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Application of optical property-enhancement film to improve efficiency and suppress angle dependence of top-emitting organic light-emitting diodes

Baeksang Sung[a*], Chul Woong Joo[b,c*], Jun Chang Yang[d*], Akpeko Gasonoo[e], Seung Wan Woo[a,b], Jae-Hyun Lee[a], Steve Park[d] and Jonghee Lee[a]

[a]Department of Creative Convergence Engineering, Hanbat National University, Daejeon, Republic of Korea; [b]Reality Display Research Section, Electronics and Telecommunications Research Institute (ETRI), Daejeon, Republic of Korea; [c]SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon, Republic of Korea; [d]Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon, Republic of Korea; [e]RIPE & 3D Printing, Hanbat National University, Daejeon, Republic of Korea

ABSTRACT
In this paper, we propose the application of an optical property-enhancement film that complements the angular dependence and total reflection of top-emitting organic light-emitting diodes (TEOLEDs). The optical property-enhancement film is composed of a porous pyramid arrangement applied on a thin-film encapsulation layer of TEOLEDs and is applied to distribute the transmitted light evenly. The results confirm that the change in the electroluminescence spectrum for each angle was effectively reduced because the TEOLEDs demonstrated uniform light distribution. In addition, reducing the total internal reflection in the film structure made it possible to improve the external quantum efficiency by approximately 35% and current efficiency by 38%.

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KEYWORDS
Organic light-emitting diodes; light extraction; pyramid structure; porous surface; patterning

1. Introduction

Organic light-emitting diodes (OLEDs) are rapidly occupying the market with unique advantages not found in other light sources. Appropriately fast response speeds, low power consumption, and self-luminous characteristics make these suitable for high-performance displays. The displays can also be configured in different forms to increase immersion because they are flexible. Moreover, their usage possibilities remain sufficient for application to new technologies such as Metaverse [1–5]. However, there are several considerations that must be respected to produce high-resolution OLED displays. High-resolution displays consist of highly integrated thin-film transistor (TFT) circuits. Therefore, it is difficult to use bottom-emitting OLED (BEOLED) structures with low aperture ratios [6–8]. Thus, top-emitting OLEDs (TEOLEDs) are essential in preventing the loss of aperture ratio. However, because the cathode of TEOLEDs is fabricated with a thin metal layer, part of the light is reflected. This has been identified as the main light loss path of TEOLEDs so consequently, different methods have been proposed [9–12]. The microcavity effect, a method using inter-electrode reflection, is a method of inducing light resonance by optimizing the distance between the semi-transparent cathode and reflective anode to the emission wavelength. Light resonance through countless reflections can pass through the cathode with strong intensity, contributing to the improvement of the current efficiency (CE) of TEOLEDs. Nevertheless, several problems have been associated with efficiency improvement using the microcavity effect. Owing to the luminance improvement centered on the front, low luminance was typically measured from the side, resulting in angular dependence where the emission color was also distorted. Consequently, light extraction technologies that suppress total reflection using a structure applied to BEOLEDs have also been introduced to TEOLEDs [13–16]. These methods, including BEOLED light extraction technology, can increase efficiency by reducing total reflection and can add a light scattering layer to suppress microcavity effects as well as reduce angular dependence [17–19]. However, unlike BEOLEDs

CONTACT Steve Park stevepark@kaist.ac.kr Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Republic of Korea; Jonghee Lee jonghee.lee@hanbat.ac.kr Department of Creative Convergence Engineering, Hanbat National University, Daejeon 34158, Republic of Korea
∗ These authors contributed equally to this work

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where a light extraction layer can be added directly to the substrate, a layer must be applied over the cathode of the TEOLEDs. This limits the device performance and could introduce stability issues. As such, there is a need for a method that can increase efficiency, maintain device stability, and reduce angular dependence.

In this study, we discuss an optical property-enhancement film that can improve the efficiency of TEOLEDs and suppress angular dependence. An optical film composed of a porous pyramidal arrangement effectively improves efficiency by extending the critical angle from the pyramidal structure of the TEOLEDs. The light transmitted through the porous surface is also evenly scattered and demonstrates light distribution characteristics close to Lambertian emission. These optical property-enhancement films applied on top of the thin film encapsulation (TFE) behave ideally without compromising device stability. The surface of the proposed optical property-enhancement film is prepared by distributing polystyrene (PS) beads on the Si mold of the intaglio pyramid and curing polydimethylsiloxane (PDMS). The film is then immersed in toluene to remove the PS beads. The complete removal of PS can be observed using scanning electron microscopy (SEM).

2. Experiment design

The precursor solution of PDMS was solution-sheared and cured on a prefabricated Si mold to prepare a pyramid-arranged film. The PDMS was sheared after placing the PS beads in the mold to add a porous surface to the pyramid structure. The PS was subsequently removed by toluene immersion to form a porous surface. Details of the film production methods have been described previously [20]. PDMS film is a suitable material with a refractive index less than that of the TFE layer (Al_2O_3, with $n = 1.65$), which contributes effectively to preventing total internal reflection.

