

Bio-derived Casein–Tannic Acid Microlens Array Films with UV-Protective Function for OLED Light Extraction

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Abstract

We devised an eco-friendly and facile external film strategy that simultaneously enhances light extraction and mitigates ultraviolet (UV)-induced degradation in organic light-emitting diodes (OLEDs). To this end, we developed a milk-derived casein (CA)–tannic acid (TA) composite external-light-extraction film, referred to as a CATA film. CA was employed as a film-forming matrix, while TA promoted the formation of a crosslinked structure and imparted UV-protective functionality through its UV-absorbing characteristics. The film was fabricated via a simple process using deionized water as the sole solvent. Additionally, a microlens array (MLA) structure was replicated on the surface of the CATA film to enhance external-light-extraction efficiency by modifying the propagation path of light and thereby alleviating total internal reflection. Among the CATA MLA films with different TA concentrations, the CATA_0.5 film yielded a 57.8% enhancement in electroluminescence intensity compared with the reference device. Furthermore, owing to the UV-absorbing capability of TA, the UV-induced reduction in the current density of OLEDs was suppressed by 71.2%. These results indicate that bio-derived CATA MLA films can simultaneously improve the optical performance of OLEDs and suppress UV-induced photodegradation. Moreover, this study demonstrated a design strategy for an eco-friendly UV-protective external-light-extraction film that can be applied through external attachment without modifying the device architecture.

1. Introduction

Conventional petroleum-based synthetic polymers have inherent limitations owing to environmental concerns associated with their production, use, and disposal [1–3]. As a result, environmental issues and sustainability have received increasing attention, with the need for eco-friendly materials being widely recognized. Accordingly, bio-based materials that can replace conventional petroleum-derived polymers have been extensively studied [4–6]. Attempts have also been made to incorporate eco-friendly materials into optical films, and bio-based materials with biodegradability and environmental compatibility have been investigated for a wide range of applications [7, 8].

Specifically, in optical film applications, wherein large amounts of polymeric materials are used in displays, lighting, and other optoelectronic devices, replacing petroleum-based polymers with sustainable alternatives has become increasingly important. At the same time, these films must retain their optical functionality, including the ability to control and extract light efficiently. In light-emitting devices, effective extraction of the generated light is essential for improving device efficiency. Organic light-emitting diodes (OLEDs) are self-emissive devices that generate light directly through charge recombination and exhibit excellent optical properties. Therefore, OLEDs have been widely utilized in high-resolution displays and lighting applications [9–11]. However, because OLEDs consist of multilayer thin-film structures, total internal reflection (TIR) occurs because of refractive index differences between the layers. Consequently, a substantial portion of the generated light is trapped inside the device through substrate and waveguided modes [12, 13]. The external quantum efficiency (EQE, η_{ext}) can be

expressed as the product of the internal quantum efficiency (IQE, η_{int}) and light-extraction efficiency ($\eta_{coupling}$), as follows [12].

$$\eta_{ext} = \eta_{int} \times \eta_{coupling} = \gamma \times \eta_{exc} \times \phi_p \times \eta_{coupling}$$

1

,

where γ is the charge balance factor; η_{exc} is the exciton generation efficiency; and ϕ_p is the intrinsic radiative efficiency resulting from radiative recombination. Among these factors, $\eta_{coupling}$ is a critical limiting factor that determines the EQE of OLEDs. Therefore, even when the IQE is high, the actual EQE is strongly limited by the light-extraction efficiency and generally remains at approximately 20–30% [12, 13].

However, the performance of OLEDs is governed not only by light-extraction efficiency but also by their stability under external environmental conditions [14]. Particularly, OLEDs are vulnerable to ultraviolet (UV) exposure, which can induce photodegradation in organic emissive layers and charge transport layers. Such UV-induced degradation causes changes in the chemical structures of organic molecules, thereby deteriorating their charge transport properties and emission efficiency. Moreover, it increases non-radiative decay pathways, resulting in the degradation of the electrical and optical performance of the device. Photodegradation is a major factor that reduces device lifetime during long-term operation [15]. Therefore, to ensure both high performance and long-term stability of OLEDs, simultaneously improving the light-extraction efficiency and suppressing UV-induced degradation is necessary.

