Spectrally independent and wide-angle light extraction of organic light emitting diodes with randomly disassembled nanostructure†

Joel Ndikumana and Kunsik An*  

Organic Light Emitting Diodes (OLEDs) have emerged as popular screen advancements in digital appliances, thanks to their unique characteristics such as self-emitting ability, transparency, true dark tone, and flexibility. In comparison to liquid crystal displays, OLED displays offer superior performance. Moreover, OLEDs are also a promising option for illuminating purposes. Although significant progress in enhancing the internal quantum efficiency (IQE) of OLEDs, achieving an external quantum efficiency that matches their full potential has proven challenging due to optical losses. This paper explores the utilization of Randomly Disassembled Nanostructures (RaDiNa) for light extraction in flexible OLEDs. The study confirms that the implementation of RaDiNa enhances light extraction efficiency, particularly at angles above the critical angle, leading to an increased external quantum efficiency (EQE). The application of a cured polydimethylsiloxane (PDMS) onto the substrate resulted in the RaDiNa-structured OLEDs demonstrating not only an extended viewing angle but also stable light emission across all angles, reinforcing the technology’s value for high-quality display applications. These findings underscore RaDiNa’s potential for significantly improving the efficiency of flexible OLEDs without altering the light emission spectrum based on the viewing angle.

Introduction

Organic Light Emitting Diodes (OLEDs) are appealing for screen technology due to their high-efficiency, minimal energy consumption, and Bendability.1–3 The advancement of highly efficient OLEDs is pivotal for numerous facets of contemporary living, ranging from the growing desire for extended battery life in handheld devices to the global call for decreased energy use. Despite significant advancements in increasing the IQE of OLEDs to approximately 100%, the external quantum efficiency (EQE) still lags due to photonic dissipation.4–8 One of the primary challenges faced by OLEDs is the low light extraction efficiency from the active layer, leading to reduced EQE and limited brightness.9

The EQE represents the proportion of electrically injected carriers to the photons that are observed externally. In the case of OLEDs, there are three primary factors that restrict the EQE. Firstly, when photons are generated within the emissive layer (EML), a fraction of these photons is dissipated at the boundary connecting the organic layers and the contact metal as the result of the surface plasmon-polariton (SPP) mode activation. Secondly, photons become confined within the substrate and fail to exit the device because of Total Internal Reflection (TIR) happening at the boundary between the air and the substrate. Lastly, the photons produced within the emissive layer (EML) becomes enclosed within the OLED as a waveguide mode given that the organic packs and transparent conducting electrode (TCE) function as a waveguide.1,10,11 To address the challenges associated with light extraction in OLEDs, numerous approaches have been developed aiming to enhance light out-coupling efficiency. These methods include the incorporation of scattering layers such as low-index grids,12 air bubbles,13 nanoparticles,14,15 nanopillars,16 micro-lenses,17 Periodic and random corrugations within OLEDs and wrinkle substrates for external light extraction.18–22 However, these approaches often suffer from limitations such as wavelength dependence, stemming from the presence of resonant coupling, or intrusiveness to the device structure.23,24 Additionally, many of these methods require complex fabrication processes, resulting in increased costs and mismatch with extensive-scale fabrication. Hence, there exists a pressing demand for a novel technique that can effectively address the outcoupling challenge without being intrusive, while also having scalability, cost effectiveness, simplicity in manufacturing, independence from wave-
length, complementarity with large-area OLED manufacturing processes.\textsuperscript{25,26} The development of such a solution would enable the creation of energy-efficient OLEDs and could have an immediate impact on the commercialization of display products. These external structures do not adversely affect the electrical characteristics of the device and may be produced separately from the organic layer. Nevertheless, in terms of cost-effectiveness and the ability to scale up OLED manufacturing, there is a need for a more straightforward and economical approach to create suitable external light outcoupling structures that enhance the efficiency of external light emission. This continues to be the primary challenge in this area of research.