The substrates used for the OLED production were rinsed with acetone, methanol, and deionized water for 15 min each and then dried in a vacuum oven for 15 min. Organic and metal layers were then deposited using a thermal vacuum evaporator. The detailed structures (Devices A to F) of the fabricated TEOLEDs are described in the Results and discussion section. The fabricated TEOLEDs were subjected to TFE passivation, and Al_2O_3 was applied to protect them from oxygen and water. Al_2O_3 was deposited by plasma-enhanced atomic layer deposition, where trimethylaluminum was used as the aluminum precursor. Argon was used as the carrier and purge gas during the TFE deposition. The base pressure of the deposition chamber was approximately 10^{-3} Torr and all layers were deposited at 95°C. The WVTR value of the Al2O3 TFE layer is below 5×10^{-5} gm^{-2} d^{-1}, which is the limiting value in the measurement using MOCON equipment.

The transmittance and haze of the prepared films were measured using UV-Vis spectroscopy (Lambda 950, Perkin Elmer). The porous pyramid film's surface was analyzed by SEM (Hitachi S-4800). Source measurement (Keithley 238) was used to measure the current density–voltage-luminance (*J-V-L*), CE, and external quantum efficiency (EQE) of the OLEDs. The angle-dependent EL intensity and Commission Internationale de l'eclairage (CIE) 1931 coordinates of the TEOLEDs were measured using a spectroradiometer (CS-2000, Minolta) and goniometer system.

3. Results and discussion

The prepared films were designed to reduce the total internal reflection of the TEOLEDs and suppress angular dependence by inducing the diffusion of light from the porous surface [19]. A pyramid-structured film was adopted to effectively reduce total internal reflection. In the case of the porous pyramid film, innumerable 2 μm-sized pores were arranged such that the transmitted light could be evenly diffused. As indicated in the SEM image in Figure 1, the manufactured film had a pyramidal structure resulting from the Si mold with countless pores formed after removal of the PS beads on the surface using toluene. Consequently, the efficiency and angular dependence of the TEOLEDs improvement could be expected. To measure the light diffusion between the pyramid film and porous pyramid film, the total transmittance (T_t) and parallel transmittance (T_p) were measured using a UV-Vis spectrometer (Figure 2). The diffuse transmittance (T_d) and haze were calculated using the following equations:

$$T_d + T_p = T_t(\text{Total transmittance})$$

$$T_d/T_t * 100 = \text{Haze}$$

The porous pyramid film exhibited an average of approximately 10% greater transmittance at all wavelengths. However, the ratio of the diffuse transmittance was greater in all sections (approximately 15) because it was greater in the porous film. The high haze characteristic of the porous pyramid structure could also be expected to improve the light distribution characteristics of the TEOLEDs.

To understand the change in luminance and efficiency of TEOLEDs with the attached optical property-enhancement films, devices were fabricated based on the three architectures displayed in Figure 3.

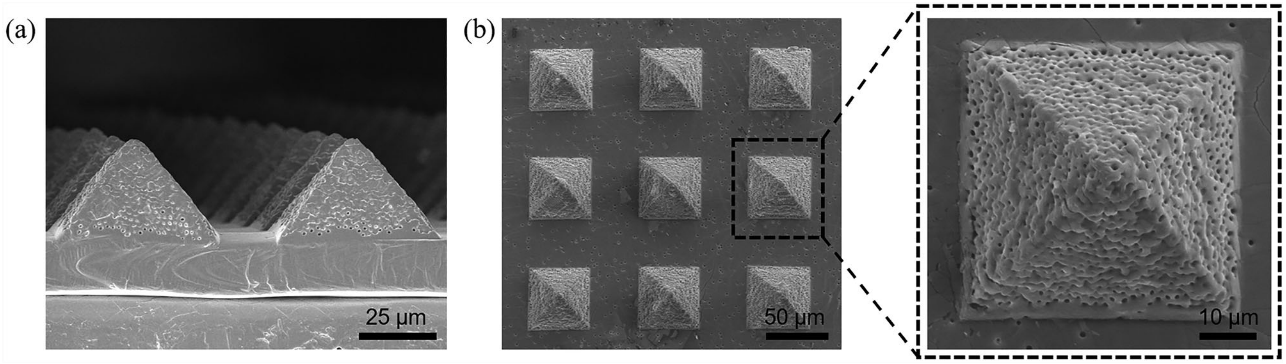

Figure 1. (a) Cross-sectional view and (b) top view of SEM images of porous pyramid film.

