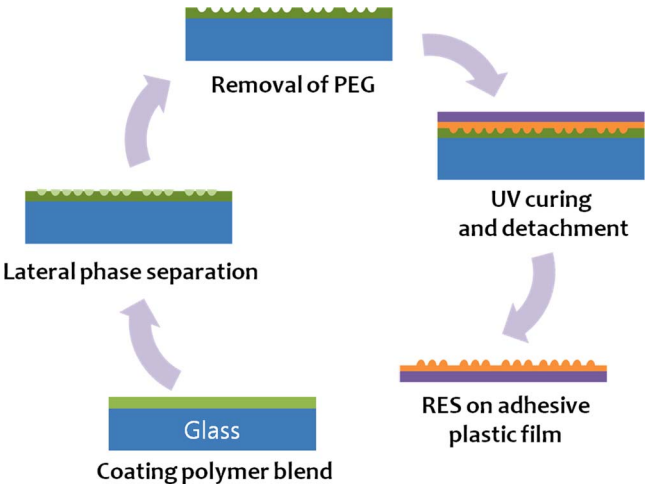


**Figure 1.** The fabrication of the phase separation film and transferring the pattern to an adhesive plastic film.

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(50nm)/HAT-CN (5nm)/TAPC (50nm)/2,6-bis-[3-(carbazol-9-yl)phenyl]pyridine [DCzPPy] doped with 10% of Bis[2-(4,6-difluorophenyl)pyridinato-$C^2$,*N*](picolinato)iridium(III) [FIrpic] (20nm)/1,3,5-Tri[(3-Pyridyl)-Phen-3-Yl]Benzene [TmPyPB] (10 nm)/5% of Li doped TmPyPB (45 nm)/LiF(1 nm)/Al (100 nm). To achieve high hole injection and electrical stability, we used a hole transport layer (HTL), in which HAT-CN and TAPC were alternating. The OLED characteristics using such an HTL have been reported elsewhere.[19] Because blue is a rather weak color in OLEDs, we intentionally choose blue phosphorescence as the emissive layer to enhance its low efficiency. To protect the organics from atmospheric degradation, the fabricated OLEDs were glass encapsulated in a glove box. The emitting area was 70 $mm^2$ (10 mm × 7 mm). The RES on the adhesive plastic film was attached to the emission surface of the fabricated OLEDs. Characteristics of OLEDs with planar and MLA were also measured and compared with the RES OLEDs. All OLEDs were fabricated in one identical batch and measured under same conditions. The current density (*J*) – voltage (*V*) characteristics of the devices were measured using a Keithley 238. Their angular spectra and luminance distributions were measured by a goniometer-equipped spectroradiometer (Minolta CS-2000).

## Results and Discussion

The surface morphologies of the samples were observed using a scanning electron microscope (SEM, Sirion FEI). By the selective removal of the PEG sites of the phase-separation film, a random hole array is formed (Fig. 2a). Fig. 2a shows that the phase separation of the PEG/PS system takes place in a lateral fashion in the thin film regime.[20] The relatively narrow distribution of the hole diameter indicates that the initial local fluctuation in capillary waves does not grow significantly during the separation process. Randomness is thought to have its origin in the liquid phase separation, in which preferences in specific crystallographic interfaces are absent. The RES was obtained by transferring random hole array patterns (Fig. 2b). Atomic force microscopy (AFM, Model: Park System, XE-100) scans showed bump diameters and heights in the ranges of 1∼2 μm and 300∼400 nm, respectively. Commercial MLA (MIRAENANOTECH Co.) was used to contrast our RES (Fig. 2d). The MLA used in this study consisted of hemispheres in a sequence of *ABAB*. The diameter was around ∼80 μm and the fill factor was near 100%. The results shown in Figs. 2a to 2c demonstrate that our transfer process is highly accurate without dimensional distortion of the original shape.

Fig. 3a shows the normal *J-V-L* characteristics of the OLED with RES and MLA and planar device. The *J-V* characteristics of the devices are almost the same. This was expected because the light extraction film is located at the external surface of the OLEDs. The *V-L* characteristics show that the normal luminance of the OLED with MLA is higher than that of the OLED with RES and the planar OLED. This is due to the selective luminance enhancement in the normal direction of the MLA. As mentioned above, the light distribution of OLEDs with a typical hemispheric MLA is concentrated