To enhance light-extraction efficiency, various technologies have been proposed, including microlens arrays (MLAs) [16, 17], nano-patterned structures [18], and scattering layers [19]. Among them, external-light-extraction strategies have the advantage of effectively extracting light lost through the substrate mode without modifying the internal device structure. However, some approaches require complicated microfabrication processes, such as photolithography, which can increase their processing costs; this limits their applicability to large-area fabrication [20, 21]. Furthermore, conventional external-light-extraction films are commonly fabricated using synthetic polymers such as polydimethylsiloxane (PDMS), poly(methyl methacrylate) (PMMA), and UV-curable resins. These materials have limitations in terms of their environmental impact and sustainability, thus necessitating their replacement with sustainable alternatives [22, 23]. Therefore, developing eco-friendly bio-based light-extraction films is highly desirable [24, 25].

To overcome these limitations, we designed a CATA composite film based on the interaction between casein (CA) and tannic acid (TA). CA is a milk-derived protein-based biopolymer obtained from dairy processing and has attracted attention as a sustainable bio-based material because of its excellent film-forming ability and biodegradability. Additionally, CA contains various functional groups, such as amide and carboxyl groups, which enable interactions with other molecules [26]. TA is a natural polyphenolic compound containing multiple phenolic groups and can form a crosslinked structure through hydrogen

bonding with proteins and exhibits light absorption characteristics in the UV region [27, 28]. Therefore, the combination of CA and TA can serve as an effective eco-friendly strategy for simultaneously achieving crosslinked network formation and UV-protective functionality.

In this study, we introduced an MLA structure onto a CATA film to control the propagation path of light, thereby alleviating TIR and improving the external-light-extraction efficiency of OLEDs. Additionally, we utilized the UV-absorbing capability of TA to suppress photodegradation of OLED devices. Through this approach, we aim to present an effective strategy for simultaneously enhancing the light-extraction efficiency of OLEDs and suppressing UV-induced photodegradation using an eco-friendly bio-based external-light-extraction film.

2. Experimental

2.1. Preparation of CATA MLA films

CA (CAS: 9005-46-3), TA (CAS: 1401-55-4), and glycerol (gly, CAS: 56-81-5) were purchased from Sigma-Aldrich (USA) and used without further purification. A CA solution was prepared by dissolving CA in deionized water at a concentration of 10 wt%, followed by stirring at 500 rpm for 1 h at 60°C. Subsequently, TA was added to 10 mL of the prepared CA solution at concentrations of 0, 0.5, 1.0, and 1.5 wt%. Next, 0.3 mL of gly was added as a plasticizer to improve the film's flexibility and reduce the brittleness. The mixture was then stirred at 60°C for 6 h. The CATA MLA films prepared with the aforementioned TA concentrations were denoted as CATA_0, CATA_0.5, CATA_1.0, and CATA_1.5, respectively. During this process, the outside of the vial was wrapped with aluminum foil to block light and minimize light-induced yellowing associated with TA absorption. The mixed solution was cast in 10 mL portions onto Petri dishes equipped with an MLA mold having a diameter and depth of approximately 20 and 10 μm , respectively. The cast films were dried overnight in an oven at 35°C. After drying, the formed CATA MLA films were peeled off from the Petri dishes and cut into $4 \times 4 \text{ mm}^2$ pieces to completely cover the OLED pixels. Before attaching the CATA MLA film to the OLED substrate, a small amount of refractive index matching oil ($n = 1.5200 \pm 0.0002$, Refractive Index Liquid Series A, Cargille Laboratories, USA) was applied onto the OLED substrate. This process minimized the refractive index mismatch caused by the air gap between the OLED substrate and the CATA MLA film.

The surface morphology of the films was observed via optical microscopy (BX43, Olympus, Japan) and field-emission scanning electron microscopy (FE-SEM; SU8230, Hitachi, Japan). Fourier transform infrared (FT-IR) spectra were obtained using an FT-IR spectrometer (Nicolet iS50, Thermo Fisher Scientific, USA) in the range of $4000\text{--}400 \text{ cm}^{-1}$. The optical transmittance was analyzed using a UV–visible–near-infrared (UV–Vis–NIR) spectrophotometer (Lambda 1050+, PerkinElmer, USA) in the wavelength range of 200–800 nm.