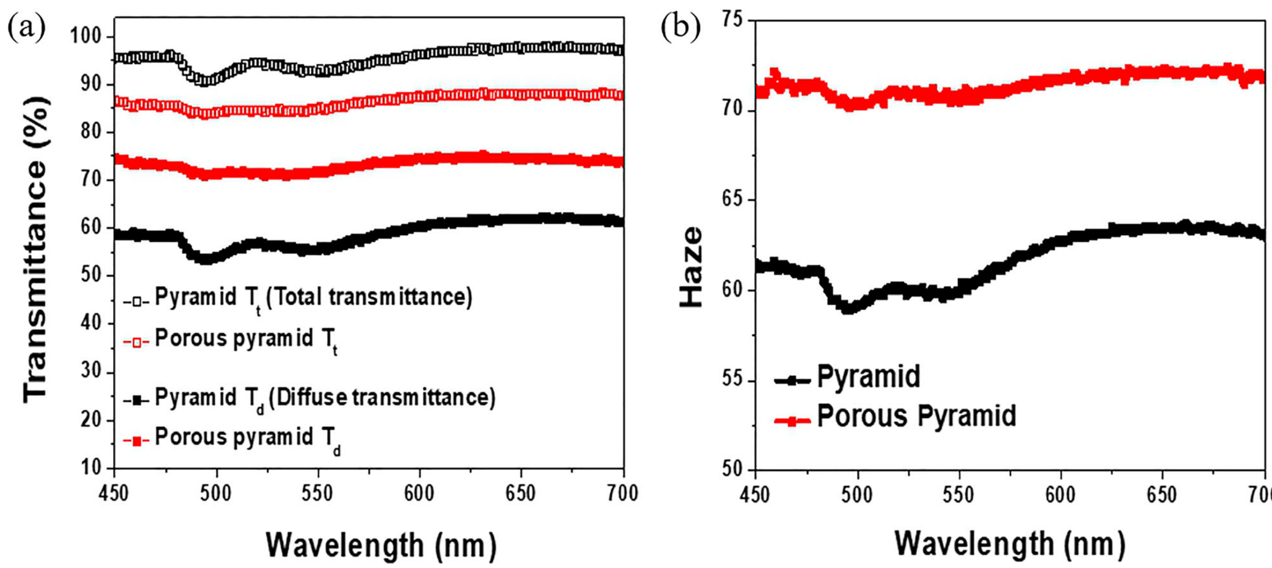

Figure 2. (a) Transmittance and haze of pyramid and porous pyramid film.

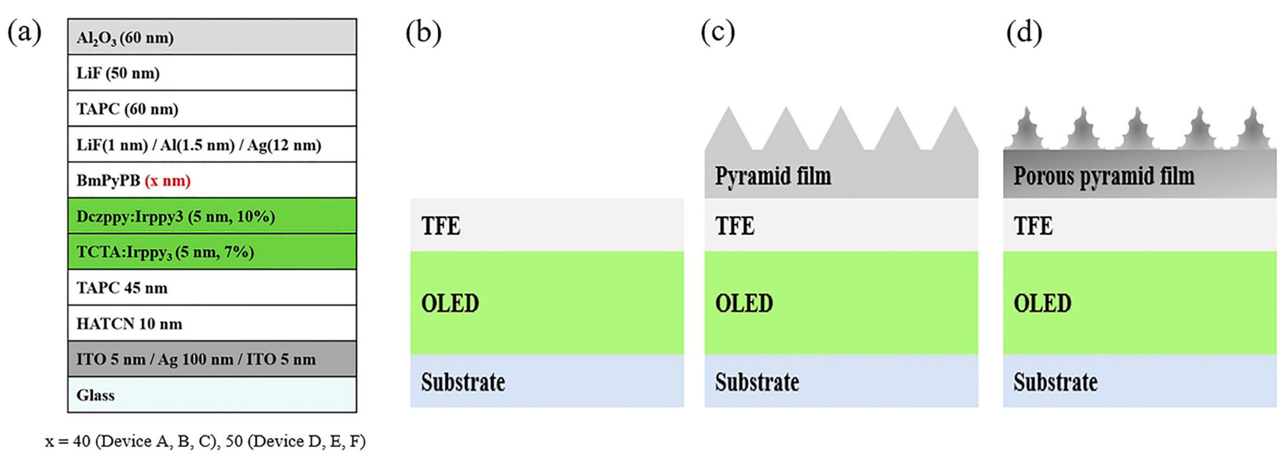

Figure 3. Device structure of (a) TEOLEDs, (b) reference OLED, (c) TEOLEDs with pyramid film, and (d) TEOLEDs with porous pyramid film.

Device A (Reference): ITO (5 nm)/Ag (100 nm)/ITO (5 nm) – Anode/Hexaazatriphenylenehexacarbonitrile (HAT-CN) (10 nm) – hole injection layer(HIL)/1,1-Bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) (40 nm) – hole transport layer/Tris(4-carbazoyl-9-ylphenyl) amine (TCTA):Tris(2-phenylpyridine)iridium(III) (Ir(ppy)$_3$) (5 nm) bis[3-(9H-Carbazol-9-yl)phenyl]pyridine (DCzPPy):(Ir(ppy)$_3$) (5 nm) – emitting layer (EML)/1,3-bis[3,5-di(pyridin-3-yl)phenyl]benzene (BmPyPB) (40 nm) – electron transporting layer (ETL)/Lithium fluoride

(LiF) (1 nm) – electron injection layer(EIL)/Al (1.5 nm)/Ag (12 nm) – Cathode/TAPC (60 nm) – capping layer (CPL)/LiF (50 nm)/Al_2O_3 (60 nm) – TFE.

Device B: Anode/HIL/HTL/EML/ETL (40 nm)/EIL/Cathode/CPL/TFE/**Pyramid film.**

Device C: Anode/HIL/HTL/EML/ETL (40 nm)/EIL/Cathode/CPL/TFE/**Porous pyramid film.**

Figure 4(a) displays the current density–voltage-luminance (*J-V-L*) characteristics of Devices A, B, and C, respectively. As expected, the luminance of the devices equipped with the optical property-enhancement films (Devices B, and C) increased under the same current density. The EQE and CE were also improved, as indicated in Figure 4(b,c), respectively. For example, at approximately 800 cd/m^2, the CE of Devices A, B, and C were 53.6, 62.1, and 63.8 cd/A, respectively. At approximately the same luminance, the EQE of Device A was 14.9%,

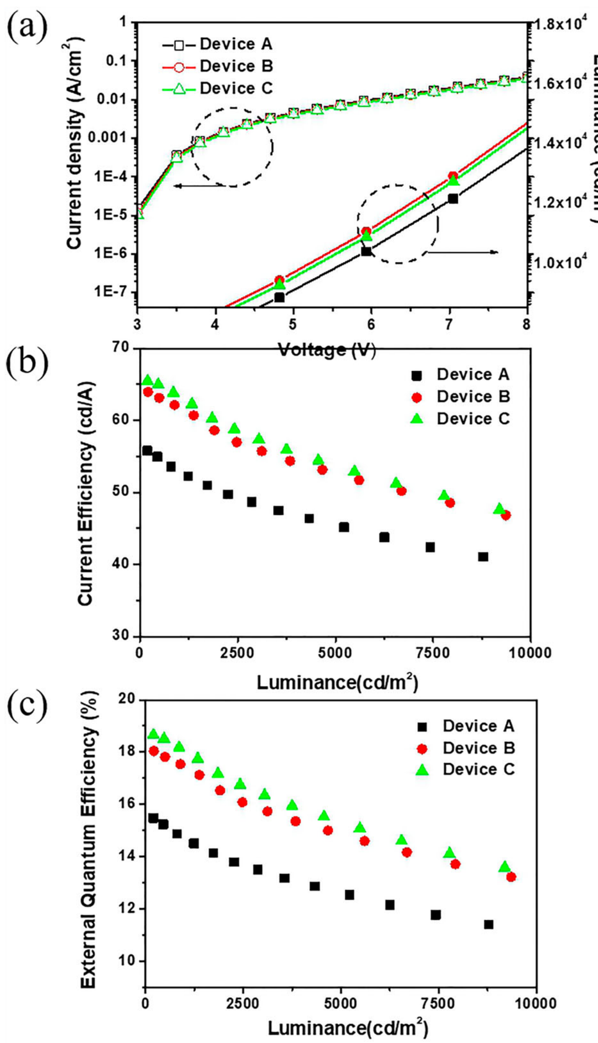

Figure 4. (a) Current density-voltage-luminance, (b) Current efficiency-luminance, and (c) External quantum efficiency characteristics of Device A, B, and C.

Table 1. Summarized light-emitting performances of Devices A, B, and C.

	cd/m^2	EQE (%)	EQE enhancement (%)	C.E (cd/A)	C.E enhancement (%)
Device A	804.1	14.9		53.6	
Device B	891.9	17.5	17.9	62.1	15.9
Device C	857.2	18.2	22.3	63.8	19.0

Note: At a driving voltage of 4.1 V.

whereas Devices B and C demonstrated enhanced EQEs of 17.5% and 18.2%, respectively. Details of the device performance are summarized in Table 1. This efficiency improvement can be attributed to the critical angle reduction from the pyramid and porous surface, which are the main components of the optical property-enhancement film. The trapped light caused by total internal reflection could be extracted into the air, leading to improved efficiency.

From the performance metrics, it was demonstrated that the improvement in EQE was greater than the improvement in CE. This can be attributed to the normalized angular intensity distribution displayed in Figure 5(a). Device A demonstrated the greatest intensity in the range of 0°–30°, which represents the luminance in the forward direction of the device. Device C was close to the Lambertian distribution, indicating that the light transmitted through the porous surface was evenly distributed. It can be inferred that this contributed to the improvement in the EQE of the TEOLEDs by controlling the light distribution characteristics of the porous film. Suppression of the angular dependence of the film is also demonstrated by the angle-dependent normalized EL spectra and CIE coordinates, as indicated in Figure 5(b). In Device A, a peak shift of 36 nm was observed in the EL spectrum from 550 nm to 514 nm as the measurement angle increased from 0° to 60°. For Device B, the peak shifted by 9 nm, from 524 nm to 515 nm. Device C indicated a peak shift of only 5 nm from 520 nm to 515 nm, demonstrating the ability of the porous pyramid film to effectively suppress the angular-dependent electroluminescence intensity (EL intensity). Figure 5(c) displays the angular-dependent characteristics of the CIE coordinates of the devices. For Device A, the difference between the maximum and minimum CIE (x,y) values was (0.1053, 0.059). For Devices B and C, however, the difference decreased to (0.055, 0.026) and (0.038, 0.016), respectively. This further highlights the advantage of optical property-enhancement films in suppressing angular-dependent changes in CIE coordinates.

An additional experiment was conducted to investigate the improvement in the luminance and suppression of the angular dependence in the TEOLEDs equipped with the fabricated enhancement films. The TEOLEDs

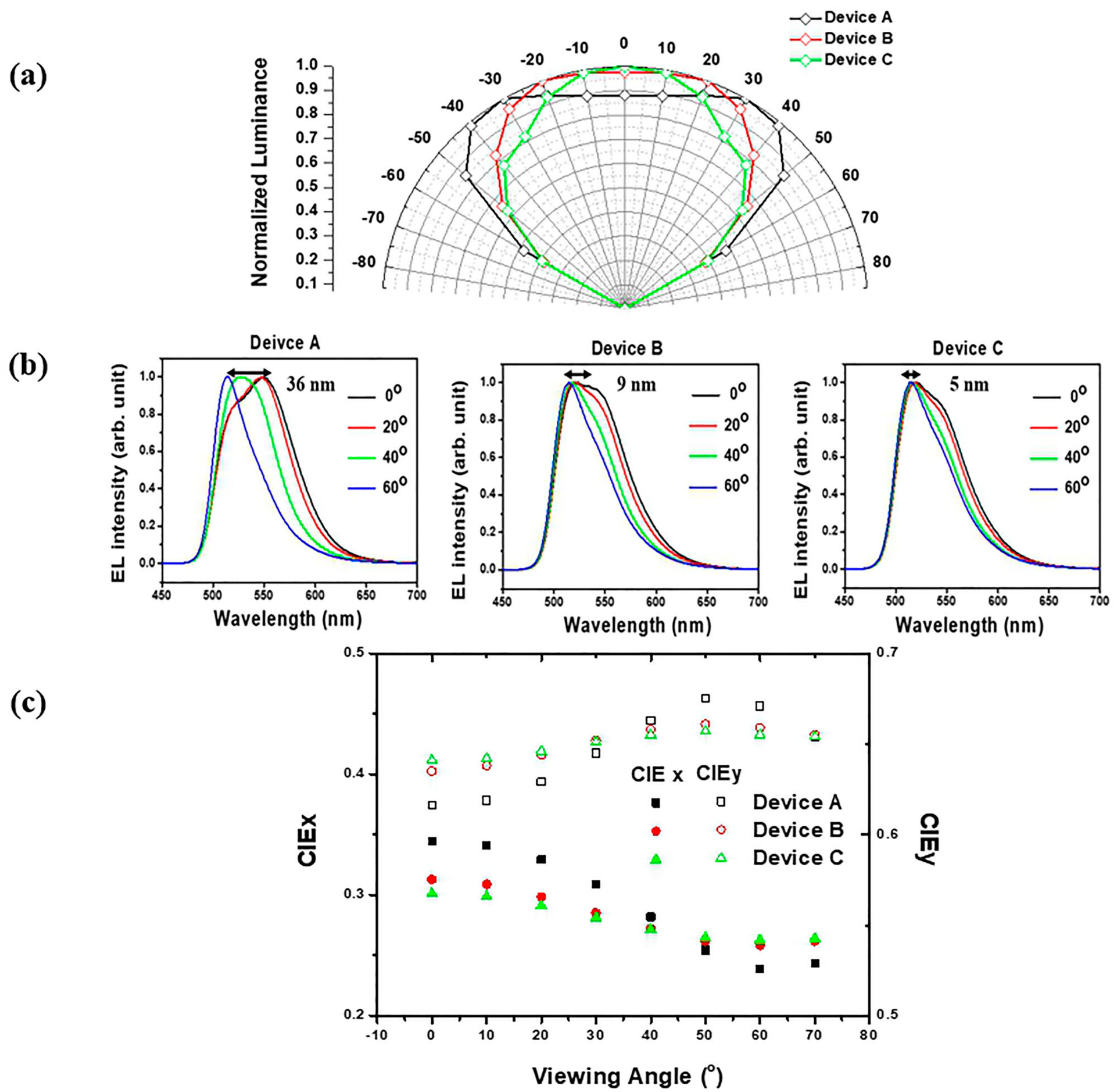

Figure 5. (a) Normalized angular distribution of luminance for Devices A, B, and C. Measured EL spectra of (b) Devices A, B, and C at 0°, 20°, 40°, and 60°. (c) CIE coordinates of Devices A, B, and C depending on viewing angles at a driving voltage of 4.1 V.

used were devices with an ETL thickness of 50 nm and were designed to have a strong luminance intensity near 50°. The device structures used in this experiment are described below.

Device D: Anode/HIL/HTL/EML/ETL (50 nm)/EIL/Cathode/CPL/TFE

Device E: Anode/HIL/HTL/EML/ETL (50 nm)/EIL/Cathode/CPL/TFE/**Pyramid film**

Device F: Anode/HIL/HTL/EML/ETL (50 nm)/EIL/Cathode/CPL/TFE/**Porous pyramid film**

The normalized angular intensity distributions of Devices D, E, and F are displayed in Figure 6(a). This indicates that Device D has a strong resonance at 50°, the devices with the attached films (Devices E and F) exhibit similar angular emission to the Lambertian distribution owing to the optical property-enhancement film that evenly distributes the light. The angle-dependent normalized EL intensity indicated in Figure 6(b) exhibits excellent color stability for Device F, which has a porous pyramid film. The peak shift from 0° to 60° was 30 nm for both Devices D and E, whereas Device F exhibited a peak shift of only 5 nm. The angular-dependent characteristics of the CIE coordinates of Devices D, E, and F are indicated in Figure 6(c). It was demonstrated that device

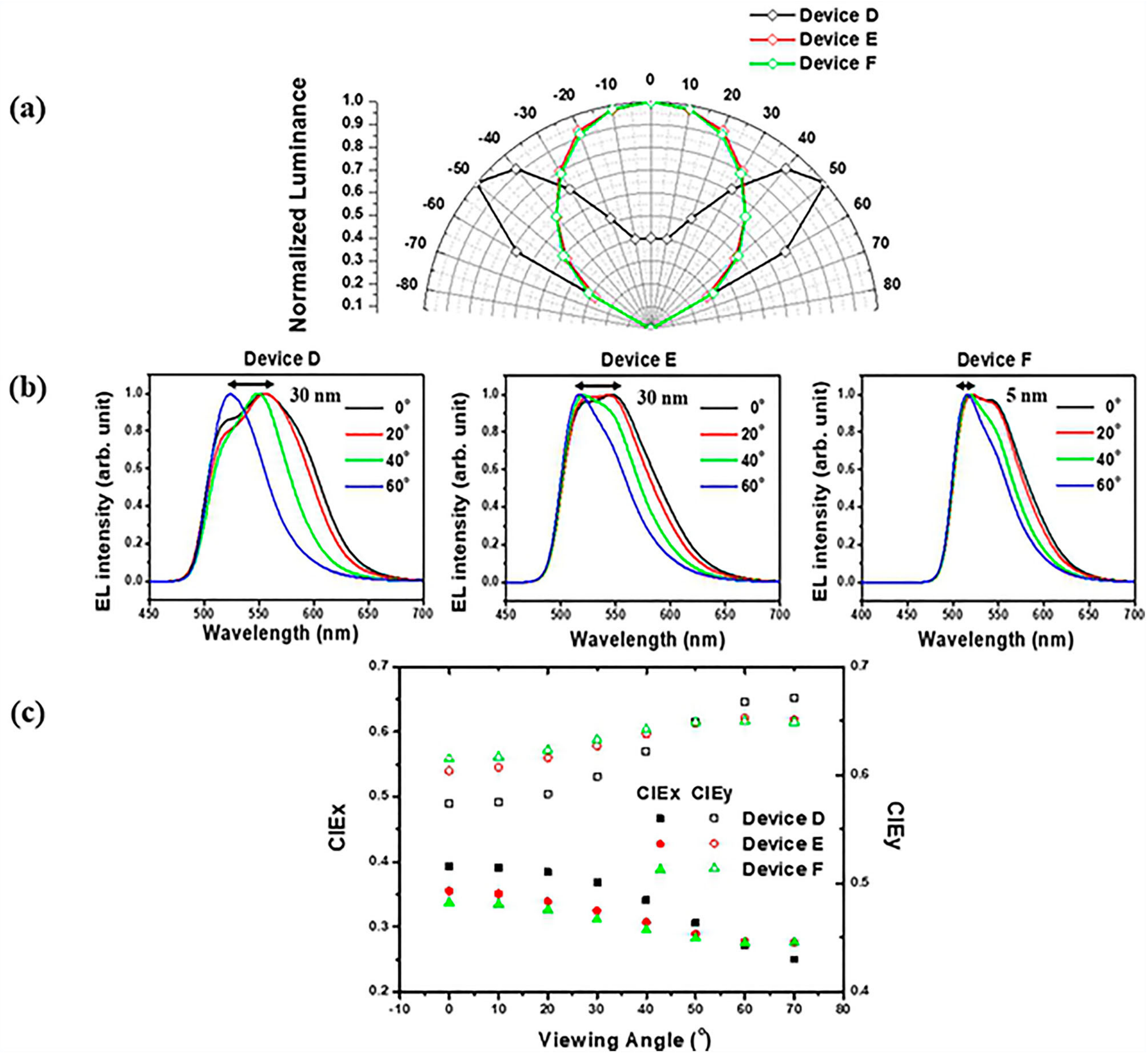

Figure 6. (a) Normalized angular distribution of luminance for Devices D, E, and F. Measured EL spectra of (b) Devices D, E, and F at 0°, 20°, 40°, and 60°. (c) CIE coordinates of Devices D, E, and F depending on viewing angles at a driving voltage of 5 V.

F recorded the least CIE coordinate variation. The CIE coordinate variation ranges Δ(x,y) for Devices D, E, and F were (0.1424,0.097), (0.079, 0.049), and (0.0611, and 0.035), respectively. This investigation clearly demonstrates the ability of the porous pyramid film to suppress the angular dependence of TEOLEDs, even with a strong microcavity.

Figure 7(a) displays the *J-V-L* characteristics of Devices D, E, and F. Compared with the previously measured TEOLEDs with an ETL of 40 nm, the significantly reduced critical angle at the porous pyramid interface allows for the greatest luminance exhibited by Device F. This clearly demonstrates the advantages of the porous pyramid film as an optical property-enhancement film. The EQE and CE of Devices D, E, and F are indicated in Figure 7(b,c), respectively. At 5 V, the EQE of Devices D, E, and F were 12.23%, 15.49%, and 16.51%, respectively. Compared to the reference device (Device D), Devices E and F demonstrated EQE improvements of 26% and 35%, respectively. Devices D, E, and F exhibited CE of 44.30, 55.88, and 61.19 cd/A, respectively. This corresponds to 26.14% and 38.12% CE improvements in Devices E and F, respectively. The performance of Devices D, E, and F is summarized in Table 2. This investigation proves the ability of performance films to distribute light uniformly over a large viewing angle. It was demonstrated that the optical property-enhancement films effectively improved luminance even in TEOLEDs with weak luminance in the forward direction.

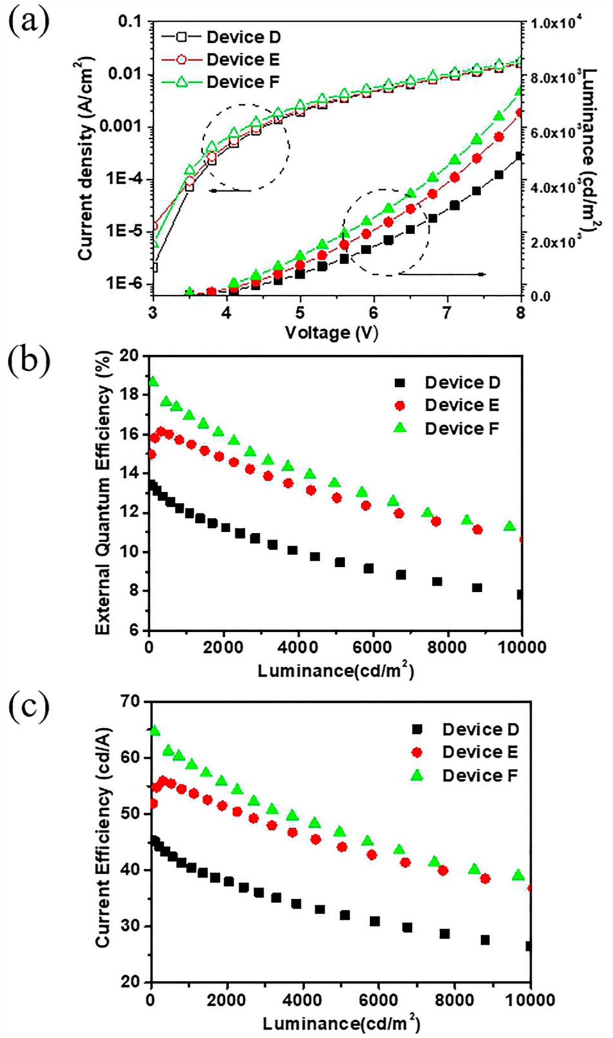

Figure 7. (a) Current density-voltage-luminance, (b) Current efficiency-luminance, and (c) External quantum efficiency characteristics of Devices D, E, and F.

Table 2. Summarized light-emitting performances of Devices D, E, and F.

	cd/m^2	EQE (%)	EQE enhancement (%)	C.E (cd/A)	C.E enhancement (%)
Device D	808	12.23		44.30	
Device E	1127	15.49	26.66	55.88	26.14
Device F	1145	16.51	34.99	61.19	38.12

Note: At a driving voltage of 5 V.

4. Conclusion

We proposed the application of a porous pyramid array optical property-enhancement film as an effective method of improving the efficiency and suppressing angular-dependent emission in advanced TEOLEDs. The TEOLEDs equipped with the optical property-enhancement films exhibited an improved EQE of 35% and a CE of 38% without changing the electrical characteristics of the pristine TEOLEDs. The films extract light trapped owing to total internal reflection at the TFE-air interface. It was further demonstrated that even when porous pyramid films were applied to TEOLEDs with biased emission, the angular-dependent EL intensity was significantly minimized by effectively controlling the emission angle, resulting in a light distribution close to Lambertian. The proposed optical property-enhancement film for TEOLEDs is versatile, process-friendly, and can be adopted in advanced OLED displays and lighting.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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Notes on contributors

Baeksang Sung received his B.S. and M.S. degree from Hanbat National University, South Korea in 2020, and 2022, respectively. He is currently taking the Ph. D course in the Department of Creative Convergence Engineering of Hanbat National University.

Chul Woong Joo received his B.S. and M.S. degrees in Polymer Science and Engineering of Organic Electronics Devices from Dankook University, South Korea in 2008 and 2010, respectively. He joined the Electronics and Telecommunications Research Institute in Daejeon, South Korea in 2011. His current research interests include device architectures in OLEDs and next-generation displays.

Jun Chang Yang received his B.S. degree in Materials Science and Engineering at Korea University, South Korea in 2016. He subsequently received his M.S. and a Ph.D. degrees in Materials Science and Engineering at the Korea Advanced Institute of Science and Technology (KAIST), South Korea in 2018 and 2022, respectively. He is currently working as a postdoctoral associate at KAIST. His research interests focus on tactile sensing electronic skin for wearable and robotic applications, 3d printing, and soft robotics.

Akpeko Gasonoo received his Bsc. degree in Computer Engineering from Kwame Nkrumah University of Science and Technology, Kumasi, Ghana in 2013, and his MEng. and PhD degrees in Electronic Engineering from Hanbat National University, Daejeon, South Korea in 2017, and 2021 respectively. He joined the Research Institute of Printed Electronics & 3D Printing, Hanbat National University, Republic of Korea as a postdoctoral researcher. His current research interests include printed organic electronics, BIPV, OLED optics and processes.

Seung Wan Woo received his B.S. degree from Hanbat National University, South Korea in 2022. He is currently taking the M.S. course in the Department of Creative Convergence Engineering of Hanbat National University.

Jae-Hyun Lee is a professor in the Department of Creative Convergence Engineering at Hanbat National University. He received his B.S. and Ph. D. degrees from the Department of Materials Science and Engineering at Korea University in 2002 and Seoul National University in 2011, respectively. He worked as a post-doctoral fellow at the Institut für Angewandte Photophysik (IAPP) of Technische Universität Dresden (TU Dresden). His research areas include flexible organic electronics and electrically doped organic semiconductors.

Steve Park is an associate professor in the Department of Materials Science and Engineering at KAIST since 2016. Prof. Park received his Bachelor's degree in Materials Science and Engineering from the University Illinois at Urbana-Champaign. He then received his Master's and a Ph.D. degree in Materials Science and Engineering at Stanford University. He went on to conduct his postdoctoral scientist work at the Electrical Engineering Department of Columbia University. Prof. Steve Park's research interests are in tactile sensing electronic skin for wearable and robotic applications, solution-based thin-film crystallization for flexible electronics, 3D printing, and biosensors.

Jonghee Lee received his B.S., M.S., and Ph.D. degrees in Chemistry from the Korea Advanced Institute of Science and Technology (KAIST), Rep. of Korea, in 2002, 2004, and 2007, respectively. He joined the Electronics Telecommunications Research Institute (ETRI), Rep. of Korea, in 2007. He moved to the Institut für Angewandte Photophysik (IAPP) at the Technische Universität Dresden (TU Dresden) in Germany as a post-doc researcher under the Alexander von Humboldt fellowship in 2010. He joined ETRI again in 2012 and has since developed device architectures and process of organic light-emitting diodes (OLEDs) as well as next generation opto-electronic devices. He has been an associate professor at Hanbat National University, Daejeon, South Korea since 2018.

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