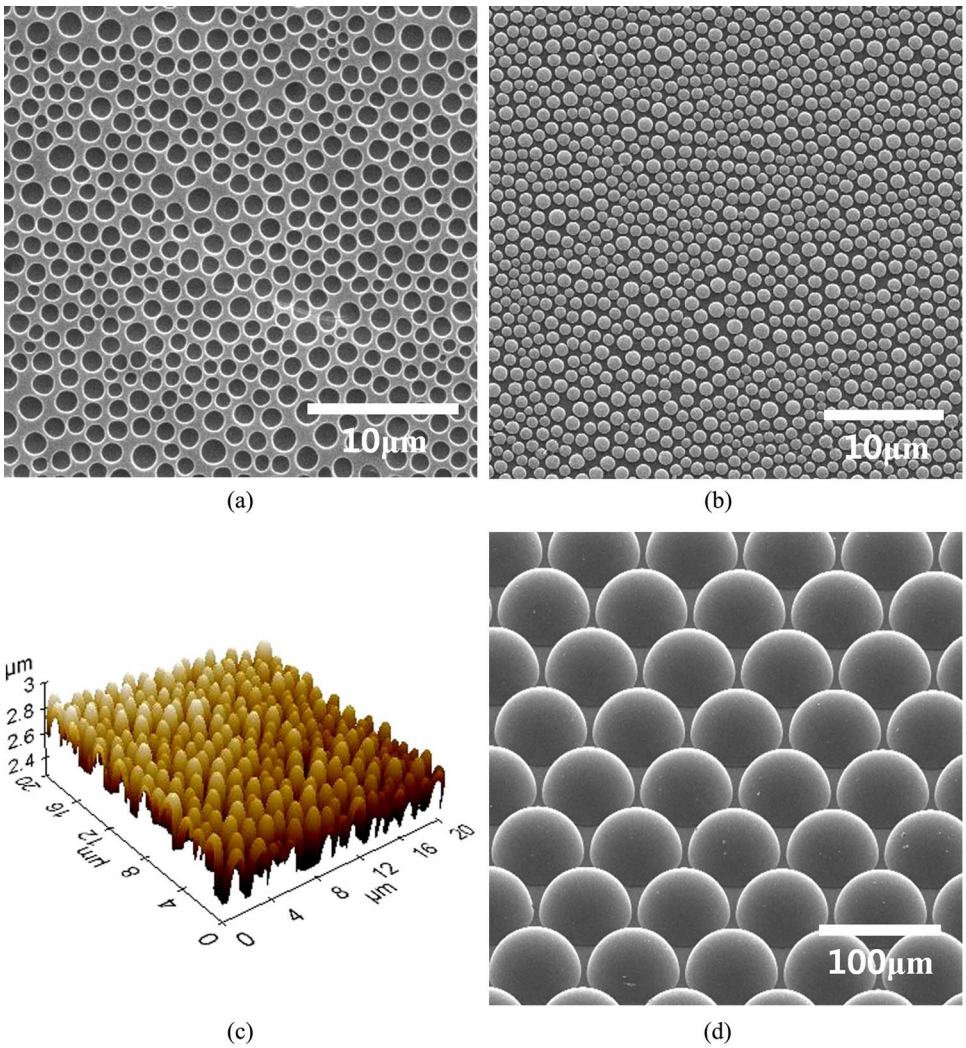
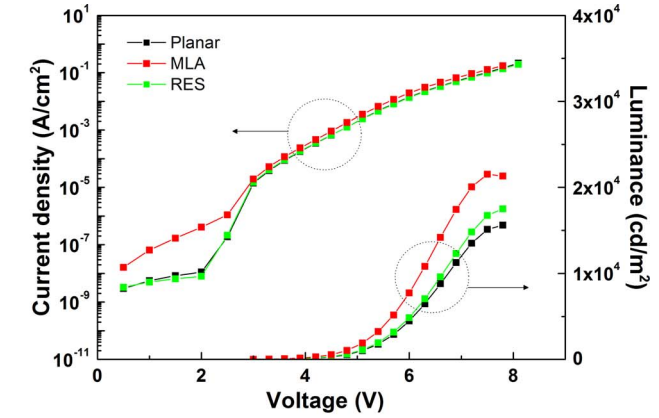
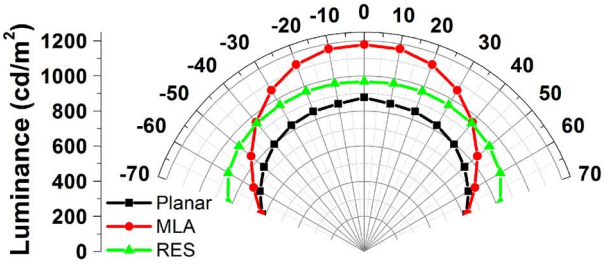


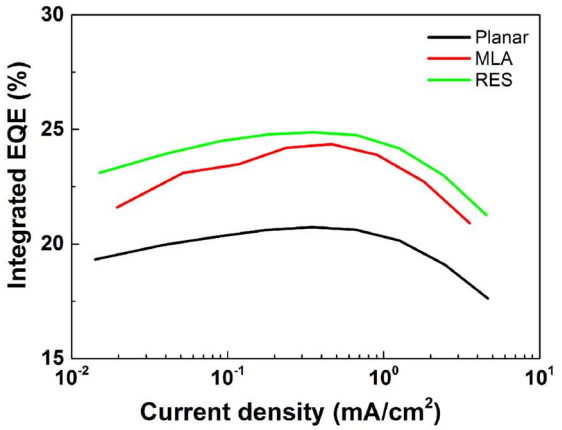

**Figure 2.** SEM images of (a) phase separation film, (b) RES, (c) AFM image of RES and (d) MLA.




(a)