2.2. OLED fabrication and characterization

Bottom-emitting OLED devices were fabricated with the following structure:

indium tin oxide (ITO) (150 nm)/dipyrzino[2,3-f:2',3'-h]quinoxaline-2,3,6,7,10,11-hexacarbonitrile (HAT-CN) (5 nm)/N,N'-di(1-naphthyl)-N,N'-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB) (55 nm)/tris(8-hydroxyquinoline)aluminum (Alq3) (40 nm)/lithium fluoride (LiF) (1 nm)/aluminum (Al) (100 nm).

Patterned ITO glass substrates were ultrasonically cleaned in acetone for 20 min, followed by cleaning in boiling isopropyl alcohol (IPA) for 20 min. The substrates were then treated with UV-ozone to remove organic contaminants and activate the surface, after which they were transferred to a thermal evaporation system. All layers, including LiF and Al, were deposited by thermal evaporation under high vacuum conditions below 1.0×10^{-6} Torr. After the deposition, the OLED devices were encapsulated in a nitrogen-filled glovebox to prevent the ingress of oxygen and moisture. The electrical and electroluminescent characteristics of the fabricated OLED devices were measured using a source meter (Keithley 2400, Tektronix, USA), a spectroradiometer (PR-650, Photo Research, USA), and a 2-inch integrating sphere (IS200-4, Thorlabs, USA). To evaluate the UV-protective effect of the CATA MLA film, OLED devices with and without the CATA_0.5 film were exposed to UV light at 184 and 254 nm for 1 and 2 h, using a UV-ozone cleaner (AH-1700, AhTech LTS, Republic of Korea).

3. Results and discussion

3.1. Formation mechanism of TA-crosslinked CA films

We propose a formation mechanism for CATA composite films based on hydrogen-bonding interactions between CA and TA. CA is a protein-based biopolymer containing functional groups such as amide (-CONH-) and carboxyl (-COOH) groups, which enable various intermolecular interactions (Fig. 1(a)) [26]. In contrast, TA is a polyphenolic compound with multiple phenolic hydroxyl (-OH) groups, which can induce interactions with proteins through these functional groups while exhibiting strong absorption in the ultraviolet region (Fig. 1(b)) [27, 28]. Based on these characteristics, a non-covalently crosslinked structure can be formed between CA and TA, mainly through hydrogen bonding.

As shown in Fig. 1(c), hydrogen bonds can form between the amide and carboxyl groups of CA and hydroxyl groups of TA, leading to the construction of a three-dimensional crosslinked network. These interactions contribute to the formation of a bonding network within the CA matrix, thus providing the structural basis for the CATA composite film [29, 30]. Based on this mechanism, the prepared CATA solution was cast onto an MLA mold, as illustrated in Fig. 1(d), to form a film with a regular microstructured surface. This microstructure can alter the propagation path of light through refraction and scattering effects, enabling trapped light to be effectively extracted and consequently contributing to enhanced external-light-extraction efficiency [17].

3.2. Surface morphology of CATA MLA films

Figure 2(a) shows an optical microscopy image of the fabricated CATA_0.5 film, while Fig. 2(b) presents the corresponding surface SEM image of the same sample. Both images confirm that the MLA structure

was uniformly formed on the film surface and that the mold structure was replicated with high fidelity. The MLA mold used in this study had a diameter and depth of approximately 20 and 10 μm , respectively.

Although surface observations confirmed the formation of a regular microstructure, they were limited in revealing the internal geometry of the replicated structure. Therefore, cross-sectional SEM analysis was performed after cutting the film, as shown in Fig. 2(c). The cross-sectional image confirmed that the concave microstructures formed on the surface extended uniformly into the depth of the film. These regular microstructures can alter the propagation path of light and induce scattering, thereby reducing internal optical losses in OLEDs and contributing to improved external-light-extraction efficiency.

3.3. Optical properties of CATA MLA films

Figure 3(a) shows the total transmittance spectra of the CATA MLA films measured using a UV–vis spectrophotometer. In the visible wavelength region of 400–800 nm, all samples generally maintained high transmittance, while the total transmittance gradually decreased with increasing TA content. Additionally, the transmittance of all samples significantly decreased in the UV region below 400 nm. This decrease is attributed to the light absorption of the aromatic phenolic structures in TA, which originates from π – π^* transitions in the UV region [31]. Particularly, the TA-free sample (CATA_0) exhibited relatively high transmittance in the UV region, whereas the TA-containing samples showed transmittance values close to zero below 350 nm. These results confirm that CATA MLA films possess excellent UV-blocking properties.

To further evaluate the light scattering properties, the diffuse transmittance (T_d) was calculated from the total transmittance (T_t) and parallel transmittance (T_p), as shown in Eq. (2), and the *haze* value was then calculated using Eq. (3) (Fig. 3(b)). *Haze* is an indicator of light scattering within a film, where higher values correspond to stronger light diffusion. In this study, the CATA MLA films exhibited a high *haze* value of approximately 85%. This result is attributed to enhanced light scattering caused by the MLA structure formed on the film surface. Particularly, the MLA structure changes the propagation path of incident light, thereby promoting light diffusion. The increased light scattering is expected to alleviate optical losses caused by total internal reflection and contribute to improved light-extraction efficiency [17].

$$T_t = T_p + T_d$$

2
,

$$haze = \frac{T_d}{T_t} \times 100$$

3
.

3.4. FT-IR analysis of CATA films

Figure 4 shows the FT-IR spectra obtained to analyze the chemical interactions in the CA-based films. The molecular interactions induced by the additives were investigated by comparing the pure CA film with the films containing TA and gly. The broad peak observed at approximately 3200–3300 cm⁻¹ corresponds to O–H and N–H stretching vibrations, indicating hydrogen-bonding interactions formed within the films [29]. Upon the addition of TA, changes in this peak were observed, which are attributed to the enhancement of hydrogen-bonding interactions.

The spectral changes were further analyzed based on the characteristic amide bands of CA, including Amide I (~ 1630 cm⁻¹), Amide II (~ 1530 cm⁻¹), and Amide III (~ 1200–1300 cm⁻¹) [32]. In the TA-containing films, changes in the intensity of the Amide I band were observed, along with slight shifts in the positions of the Amide II and Amide III peaks (Table 1). These peak shifts suggest the formation of intermolecular interactions between the functional groups of CA and the hydroxyl groups of TA [29].

Upon the addition of gly, a new peak appeared at approximately 1000–1100 cm⁻¹, which is attributed to the C–O stretching vibration of gly [33]. This result indicates that gly was uniformly dispersed within the film matrix. Overall, the FT-IR results demonstrate the presence of hydrogen-bonding interactions between CA and TA, supporting the formation of a crosslinked network structure within the films.

Table 1
Characteristic FT-IR peak positions (cm⁻¹) of CA-based films

CA	O-H/N-H	Amide I	Amide II	Amide III	C-O
	3280	1632	1533	1239	1080
CAGly	3274	1630	1535	1237	1040
CATA (w/o gly)	3281	1636	1530	1201	1041
CATA (w/ gly)	3275	1636	1534	1201	1039

3.5. OLED performance of CATA MLA films

To evaluate the external-light-extraction performance of the CATA MLA films, they were attached to the outer surface of the OLED glass substrate, and the device characteristics were measured. Figure 5(a) shows the OLED device structure used in this study and the position of the externally attached CATA MLA film. Figure 5(b) presents the current density–voltage (*J*–*V*) characteristics before and after the film attachment. Negligible changes in *J* and turn-on *V* were observed for all devices, indicating that the CATA MLA films did not affect the electrical characteristics of the OLED devices. Figure 5(c) shows the electroluminescence (EL) spectra measured using an integrating sphere. The EL intensity increased for all devices with CATA MLA films compared with the reference device. Particularly, the CATA_0 film exhibited the highest EL intensity, which can be attributed to its high optical transmittance and low absorption.

However, while the TA-free CATA_0 film showed excellent light-extraction performance, it had limitations in terms of additional functionalities, such as UV-blocking capability and crosslinked network formation. Therefore, when both light-extraction performance and functionality are considered, TA-containing films can be a more suitable option. Among them, the CATA_0.5 film showed a promising balance between these two properties. The device with the CATA_0.5 film exhibited an approximately 57.8% increase in EL intensity compared with the reference device (Table 2). This improvement can be attributed to the light scattering by the surface MLA structure, along with the relatively high transmittance of the film.

As the TA content increased, the EL intensity gradually decreased. This trend can be attributed to the decrease in film transmittance caused by increased light absorption with increasing TA content, as confirmed in Fig. 3 [31]. Although the *haze* values remained at a similar level or slightly increased with increasing TA concentration, the reduction in transmittance became more dominant, leading to an overall decrease in light-extraction efficiency. Figure 5(d) shows the calibrated EQE values estimated by scaling the measured EQE of the reference OLED device with the relative enhancement in EL intensity obtained from integrating sphere measurements. These values were used to evaluate the relative efficiency enhancement among the devices. The reference device exhibited an EQE of 0.628%, whereas the EQE increased to 0.991% when the CATA_0.5 film was applied, corresponding to a 1.58-fold enhancement. Thus, these results demonstrate that the application of the CATA MLA film effectively improved the light-extraction efficiency of OLEDs. Notably, this enhancement was achieved through simple external attachment without modifying the OLED device structure.

Table 2
Relative EL intensity and calibrated EQE values of OLED devices incorporating CATA MLA films

Reference	Integrated EL intensity (a.u.)	Relative EL intensity (%)	Calibrated EQE (%)
	2.957	100	0.628
CATA_0	4.918	166.3	1.044
CATA_0.5	4.667	157.8	0.991
CATA_1.0	4.552	152.9	0.960
CATA_1.5	4.133	139.8	0.878

3.6. UV-protective effect of CATA MLA films

TA contains multiple phenolic groups, and these structures can effectively absorb light in the UV region. To investigate the effect of this property on the photodegradation of OLED devices, the device characteristics after UV irradiation were compared between devices with and without the CATA_0.5 film, which was selected based on its balanced light-extraction and UV-protective performance. Figures 6(a) and 6(b) show the J–V characteristics after 1 and 2 h of UV irradiation at 184 and 254 nm, respectively. J

decreased in all devices upon UV exposure. The device without the film (w/o film) exhibited a substantial decrease, whereas the device with the CATA_0.5 film (w/ film) showed significantly suppressed degradation.

At 6 V, J of the device without the film decreased markedly from approximately 220 mA cm^{-2} before UV irradiation to approximately 105 mA cm^{-2} after 2 h of irradiation. In contrast, J of the device with the CATA_0.5 film decreased only to approximately 160 mA cm^{-2} , indicating that degradation was effectively suppressed. This result quantitatively demonstrates the difference in degradation behavior depending on the presence or absence of the film under the same voltage condition. This trend is more clearly observed in J reduction ratio shown in Fig. 6(c). Based on this reduction ratio, the UV-induced reduction in J after 1 h of UV irradiation was suppressed by approximately 71.2% upon the application of the CATA_0.5 film.

A similar trend was observed in the luminance characteristics. Figures 6(d) and 6(e) show the luminance–voltage (L – V) characteristics after UV irradiation. L also decreased upon UV irradiation; however, its reduction was markedly suppressed in the device with the film. At 6 V, L of the device without the film decreased from approximately 3400 cd m^{-2} before UV irradiation to approximately 1750 cd m^{-2} after 2 h of irradiation. In contrast, the device with the CATA_0.5 film maintained an L of approximately 2530 cd m^{-2} . As shown in Fig. 6(f), the L reduction ratio after 1 h of UV irradiation was also suppressed by approximately 60%, showing a trend consistent with the J results.

These results suggest that incident UV light was effectively blocked by the UV-absorbing characteristics of TA, thereby suppressing UV-induced photodegradation [31]. Thus, the CATA MLA film served not only as an external-light-extraction layer but also as an effective UV-protective layer capable of mitigating device performance degradation under UV exposure.

4. Conclusions

In this study, a bio-derived casein–tannic acid (CATA) composite film with a microlens array (MLA) structure was developed and applied as an external-light-extraction layer for OLED devices. The incorporation of TA induced the formation of a hydrogen-bonding-based crosslinked network within the CA matrix while simultaneously imparting UV-blocking properties. OLED devices with the CATA_0.5 film exhibited a 57.8% enhancement in electroluminescence (EL) intensity compared with the reference device. This improvement is attributed to light scattering induced by the MLA structure, which alleviates optical losses caused by total internal reflection and enhances light-extraction efficiency.

Additionally, owing to the UV-absorbing characteristics of TA, the decreases in current density and luminance of OLED devices after UV irradiation were suppressed by 71.2% and 60%, respectively, thereby effectively mitigating UV-induced photodegradation and performance degradation. Notably, the proposed CATA MLA film can be applied through simple external attachment to OLED devices, thereby enabling improved optical performance without modifying the existing device structure, and highlighting

its practical applicability. These results demonstrate that eco-friendly bio-based CATA MLA films provide an effective strategy for simultaneously enhancing light-extraction efficiency and suppressing UV-induced photodegradation in OLED devices.

Declarations

Author Contribution

S.K., S.H. and J.L. conceived the idea and designed the experiments. S.K., S.H., B.S., S.O, and H.K. fabricated and characterized the devices. S.K., H.J. and S.J. analyzed the data. All authors discussed the results and contributed to the manuscript. B.S. J.-H.L and J.L. guided all aspects of the work.

References

1. R. A. Gross, B. Kalra, Biodegradable Polymers for the Environment, *Science* 297 (5582), 803–807 (2002)
2. R. Geyer, J. R. Jambeck, K. L. Law, Production, use, and fate of all plastics ever made, *Sci. Adv.* 3 (7), e1700782 (2017)
3. J. R. Jambeck, R. Geyer, C. Wilcox, T. R. Siegler, M. Perryman, A. Andrady, R. Narayan, K. L. Law, Plastic waste inputs from land into the ocean, *Science* 347 (6223), 768–771 (2015)
4. M. Shen, B. Song, G. Zeng, Y. Zhang, W. Huang, X. Wen, W. Tang, Are biodegradable plastics a promising solution to solve the global plastic pollution?, *Environ. Pollut.* 263, 114469 (2020)
5. A. Gandini, Polymers from renewable resources: a challenge for the future of macromolecular materials, *Macromolecules* 41 (24), 9491–9504 (2008)
6. J.-W. Rhim, H.-M. Park, C.-S. Ha, Bio-nanocomposites for food packaging applications, *Prog. Polym. Sci.* 38 (10–11), 1629–1652 (2013)
7. H. Jang, J. Lee, S. Han, B. Sung, C. W. Joo, H. Park, J. Kim, H. Kang, S. Kim, J.-H. Lee, Y. H. Kim, J. Lee, Light extraction film for organic light-emitting diodes based on an eco-friendly template comprising cellulose and polyphenon 60, *J. Inf. Disp.* 26 (4), 369–379 (2025)
8. S. Han, B. Sung, H. Jang, J. Lee, S. Woo, D. Kim, H.-R. Joe, J. W. Han, Y. H. Kim, J.-H. Lee, J. Lee, Spontaneously formed cellulose-based random micro-textured film for light extraction in organic light-emitting diodes, *J. Inf. Disp.* 25 (4), 341–348 (2024)
9. J. Kang, G. W. Beak, J. Y. Lee, J. Kwak, J.-H. Park, Advances in display technology: augmented reality, virtual reality, quantum dot-based light-emitting diodes, and organic light-emitting diodes, *J. Inf. Disp.* 25 (3), 219–234 (2024)
10. H. Lee, H. Cho, C.-W. Byun, J.-H. Han, B.-H. Kwon, S. Choi, J. Lee, N. S. Cho, Color-tunable organic light-emitting diodes with vertically stacked blue, green, and red colors for lighting and display applications, *Opt. Express* 26 (14), 18351–18361 (2018)

11. S.-J. Zou, Y. Shen, F.-M. Xie, J.-D. Chen, Y.-Q. Li, J.-X. Tang, Recent advances in organic light-emitting diodes: toward smart lighting and displays, *Mater. Chem. Front.* 4 (3), 788–820 (2020)
12. K. Saxena, V. K. Jain, D. S. Mehta, A review on the light extraction techniques in organic electroluminescent devices, *Opt. Mater.* 32 (1), 221–233 (2009)
13. W. Brütting, J. Frischeisen, T. D. Schmidt, B. J. Scholz, C. Mayr, Device efficiency of organic light-emitting diodes: Progress by improved light outcoupling, *Phys. Status Solidi A* 210 (1), 44–65 (2013)
14. H. Aziz, Z. D. Popovic, Degradation phenomena in small-molecule organic light-emitting devices, *Chem. Mater.* 16 (23), 4522–4532 (2004)
15. S.-K. Kwon, J.-H. Baek, H.-C. Choi, S.-K. Kim, R. Lampande, R. Pode, J.-H. Kwon, Degradation of OLED performance by exposure to UV irradiation, *RSC Adv.* 9, 42561–42568 (2019)
16. J. Lee, B. Sung, S. Han, H.-B. Kim, J. W. Han, Y. H. Kim, J.-H. Lee, J. Lee, Bio-material-based deformable out-coupling film with UV-blocking properties for stabilized organic light-emitting diodes, *J. Inf. Disp.* 26 (3), 197–207 (2025)
17. J.-H. Han, J. Moon, D.-H. Cho, J.-W. Shin, H.-Y. Chu, J.-I. Lee, N. S. Cho, J. Lee, Luminescence enhancement of OLED lighting panels using a microlens array film, *J. Inf. Disp.* 19 (4), 179–184 (2018)
18. J.-W. Shin, D.-H. Cho, J. Moon, C. W. Joo, S. K. Park, J. Lee, J.-H. Han, N. S. Cho, J. Hwang, J. W. Huh, H. Y. Chu, J.-I. Lee, Random nano-structures as light extraction functionals for organic light-emitting diode applications, *Org. Electron.* 15 (1), 196–202 (2014)
19. H.-W. Chang, J. Lee, S. Hofmann, Y. H. Kim, L. Müller-Meskamp, B. Lüssem, C.-C. Wu, K. Leo, M. C. Gather, Nano-particle based scattering layers for optical efficiency enhancement of organic light-emitting diodes and organic solar cells, *J. Appl. Phys.* 113 (20), 204502 (2013)
20. Y. R. Do, Y. C. Kim, Y.-W. Song, H. Jeon, S.-H. Kim, Y.-H. Lee, Enhanced Light Extraction from Organic Light-Emitting Diodes with 2D SiO₂/SiN_x Photonic Crystals, *Adv. Mater.* 15 (14), 1214–1218 (2003)
21. Y. Sun, S. R. Forrest, Enhanced light out-coupling of organic light-emitting devices using embedded low-index grids, *Nat. Photonics* 2 (8), 483–487 (2008)
22. Y. Sun, S. R. Forrest, Organic light emitting devices with enhanced outcoupling via microlenses fabricated by imprint lithography, *J. Appl. Phys.* 100 (7), 073106 (2006)
23. S. Möller, S. R. Forrest, Improved light out-coupling in organic light emitting diodes employing ordered microlens arrays, *J. Appl. Phys.* 91 (5), 3324–3327 (2002)
24. Y. Habibi, L. A. Lucia, O. J. Rojas, Cellulose Nanocrystals: Chemistry, Self-Assembly, and Applications, *Chem. Rev.* 110 (6), 3479–3500 (2010)
25. D. Klemm, B. Heublein, H. P. Fink, A. Bohn, Cellulose: Fascinating Biopolymer and Sustainable Raw Material, *Angew. Chem. Int. Ed.* 44 (22), 3358–3393 (2005)
26. A. Gennadios, Edible films and coatings from proteins, in *Proteins in Food Processing*, R. Y. Yada, Ed., Woodhead Publishing Ltd, 442–467 (2004)

27. C. Chen, H. Yang, X. Yang, Q. Ma, Tannic acid: a crosslinker leading to versatile functional polymeric networks: a review, *RSC Adv.* 12 (13), 7689–7711 (2022)
28. A. Scalbert, Antimicrobial properties of tannins, *Phytochemistry* 30 (12), 3875–3883 (1991)
29. M. L. Picchio, Y. G. Linck, G. A. Monti, L. M. Gugliotta, R. J. Minari, C. I. A. Igarzabal, Casein films crosslinked by tannic acid for food packaging applications, *Food Hydrocoll.* 84, 424–434 (2018)
30. W. Zhang, S. Roy, P. Ezati, D.-P. Yang, J.-W. Rhim, