



Fully biocompatible poly(butylene adipate-co-terephthalate) based microstructures for eco-friendly light extraction in organic light-emitting diodes

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Abstract

Organic light-emitting diodes (OLEDs) inherently suffer from severe optical losses, with most generated photons confined by total internal reflection. To mitigate these losses, conventional light-extraction methods rely on complex, energy-intensive fabrication using non-biodegradable materials. Thus, this study presents an eco-friendly and biocompatible strategy that combines poly(butylene adipate-co-terephthalate) (PBAT) as a biodegradable template with carboxymethyl cellulose and tannic acid (ECO-UV) films. Templates and films were fabricated through a simple, low-temperature, solution-based replication process without lithography or vacuum processes, forming irregularly distributed microstructures that enhance light scattering and suppress internal reflection. The OLEDs incorporating the ECO-UV film exhibited up to 36% enhancement in electroluminescence intensity, demonstrating that even a fully biodegradable, solution-processed system can achieve light-extraction performance comparable to that of conventional synthetic counterparts.

Keywords Organic light-emitting diodes · Light extraction · Biodegradable polymers · Ultraviolet shielding

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1 Introduction

Organic light-emitting diodes (OLEDs) are regarded as promising candidates for next-generation display and lighting technologies due to their high brightness, wide viewing angle and mechanical flexibility. However, OLEDs inherently suffer from significant optical losses as most generated photons are trapped within the device by total internal reflection (TIR), resulting in limited light extraction efficiency [1–3]. To address this issue, various light extraction techniques such as microlens arrays, scattering layers and photonic crystals have been investigated [4–9]. Although these methods can effectively improve light outcoupling, they typically rely on multi-step lithographic patterning, reactive ion etching processes that consume high energy and employ non-biodegradable synthetic materials. Such fabrication routes increase production costs, impose environmental burdens and reduce scalability for manufacturing [10–13].

To overcome these limitations, recent studies have explored the incorporation of sustainable and environmentally friendly materials into OLEDs devices [14–16]. Among biodegradable polymers, poly(butylene adipate-co-terephthalate) (PBAT) has emerged as a promising material because it combines flexible aliphatic segments with rigid aromatic units enabling adjustable crystallinity and mechanical strength [17–19]. These molecular characteristics allow PBAT to make it well suited for light-extraction applications through a simple low temperature solution-based process. Although PBAT is synthesized from petroleum-based monomers, it can decompose through enzymatic hydrolysis of its ester bonds generating carbon dioxide, water and biomass [20–22]. This degradation pathway minimizes long term environmental impact positioning PBAT as a practical and sustainable material.

In this study, a microstructured template was fabricated using PBAT and an external light-extraction film was fabricated by casting carboxymethyl cellulose (CMC) and tannic acid (TA) solution onto the PBAT surface. CMC provides high visible transparency and excellent film-forming ability [23–25] while the phenolic hydroxyl groups of TA impart strong ultraviolet (UV) absorption and potential antioxidant functionality [26–28]. The replicated microstructures enhance light scattering and optical haze thereby improving outcoupling efficiency, also the UV-blocking property of TA may help mitigate device degradation caused by photochemical reactions [15, 29]. When applied to bottom-emission OLEDs, the biodegradable light-extraction film yielded up to a 36% increase in electroluminescence intensity compared with reference devices. This study demonstrates a sustainable and scalable approach to improving

OLED performance through the integration of eco-friendly materials and low temperature solution-based fabrication.

2 Experimental

2.1 Materials

The templates were fabricated using PBAT, silicon wafers, and chloroform (CAS 67–66-3). The films were fabricated using CMC (CAS 9004–32-4), TA (CAS 1401–55-4), and glycerol (CAS 56–81-5). OLED fabrication used indium tin oxide (ITO), dipyrzino[2,3-f:2',3'-h]quinoxaline-2,3,6,7,10,11-hexacarbonitrile (HAT-CN), N,N'-di(1-naphthyl)-N,N'-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB), tris(8-hydroxyquinoline) aluminum (Alq₃), lithium fluoride (LiF), and aluminum (Al). All materials were obtained from Sigma-Aldrich and EMNI and used without further purification.

2.2 Fabrication of PBAT template and ECO-UV film

Figure 1 illustrates the fabrication process for the PBAT templates and ECO-UV films. The ECO-UV films composed of CMC, TA and glycerol served as biodegradable light-extraction layers. Used in template 6-inch silicon wafers (4science) were diced into 20 mm×20 mm pieces (Supplementary Fig. 4). Templates were prepared by dissolving PBAT at 2 wt% (PBAT_{low}) and 5 wt% (PBAT_{high}) in 10 mL of chloroform. The solutions were stirred at 50 °C for 2 h to ensure homogeneity and then spin-coated onto silicon wafers at 800 rpm for 8 s. The coated templates were annealed at 50 °C for 30 s to remove residual solvent and form microstructured surfaces. ECO-UV films were fabricated by mixing CMC and TA at a 7.5:1 mass ratio in deionized water. Glycerol was added at a 1:1 mass ratio relative to CMC to enhance flexibility. The mixture was stirred overnight at 30 °C, drop-cast onto the PBAT templates and dried at 30 °C for 8 h. The films were then delaminated without physical damage confirming effective structure transfer. Surface morphology was characterized using optical microscopy (BX43, Olympus) and field-emission scanning electron microscopy (S-4800, Hitachi High-Tech). Optical transmittance was measured using a UV–vis spectrophotometer (Lambda 1050, PerkinElmer).

2.3 Fabrication of OLED devices

Bottom-emission OLEDs were fabricated with the structure ITO (150 nm) / HAT-CN (5 nm) / NPB (55 nm) / Alq₃ (40 nm) / LiF (1 nm) / Al (100 nm). ITO-patterned glass substrates (20 mm×20 mm) with 2 mm×2 mm active

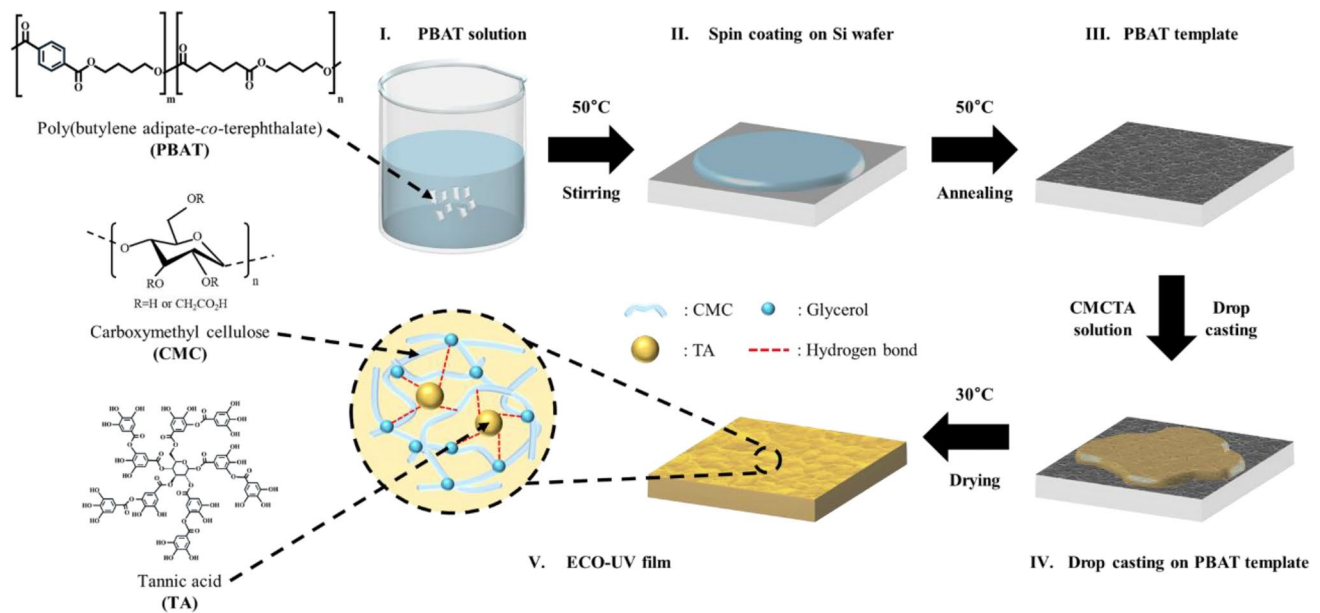


Fig. 1 Molecular structure and schematic of the fabrication process of PBAT template and ECO-UV film

emission areas were used (Supplementary Fig. 5). The substrates were ultrasonically cleaned in acetone for 15 min (JAC Ultrasonic 4020P, KODO), rinsed in isopropyl alcohol at 100 °C for 20 min (HSD180, GYROZEN), dried in an oven and treated with UV/ozone (AH-1700, AHTECH LTS) to improve surface adhesion. All organic and metal layers were thermally evaporated under 1.0×10^{-7} Torr. LiF was deposited at a rate of 0.5 Å/s while all other layers were deposited at 1.0 Å/s. The devices were encapsulated using a UV-curable epoxy resin inside a nitrogen-filled glove box to prevent degradation from oxygen and moisture (Supplementary Fig. 6). The effective refractive index of the ECO-UV film was estimated to be approximately 1.52, closely matching that of the index-matching liquid minimizing reflection losses at the interface. For optical evaluation, ECO-UV films were attached to the glass surface of the OLEDs using index-matching liquid ($n_D = 1.5200 \pm 0.0002$, Series A, Cargille Labs) to eliminate air gaps. Current density–voltage–luminance (J – V – L) characteristics and angular emission profiles were measured using a source meter (Keithley 2400, Tektronix), a spectroradiometer (PR-650, Photo Research) and a 2-inch integrating sphere (IS200-4).

3 Results and discussion

Figure 2 shows the surface morphologies of PBAT templates and ECO-UV films analyzed using optical microscopy (OM) and field-emission scanning electron microscopy (FE-SEM). The OM images reveal that both templates exhibit randomly distributed microscale surface features

ranging from several to tens of micrometers. The FE-SEM images show that PBAT_low forms larger and more microstructures with wider spacing, whereas PBAT_high contains smaller and irregular structures densely packed within narrower spacings. These morphological differences arise from viscosity flow behavior during spin coating [30, 31]. Rapid solvent evaporation during spin coating induces primary surface roughness. Meanwhile, gradual solvent removal during drying allows polymer chain rearrangement under surface tension forces, which stabilizes the transferred morphology. The lower viscosity of PBAT_low facilitates spreading and self-leveling, enabling the formation of well-defined and larger microstructures with uniform geometry. In contrast, the higher viscosity of PBAT_high restricts fluid mobility and promotes local accumulation, resulting in irregular and closely spaced features. In addition, the FE-SEM images of ECO-UV_low and ECO-UV_high confirm pattern replication from their respective PBAT templates. ECO-UV_low exhibits slightly larger and more uniformly distributed surface protrusions compared with ECO-UV_high. This reflects the larger and more continuous morphology derived from the PBAT_low template. Supplementary Figs. 1 and 2 further demonstrate consistent pattern replication across wide areas. These results suggest that surface structure facilitates balanced light scattering and suppresses internal reflection, thereby contributing to enhanced light extraction efficiency in OLEDs.

The optical properties of the ECO-UV films were evaluated by analyzing their visible transmittance, UV-blocking, and light-scattering characteristics. Figure 3 shows the total transmittance and haze spectra of the ECO-UV films. The

Fig. 2 **a** Optical microscope images of the PBAT_low and **b** PBAT_high template. **c** Field-emission scanning electron microscope images of PBAT_low and **d** PBAT_high template and **e** ECO-UV_low and **f** ECO-UV_high film

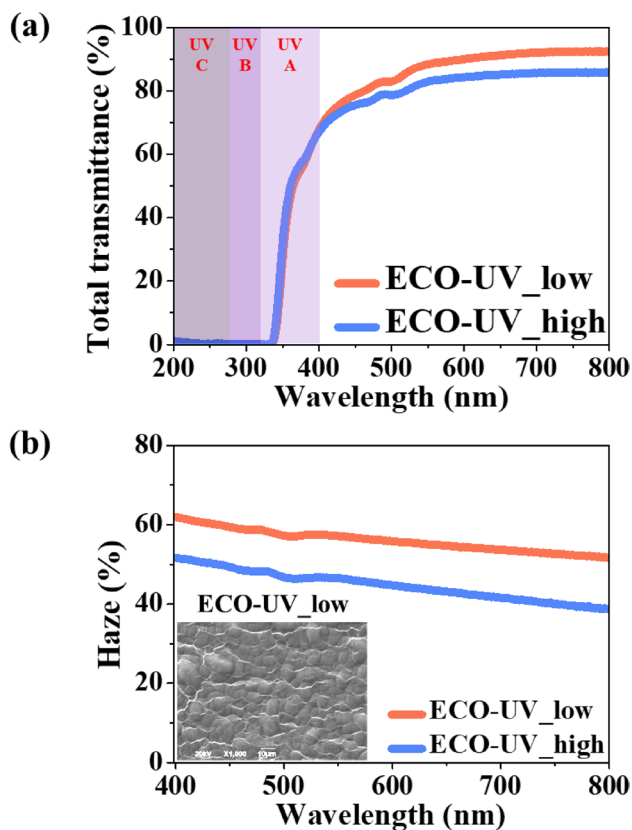
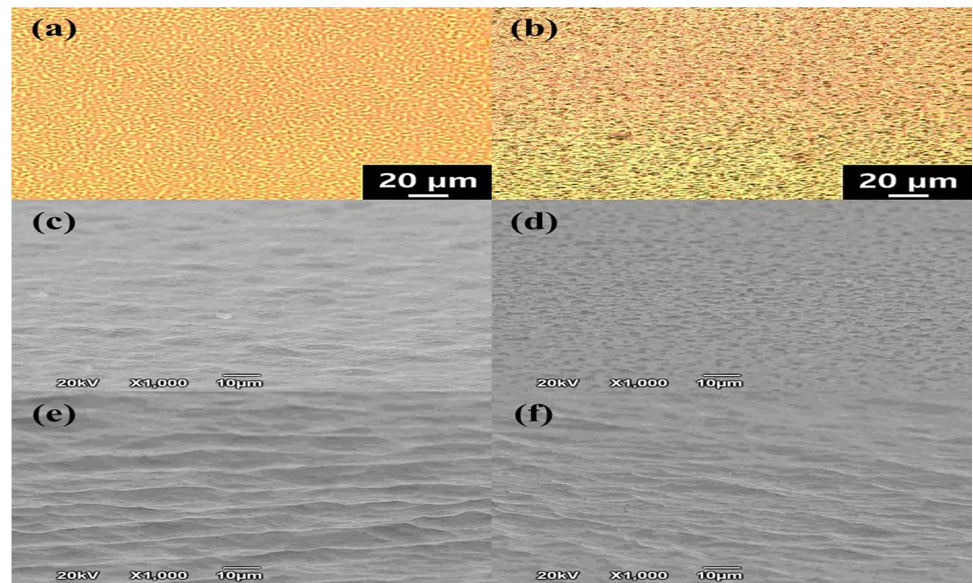


Fig. 3 **a** Total transmittance and **b** haze of ECO-UV_low and ECO-UV_high film. The inset in **b** shows field-emission scanning electron microscope image of ECO-UV_low film

total transmittance (T_t) is defined as the sum of the parallel transmittance (T_p) and diffusive transmittance (T_d), as expressed in Eq. (1). The haze value was calculated as the ratio of diffused to total transmittance, as shown in Eq. (2).

$$T_t = T_p + T_d \quad (1)$$

$$\text{Haze} = \frac{T_d}{T_t} * 100 \quad (2)$$

Both ECO-UV_low and ECO-UV_high exhibited high total transmittance (60–100%) across the visible range (400–800 nm), meeting the basic transparency requirements for light-extraction layers. ECO-UV_low showed slightly higher transmittance, attributed to its more uniformly replicated microstructures that reduce surface reflection and scattering loss [32, 33]. In the UV region (200–400 nm), both films exhibited sharp transmittance decreases, confirming strong UV-blocking property primarily originating from TA. The multiple phenolic hydroxyl groups of TA absorb UV radiation through π - π^* electronic transitions particularly in the UVA (315–400 nm) and UVB (280–315 nm) regions [14, 29]. Meanwhile, further reduction in the UVC range (100–280 nm) arises from intrinsic absorption and scattering of the polymer matrix [34]. Despite the relatively small TA content, both films achieved effective UV protection while maintaining high visible transparency, achieving a favorable balance between blocking and optical clarity. Haze analysis revealed that ECO-UV_low maintained approximately 55–60% haze across the visible range, whereas ECO-UV_high exhibited lower haze values of 40–50%. The higher haze of ECO-UV_low corresponds to its distributed microstructures observed in the FE-SEM inset, which promote efficient light scattering and suppress TIR.

Figure 4 compares the electroluminescence (EL), calibrated external quantum efficiency (EQE), and calibrated luminous efficiency (LE) of reference OLEDs and devices incorporating ECO-UV films. All devices exhibit similar

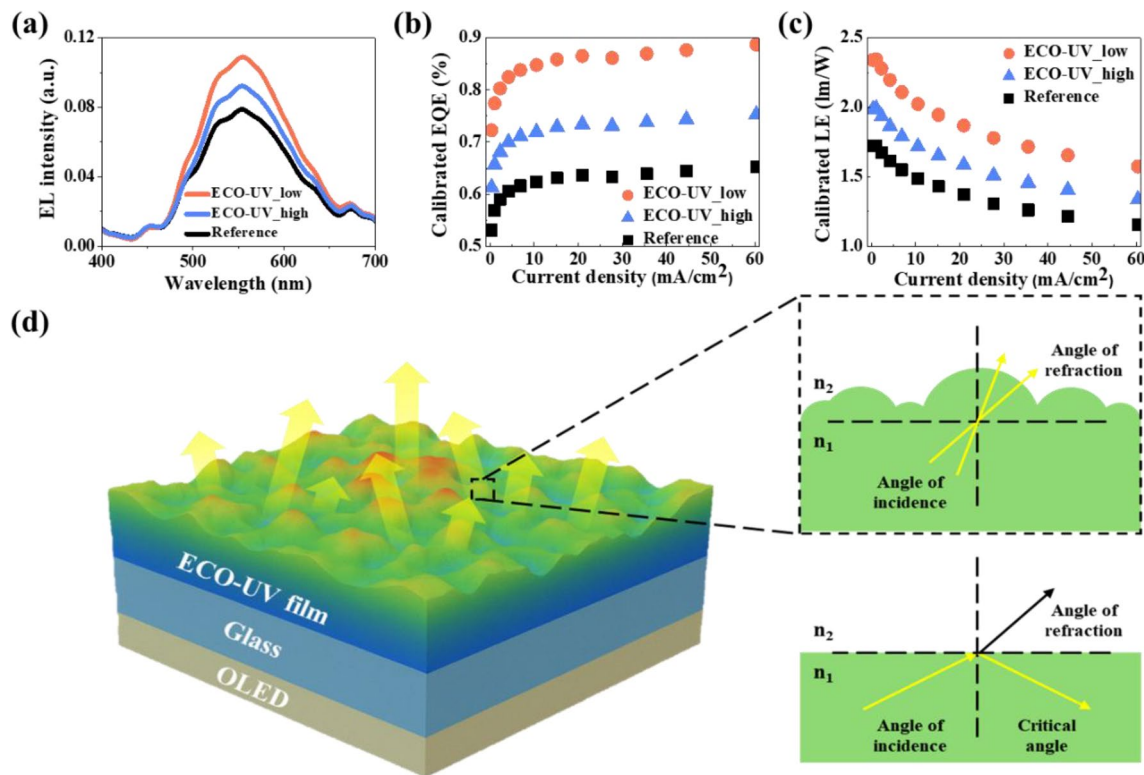


Fig. 4 **a** Electroluminescence, **b** calibrated external quantum efficiency, **c** calibrated luminous efficiency of reference OLEDs with ECO-UV film, and **d** schematic illustration of the light extraction mechanism by ECO-UV film

emission peaks near 550 nm, confirming that film attachment does not alter the intrinsic emission characteristics of the OLEDs. To quantify the light-extraction effect, the EL intensity curves of each device were integrated to calculate the total emitted intensity. Then, enhancement factors were determined by comparing the integrated EL areas of the reference and film-attached devices. As a result, ECO-UV_low and ECO-UV_high showed enhancement factors of approximately 36.08% and 15.48%, respectively. Calibrated EQE and LE values were then derived by applying these enhancement factors to the J - V - L characteristics of the reference OLEDs. Since the J - V - L data represent the emission characteristics measured along the normal direction of the device, incorporating the integrated EL enhancement factors provides a complementary means of evaluating efficiency improvements over a broader angular range. This approach offers a more comprehensive assessment of the light-extraction effect. Thus, the EQE increased from 0.65% for the reference to 0.89% for ECO-UV_low, while the LE improved from 1.72 to 2.35 lm/W. The enhancement achieved with ECO-UV_low is closely related to its surface morphology, characterized by larger and more uniformly replicated microstructures. These features promote multiple scattering and redirect confined photons into angles below the critical limit, thereby reducing TIR. In contrast, the denser but irregular surface features of ECO-UV_high

scatter light less efficiently, resulting in smaller improvements. The schematic illustration in the Figs. 4 further clarifies this mechanism, showing that the replicated microtextures enhance diffusive scattering. Overall, these results highlight that surface morphology is critical for improving efficiency, and that the ECO-UV film provides a more effective optical configuration for enhancing light extraction in OLEDs.

4 Conclusions

Biodegradable microstructured templates based on PBAT and external light-extraction films (ECO-UV) composed of CMC and TA were fabricated entirely through low temperature, solution-based processes, ensuring simplicity and environmental compatibility. The ECO-UV films exhibited high visible transmittance and strong UV blocking, maintaining a balance between transparency and protection. When applied to OLEDs, the ECO-UV film improved omnidirectional EL intensity by about 36% through multiple scattering and reduced TIR enabled by replicated microstructures. These results demonstrate that integrating biodegradable and naturally derived materials with simple, scalable processing offers a sustainable route to enhance OLED efficiency for next-generation eco-friendly display technologies.

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Data availability No datasets were generated or analysed during the current study.

Declarations

Conflict of interest The authors declare no competing interests.

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