(b)




(c)

**Figure 3.** (a) The current density-voltage-luminance characteristics, (b) the luminance distribution of the devices at current density level of 2.0mA/cm$^2$ with RES, MLA and planar OLEDs and (c) integrated EQE.

in the normal direction. Fig. 3b shows the luminance distribution of the devices at a constant current density level of 2.0 mA/cm$^2$. The luminance distribution of the OLED with MLA is mainly extended in the normal direction. Light extraction using an MLA is strongly influenced by the size and aspect ratio of lenses.[13,21,22] S. Lee et al. reported strong luminance concentration in the normal direction with the use of several hundred micrometer-sized MLAs.[13] Sun and Forrest reported the absence of preferential enhancement in the normal direction with the use of a periodic MLA of several micrometers. This result was interpreted as the effect of the small size of the MLA. However, the MLA aspect ratio also affects the outcoupling efficiency of OLEDs.[22] For diameters bigger than 5μm, outcoupling efficiency gradually increases as the aspect ratio increases. According to the simulation results, the outcoupling enhancement using an MLA becomes saturated as the diameter of the MLA reaches a certain size (∼5 μm), for a given aspect ratio (See Fig. 1(b) in Ref. 22). As the aspect ratio of the lenses decreases, normal direction efficiency also decreases. Instead, the enhancement in the high viewing angle became relatively large. Invoking this result, for a given size of MLA, the aspect ratio of the lenses turns out to be more important than MLA size. Our RES has an aspect ratio of approximately 0.6. This value is small compared to that of an MLA, which is typically 1. As seen in Fig. 3b, by using the RES it was possible to enhance the luminance over the whole angle range. This feature should be contrasted to the MLA case, in which enhancement took place preferentially in the normal direction. The uniform angular enhancement by using the RES is thought to be due to the low aspect ratio. When the feature size exceeds 50 μm, which is typical for an MLA, the substrate mode is extracted by refraction at the MLA/air interface. The diameter and height of our RES were 1∼2 μm and 300∼400 nm, respectively. In this size range, visible light cannot only refract but also scatter. Thus, while it is not easy to pinpoint definitely, it seems that both refractive and scattering mechanism are operative. We plan to investigate this problem by optical simulations.

To evaluate the efficiencies correctly, we have used the luminance angular distributions. Previous works have only considered the normal incidence in obtaining efficiencies, which is only correct for the Lambertian distribution.[13,22] We calculated the integrated EQE over an angle range of 0°∼70°. The integrated EQE of the planar, MLA-OLED, and RES-OLED were 19.4%, 22.4% and 23.3%, respectively. The enhancement of the integrated EQE of the MLA and RES OLEDs were 15% and 20%, respectively. In the normal direction, use of the MLA is an effective method to extract light. However, when the distribution is considered, the RES OLED shows higher outcoupling efficiency than the MLA OLED. Though the diameter and height of RES are smaller than those of the typically used hemispheric MLA, the RES OLED shows better efficiency with uniform light distribution. Fig. 3c shows the integrated EQE versus current density, which is obtained as follows. The ratio of the integrated EQE and the normal direction EQE at the current density of 2.0 mA/cm$^2$ is calculated. This ratio is applied to other current density levels. Then we can calculate the integrated EQE at each current density level. The RES OLED shows a higher integrated EQE overall current density level.

Fig. 4a to 4c shows the normalized angular EL spectra of the devices. For lighting, angular spectrum distortion needs to be minimized because we see the reflected light from objects. There is no shift of the main peak. However, in the EL spectra of a planar OLED, an undesirable shoulder peaks develops as the viewing angle changes, which is due to the intrinsic microcavities existing in OLEDs. On the contrary, the shoulder peak of an OLED with MLA and RES are stabilized. The variance of shoulder peaks is reduced in case of MLA and RES OLEDs because the light from the cavity effect is diffused. Fig. 4d shows the 1931 CIE color coordinates with the emission angle. The coordinates of the planar OLED change as the viewing angle changes. The RES and MLA are observed to stabilize the coordinates. Compared to the MLA OLED case, the RES OLED shows better spectral stability at a higher viewing angle (>40°). This stable angular spectral characteristic is due to the light diffusion effect occurring at the interface of RES/air. Fig. 4 shows that both MLA and RES can suppress the dependency of EL spectra on viewing angle. However, due to the random distribution and the light diffusing effect, the CIE coordinates are more stable when RES is used. In conclusion, RES offers better light extraction and spectral stability than commercial MLA's. In particular, RES offers uniform enhancement over a wide angle range. We attribute the advantages to the random distribution of the RES. Our results suggest that, in a regime where diffraction and refraction overlaps, random distribution is very useful in achieving not only light extraction but also spectral stability over a broad angle range.