Here we report OLEDs with an improved light outcoupling and reduced angular dependence by introducing a polydimethylsiloxane (PDMS) laminate with the holes created by utilizing Zinc Oxide Nanorods (ZnO NRs). We refer to this structure as “randomly disassembled nanostructure (RaDiNa)”. This layer plays a crucial role in scattering the extracted light, thereby widening the OLED viewing angle. Previous studies have reported the use of wrinkled PDMS layers as light outcoupling structures for white OLEDs.\textsuperscript{18,27–30} However, these films exhibit wavelength dependence in their scattering properties, leading to issues such as image blurring, non-uniform light extraction, and variations in efficiency across the device. Additionally, they may also introduce color deformation and angular dependence of color, further affecting the overall performance.\textsuperscript{31–33} While perpendicularly oriented ZnO NRs have been applied to enhance light outcoupling in inorganic devices, their high-temperature synthesis process makes them unsuitable for organic devices. In contrast, the formation of the RaDiNa film avoids involvement of elevated-temperature phase since it utilizes pre-existing NRs as a template. Moreover, while conventional ZnO NRs require a high-temperature annealing process, the RaDiNa-attached OLEDs merely necessitates further annealing at a temperature below 100 °C. Thus, the RaDiNa surface offers a distinctive solution which overcomes the limitations of traditional ZnO NRs as a layer for light outcoupling, making it applicable to both organic and inorganic displays.

Results and discussion

In the construction process of the RaDiNa layer, a series of precise and carefully arranged steps were undertaken, as depicted in Fig. 1a–f. The process initiated with the deposition of a ZnO seed layer, denoted as Fig. 1a, which serves as the foundation for subsequent growth. This was followed by the controlled growth of ZnO NRs, as illustrated in Fig. 1b, subsequently, a self-assembled monolayer (SAM) treatment, depicted in Fig. 1c, was administered to ease the detachment of PDMS. The fabrication process further encompassed the precise deposition of a PDMS layer, as demonstrated in Fig. 1d, within which NRs-shaped holes are formed. Fig. 1e elucidates the subsequent steps of substrate integration and PDMS curing, where careful attention was dedicated to achieving uniformity and structural integrity. The process culminated in Fig. 1f, whereby the substrate detachment and OLED fabrication were carried out, bringing together the carefully engineered layers and components to form a functional OLED device. While further in-depth details are available in the dedicated methods section, the summarized depiction in Fig. 1a–f serves as a visual guide to the sequential stages that collectively contribute to the successful fabrication of OLEDs.

After the detachment of PDMS, the ZnO NRs were examined using cross-sectional SEM images, with their average length serving as a parameter as illustrated in Fig. 2. These images revealed that the PDMS layer separated from a medium featur-
ing random interfaces, lacking any periodic characteristics. Although periodic structures in OLED light extraction layers have significant advantages, such as enhanced light extraction efficiency,34–36 directional emission control,21,37 spectral control,38 compatibility with large-area manufacturing,39 and complementarity with other light extraction techniques,40 they also have certain disadvantages. Firstly, they can lead to potential optical interference, such as the occurrence of Moiré patterns.38,41,42 Additionally, the periodic structures possess a clearly specified grating vector and orientation, causing the extracted light to exhibit strong dependencies on both angle and spectrum.1,34,43–45

These result in a pronounced angular spectral profile, which is generally unwanted in display and lighting uses. The dimensions of the ZnO NRs prior to PDMS covering were adjusted by managing the development period of the ZnO NRs precursor from 2 h to 3 h 30 min, resulting in dimensions ranging from 300 nm to 700 nm, as illustrated in Fig. 2. Additionally, it was observed that the quantity of leftover PDMS residue varied with the length of the ZnO NRs post-separation. ZnO NRs measuring 300 nanometers in length were primarily enveloped by the PDMS remains, indicating insufficient molding of the removed RaDiNa layer. Conversely, as the length of the NRs increased, the proportional amount of PDMS remainder to ZnO NRs decreased, indicating clearer molding of the sculpture by the detached PDMS film. It can be concluded that securing a sufficient length of the NRs is crucial for efficient outcoupling, as the intention of using the NRs is to create well-defined and effective rodlike holes within the PDMS. Nonetheless, a greater amount of PDMS remnants persisted on the 700 nm ZnO NRs in contrast to the 600 nm ZnO NRs which is against the anticipated pattern. This discrepancy can be attributed due to the large surface area of the 700 nm ZnO NRs, leading to excessive adhesive interaction between the PDMS and ZnO NRs. Consequently, this obstructed the creation of a distinct three-dimensional configuration in the simplified RaDiNa architecture, leading to suboptimal optical properties. Further discussion and analysis of these experimental results will be presented in the following sections.

Fig. 3 illustrates the optical simulation conducted to validate the light outcoupling capability of RaDiNa. The simulation utilized COMSOL Multiphysics, employing wave optic theory to analyze the propagation of light within the RaDiNa film. Specifically, the simulation focused on the characteristics of light propagation from the underside PDMS to the upper-side atmosphere when illuminating wavelengths of 500 nm, 550 nm, 600 nm, and 650 nm, with incident angles ranging from 0° to 70°. Each outcome of the simulation was evaluated against PDMS film devoid of any external patterning. In the simulation, the RaDiNa structure was modelled with random depths and widths, utilizing parameter values for vertically aligned pore arrays. The widths varied randomly between 50 nm and 100 nm, while the heights ranged from 300 nm to 600 nm. The magnitude of light transmitted at the planar interface plane decreases with increasing incident angles, with a corresponding decrease in field intensity as demonstrated in Fig. 3. This agrees with the Fresnel equations, which describe the behaviours of light during reflection and refraction at the boundary between two distinct optical media.46 The Fresnel equations explain how light is partitioned into reflected and refracted components upon encountering a boundary between

Fig. 2 SEM images of ZnO NRs, taken in a cross-sectional view, were obtained after PDMS separation, revealing average ZnO NRs lengths of (a) 300 nm, (b) 400 nm, (c) 600 nm, and (d) 700 nm.
media of differing refractive indices. Eqn (1) and (2) details the
transmittance with $T_s$ and $T_p$ representing the transmittance values for s-polarized (perpendicular) and p-polarized (parallel) light, respectively. The terms $I_s$ and $I_p$ denote the incident light's magnitudes for perpendicular and parallel polarizations, while $n_1$ and $n_2$ correspond to the refractive indices of the incident and transmitting media. The angles $\theta_i$ and $\theta_t$ correspond to the incident and transmission angles of the light, respectively.

$$T_s = \frac{2n_1 \cos \theta_i}{n_1 \cos \theta_i + n_2 \cos \theta_t} I_s \quad (1)$$

$$T_p = \frac{2n_1 \cos \theta_i}{n_1 \cos \theta_i + n_2 \cos \theta_t} I_p \quad (2)$$

However, it is important to note that the Fresnel equations are only valid for planar interfaces. In the case of complex structures within the wavelength of light, light propagation follows the effective medium approximation. This theory portrays the effective refractive index as a bulk characteristic, representing the adjusted mean of the constituent materials adjusted by their fractions. In the case of RaDiNa featuring an 80 nm hole diameter, less than the emission wavelength of the fabricated OLED, the effective refractive index was calculated using eqn (3), where $n_{\text{eff}}$, $n_{\text{PDMS}}$, and $n_{\text{air}}$ represent the refractive indices of the effective medium, RaDiNa, and atmosphere, correspondingly. The volume fraction of RaDiNa is denoted as $f_{\text{RaDiNa}}$.

$$n_{\text{eff}} = \left[\frac{n_{\text{PDMS}}^2 f_{\text{RaDiNa}} + n_{\text{air}}^2 (1-f_{\text{RaDiNa}})}{2}\right]^{1/2} \quad (3)$$

With a PDMS refractive index of 1.43 and an air refractive index of 1.00, the resultant effective refractive index of RaDiNa falls lies within these ranges. This observed effect, rooted in the effective medium theory, led to a significant enhancement in light extraction across a wide angular range from 0° to 70° at all wavelengths relative to the flat interface surface, as clearly demonstrated in Fig. 3. The RaDiNa, fabricated using a haphazardly aligned ZnO NRs array, exhibited a gradual decrease in refractive index from its bottom to top. This ensured a minimized refractive index difference at the interfaces between the RaDiNa and air. Such subtle index differences effectively suppressed internal reflections returning to the device, further augmenting light extraction. This effective

**Fig. 3** Field distribution simulations conducted to analyze the extracted electric field from both the RaDiNa and flat surfaces under varying light propagation angles ranging from 0° to 70°, while using illumination wavelengths of 500 nm, 550 nm, 600 nm, and 650 nm.
refractive index can be engineered to be closer to that of air, reducing the mismatch at the interface and thus reducing the critical angle. Light that would normally undergo TIR at a lower angle can now escape, leading to enhanced intensity at angles above the original critical angle. The comparable field distribution across all applied wavelengths: 500 nm, 550 nm, 600 nm, and 650 nm at similar angles, indicates that the RaDiNa layer’s irregular structure does not evidently influence the emitted color. Consistent field strength, which denotes color distribution, across these wavelengths at all angles suggests that the light extraction technique is wavelength independent. Such wavelength independence is a desirable characteristic, ensuring that the OLED’s color spectrum remains stable and consistent, regardless of the viewing angle. As depicted in Fig. 3, the field intensity strengthens notably as the angle exceeds the critical angle. This enhancement in intensity at emission angles beyond the critical angle between the layer and air is likely due to the role of the patterned PDMS layer embedded with randomly oriented nanostructures, which facilitates improved light extraction.

Fig. 4a and b present a diagrammatic representation of the device configuration with RaDiNa layer and schematic energy diagram of OLED. The OLED is constructed with 20 nm-thick molybdenum trioxide (MoO₃) hole injection layer (HIL), a 20 nm-thick TAPC hole transport layer (HTL), a 30 nm-thick layer of CBP:Ir(ppy)₃ emissive layer, a 40 nm-thick 1,3,5-tris(N-phenylbenzimidazole-2-yl)benzene(TPBi) electron transport layer (ETL), a 0.5 nm layer of LiF for electron injection, and a 100 nm layer of Al.

Device performance evaluation is illustrated in Fig. 5. Fig. 5(a) present the normalized EL spectra of OLEDs with RaDiNa, plotted against the average ZnO NRs length. The electroluminescence (EL) spectra exhibit minimal variation across all ZnO NRs length. This underscores the absence of grating effects and confirms a wavelength-independent response, essential for enhanced light extraction throughout the visible light spectrum. The unnormalized EL spectra at each average length of ZnO NRs, as well as without RaDiNa, are provided in ESI Fig. S1 (a, b, c, d, and e for 300 nm, 400 nm, 500 nm, 600 nm, and no RaDiNa, respectively).† The current-density–voltage (J–V) and luminance–voltage (L–V) characteristics as shown in ESI Fig. S2(a) and S2(b)† respectively, demonstrate a high degree of similarity across various nanorod lengths. This consistency indicates that the electrical and optical properties of the OLEDs remain stable and reliable regardless of the variations in nanorod dimensions. In Fig. 5(b), we analyzed the EQE as a function of current density for OLEDs with varying average lengths of ZnO NRs. This analysis was conducted by comparing the relative EQE improvements of devices with RaDiNa layers of different nanorod dimensions to those without RaDiNa layers. The optimized RaDiNa-attached OLED achieves EQE enhancements over the reference device by 16.5%, 43.3%, 74.6%, and 81.1% for nanorod lengths of 300 nm, 400 nm, 500 nm, and 600 nm, respectively, which indicates that the EQE increases proportionally with the average length of the ZnO NRs. At each specified length, a subtle rise in EQE is noted when the electrical current per unit area increases from 0 mA cm⁻² to 0.010 mA cm⁻². However, a marked decrease is observed as the electrical current rises from 10 mA cm⁻² to 100 mA cm⁻². It is noteworthy that the difference in EQE between 500 nm and 600 nm is minimal. Clearly, employing longer RaDiNa can markedly enhance light extraction; the same current density produces a more pronounced EQE without introducing spectral distortions. To illustrate the advantages of RaDiNa in enhancing EQE, we compared our results with EQE data from other external light extraction methods reported in the literature. The table below summarizes the EQE improvements achieved by various methods, including our RaDiNa approach (Table 1).

The RaDiNa-based light extraction technology shows a notable EQE improvement of 63%, comparable to many existing external light extraction methods used in OLEDs. For comparison, TiO₂-based scattering layers and SnOx nanocone structures achieve EQE improvements of 52% and 23%,
respectively, while disordered Nano-Hemisphere MoO$_3$ and polyimide scattering layers with air voids exhibit improvements of 64.5% and 65%, respectively. Many established methods, such as dielectric Bragg gratings (micro/nano-structures), micro-lens arrays, and photonic crystals, primarily focus on modifying the outer surface of OLEDs. These techniques often involve complex fabrication processes and sophisticated designs. For instance, dielectric Bragg gratings require precise layer deposition (laser ablation and nano imprinting), which can damage substrates due to high power, while nanoimprinting involves complex processes and limitations on long periods. Similarly, micro-lens arrays, which are engraved on the substrate layer, improve light intensity and EQE by focusing and redirecting light, but their fabrication involves meticulous patterning and alignment, which can be complex and time-consuming. Methods like hot embossing, inkjet printing, and laser-based fabrication require precise control and high-quality equipment, which can increase production costs and complexity. Photonic crystals enhance light extraction through diffraction and resonance effects but also demand high precision in creating periodic structures with specific refractive indices, which can be complex and costly to fabricate. These structures can also introduce wavelength dependence and angular spectral profiles, leading to potential issues such as color deformation and variations in efficiency across the device.

In contrast, the RaDiNa approach offers a simpler and more scalable solution. RaDiNa utilizes ZnO NRs to create a disordered nanostructure embedded in a PDMS layer, which scat-

---

**Table 1** Summary comparing the EQE data published literature using external only light extraction methods with the EQE data of this article

<table>
<thead>
<tr>
<th>Light extraction technology/structure</th>
<th>EQE improvement (%)</th>
<th>Study/ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>RaDiNa</td>
<td>81</td>
<td>This work</td>
</tr>
<tr>
<td>TiO$_2$-based scattering layers</td>
<td>52</td>
<td>47</td>
</tr>
<tr>
<td>Disordered nano-hemisphere MoO$_3$</td>
<td>64.5</td>
<td>48</td>
</tr>
<tr>
<td>SnO$_x$ nanocone structure</td>
<td>23</td>
<td>49</td>
</tr>
<tr>
<td>Polymide scattering layers with air voids</td>
<td>65</td>
<td>50</td>
</tr>
<tr>
<td>Ordered microlens array (PDMS lenses attached to the glass substrates)</td>
<td>50</td>
<td>51</td>
</tr>
<tr>
<td>Parylene film</td>
<td>45.8</td>
<td>52</td>
</tr>
<tr>
<td>Ag nanoparticles scattering layer</td>
<td>24</td>
<td>53</td>
</tr>
<tr>
<td>Phase-separated polymer blends of PAA/PI embedded with SiO$_2$ nanoparticles</td>
<td>42</td>
<td>54</td>
</tr>
<tr>
<td>PMMA structure + ZnO</td>
<td>31.1</td>
<td>55</td>
</tr>
<tr>
<td>Random bowls textured (RBT) optical film</td>
<td>27</td>
<td>56</td>
</tr>
<tr>
<td>Polymide/silica hybrid film</td>
<td>21</td>
<td>57</td>
</tr>
<tr>
<td>Polymer-metal oxide composite scattering film (Al$_2$O$_3$ + PS)</td>
<td>40</td>
<td>58</td>
</tr>
</tbody>
</table>

---

**Fig. 5** Device performance. (a) shows normalized EL spectra of the RaDiNa appended OLEDs in relation to the average length of the ZnO NRs, (b) relative EQE measurements of OLEDs vs. current density, (c) normalized luminance vs. emission angle of all RaDiNa lengths.
ters and redirects trapped light, enhancing light extraction efficiency by 81%. The fabrication process of RaDiNa is relatively straightforward, involving the growth of ZnO NRs, application of a SAM, and deposition of PDMS, followed by the detachment of the ZnO template to form the nanostructured layer. This method avoids the high-temperature synthesis required by conventional ZnO NRs, making it more compatible with organic devices. Additionally, RaDiNa does not alter the light emission spectrum based on the viewing angle, ensuring stable light emission across all angles. This simplicity in manufacturing, combined with cost-effectiveness and scalability, makes RaDiNa a promising alternative to more complex light extraction techniques in OLEDs.

Fig. 5c graphically presents the angular dependence of emitted light intensity. While the conventional device emits a pattern similar to the Lambertian distribution, a distinct alteration emerges with the incorporation of the RaDiNa structure. As the RaDiNa length increases, a marked increase in side emission becomes evident. This heightened side emission is primarily due to the enhanced light outcoupling capabilities of RaDiNa, which facilitates the redirection of photons that would otherwise be trapped within the OLED structure due to TIR. Fig. 5(d) illustrates the normalized EL intensity in relation to the emission angle. The peak luminance intensity is notably observed at emission angles exceeding 45 degrees. The difference between the total (T) and the reference, termed as the incremental portion (I), is considerably higher. This highlights RaDiNa ability to extract more light that would otherwise be internally reflected due to TIR. This manifestation of phenomenon aligns with simulation results, demonstrating intensified field strengths at higher angles. The underlying reason for this heightened light extraction, especially at high angles, can be attributed to the physics of light propagation in layered media and the effective use of nanostructures within the device. Light that typically undergoes TIR at angles higher than the critical angle is often trapped inside a conventional OLED, resulting in optical losses. However, the introduction of a patterned layer with randomly oriented nanostructures helps to scatter and redirect this trapped light, which would otherwise remain confined within the device. These nanostructures disrupt the smooth internal interface and create localized disruptions in the TIR conditions, enabling the escape of light at angles that surpass the critical threshold. The significant difference in light extraction between a standard OLED and one enhanced with RaDiNa layer is most notable at these high angles. The random orientation of nanostructures not only maximizes the scattering effect but also widens the distribution of possible emission angles.

The Fig. 6 illustrates the computed effective refractive index profile of the RaDiNa layer as a function of its height, derived from analyzing the volume fractions of air and PDMS using SEM images. The profile is shown at each average height from analyzing the volume fractions of air and PDMS using SEM images. The profile shows that the effective refractive index gradually decreases from around 1.4 to 1.0 as the height increases, indicating the dominance of PDMS at the top and air at the bottom. This gradient ensures a more efficient transition of light from the high-index environment inside the OLED to the low-index air outside, enhancing light extraction, especially at angles prone to TIR.

The trend of increasing extracted intensity with the length of the nanostructures can be explained by considering the interaction dynamics of light within the patterned layer. Longer nanostructures provide a greater interaction volume and increased surface area for light to scatter. This results in an increased probability of light being redirected out of the device before it can undergo TIR, particularly at angles above the critical angle. Additionally, the longer nanostructures contribute to a more gradual transition in the effective refractive index from the RaDiNa material to air, reducing the abruptness of the index change at the interface and minimizing the conditions for TIR. The extended length of the nanostructures also facilitates multiple scattering events, allowing for a cumulative enhancement in light extraction. This is especially significant for light traveling at high angles, as it interacts with the nanostructures over a longer path, leading to a higher likelihood of redirection and escape, resulting in increased intensity observed at these angles. We conducted experiments to determine the effectiveness of different nanorod lengths, extending up to 700 nm. However, our findings revealed that lengths of 700 nm could not be fabricated effectively due to very high adhesion between the nanorods and PDMS. This
Excessive adhesion caused substantial portions of PDMS to remain adhered to the nanorods, compromising the integrity of the formed holes and thus the overall quality of the RaDiNa structure.

For any lighting application aspiring for high quality, it is imperative that the color remains consistent across various viewing angles. Fig. 7(a–d) serves as evidence to RaDiNa ability in this domain. The angular electroluminescence spectra of OLEDs integrated with RaDiNa at varying average ZnO NRs lengths of 300 nm, 400 nm, 500 nm, and 600 nm were thoroughly analyzed across a spectrum of viewing angles: 0°, 20°, 40°, 60°, and 80°. Considering that Fig. S3† illustrates the angular EL spectra of the device without RaDiNa, a striking observation from this analysis is the remarkable consistency in the spectral shape, irrespective of the specific RaDiNa conditions or the viewing angle. This is in line with our simulation results, which also predicted a negligible variation in the spectral shape with changing angles. Such consistency ensures that the emission color of the OLEDs remains unaffected by the irregular structure of the RaDiNa layer. The slight variations in the EL spectra across different viewing angles observed in Fig. 6 and Fig. S3† are real and can be attributed to several factors. Primarily, these differences are due to cavity effects inherent in the OLED structure. The microcavity effect, resulting from the interference of light within the thin-film layers of the OLED, can cause minor shifts in the emission spectrum at different angles. Additionally, variations in layer thicknesses within the device can slightly alter the optical path length, contributing to these spectral differences. Measurement errors, such as instrument noise and slight misalignments during spectral recording, can also introduce minor discrepancies. Despite these variations, the overall consistency of the EL spectra is maintained, as evidenced by the stable primary emission peak across all viewing angles. This confirms that the color remains consistent, validating the robustness of the OLED design.68

Conclusions

In our research, we introduced and detailed the integration of a light extraction structure, termed RaDiNa, in OLED devices. The PDMS film with ZnO nanorods-induced holes enhances light extraction efficiency, especially at angles above the critical angle, a notable breakthrough that could dramatically increase the utility of OLEDs in various applications. We have provided substantial evidence through film analyses, optical simulations, and device characterizations that the integration of RaDiNa leads to a pronounced increase inside emission, particularly at angles that would typically suffer from TIR. Importantly, this enhancement does not come at the cost of color fidelity; the light emission spectrum remains consistent across all viewing angles. The implications of our findings are significant, offering a method to not only improve external quantum efficiency but also maintain color stability in OLED displays, ensuring high-quality visual performance across a range of angles. This consistency in the emission spectrum, independent of the viewing angle, further underscores the flexibility of the RaDiNa structure in maintaining the desired optical properties of OLEDs.

Fig. 7 angular EL spectra of RaDiNa-attached OLEDs at various average lengths of ZnO NRs (a) 300 nm, (b) 400 nm, (c) 500 nm, and (d) 600 nm across distinct viewing angles (0°, 20°, 40°, 60°, and 80°).

This journal is © The Royal Society of Chemistry 2024

Published on 26 July 2024. Downloaded on 8/4/2024 5:43:16 AM.
Experimental section

Synthesis of ZnO NRs

The synthesis of ZnO NRs was conducted based on previously established procedures, with minor adjustments. In summary, a triple-necked round-base flask was used to prepare a blend consisting of 2 Grams of zinc acetate dihydrate (Zn(CH₃COO)₂·2H₂O) and 80 milliliters of methanol. The flask was heated to 60 °C under an N₂ atmosphere. Once the temperature stabilized, a solution of potassium hydroxide (KOH) mixed in methanol at a concentration of 23.25 mg mL⁻¹ was infused to the flask at a rate of 238.5 mL h⁻¹. The reaction proceeded for 150 minutes, followed by a 12-hour precipitation of the Zinc Oxide Nanoparticles (ZnO NPs). The resulting solution was then subjected to centrifugation at 4000 rpm used for 3 minutes to isolate the Undiluted ZnO NPs from the blend. Lastly, the ZnO NPs were respread in 1-butanol.

Radina fabrication

The production process of the Radina layer is outlined in the schematic illustration provided in Fig. 1. A self-assembled monolayer (SAM) is applied to the NRs through a spin-coating technique. The SAM possesses a thickness that closely approximates that of a single molecule layer. The purpose of these SAM-coated ZnO NRs is to assist the subsequent separation of the PDMS layer. Upon the completion of the SAM coating, the PDMS layer is coated on top the SAM-coated ZnO NRs. The next step involves detaching the substrate, resulting in the formation of NRs-molded apertures in the PDMS layer. This detachment process gives rise to the creation of the Radina layer, positioned at the bottom of the device. The Radina layer, characterized by its NRs-shaped holes, plays a significant role in the functioning of the overall structure.

Device fabrication

Flexible OLEDs were manufactured on PET substrates using the following device structure: a 150 nm hole injection layer (HIL)/a 20 nm hole transport layer (HTL)/a 30 nm layer of CBP: Ir(ppy)₃, a 40 nm electron transport layer (ETL), a 0.5 nm layer of LiF, and a 100 nm layer of Al. The HIL and HTL consist of LiF, and a 100 nm layer of Al. The HIL and HTL consist of carbon, molybdenum trioxide (MoO₃) and TAPC, correspondingly. The ETL poses the ETL. The 4,4′-N,N′-dicarbazolobiphenyl (CBP) doped with fac-tris(2-phenylpyridine)iridium(III) [Ir(ppy)₃] deposited by thermal evaporation were used to make the emissive layer, details of fabrication conditions are described elsewhere. To finalize the fabrication, the PDMS layer, now housing the Radina layer, is attached to the bottom surface of PET substrate in the OLED. This attachment ensures a secure and stable integration of the PDMS layer with the PET substrate. The entire fabrication process, as outlined in Fig. 1, allows for the successful construction of the Radina layer within the device, paving the way for its desired functionality and performance.

Optical simulation

The light extraction for the OLED was simulated using COMSOL Multiphysics 5. Electromagnetic waves were introduced through the bottom port, and periodic conditions employing Floquet periodicity were configured for the left and right ports. The upper port was configured with a dispersive boundary condition. The simulation covered incident angles ranging from 0° to 70°. The acquired simulation findings were afterward assessed in relation to those of the planar layer.

The OLED devices characterization

Cross-sectional images were obtained using a scanning electron microscope (SEM) from Hitachi, model S-4800. J–V–L (current density–voltage–luminance) characteristics were measured utilizing a source measurement unit (Keithley, model 236) in conjunction with a photomultiplier tube (PMT), an Si photodiode (Hamamatsu, model S5227-1010BQ), and a multimeter (Keithley, model 2000). EL (electroluminescence) spectra analysis was conducted utilizing a Konica Minolta spectroradiometer, model CS-2000. For studying the angular emission dispersion, EL intensities were recorded in relation to the angle utilizing a PMT coupled with a monochromator through an optical fiber. The specimens were boarded on a rotating platform for the measurements.

Data availability

The data supporting this article have been included as part of the ESL.†

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This research was funded by Konkuk University in 2024.

References