## Conclusions

We have demonstrated the RES as a structure for achieving uniform light extraction and spectral stability in OLED applications.

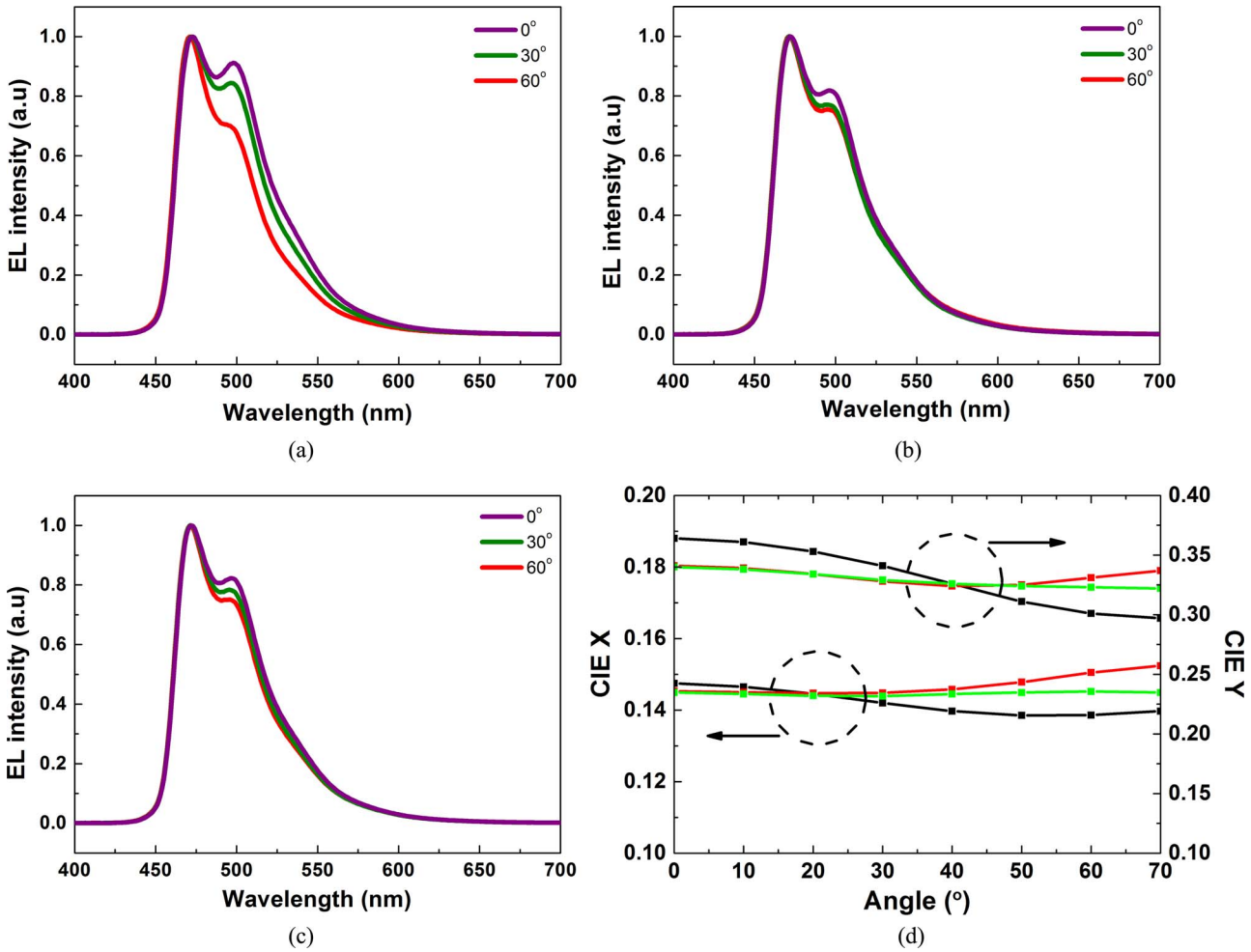



**Figure 4.** The angular EL spectra of (a) planar, (b) MLA and (c) RES OLED, (d) the color coordinates with the emission angle.

Phosphorescent blue OLEDs with RES show stable spectral characteristics and uniform light distribution as well as enhancement of the integrated EQE. Our fabrication of RES does not require a complex patterning fabrication method; thus, the proposed structure provides a practical approach for improving the performance of OLEDs and other existing light-emitting devices.

## Acknowledgment

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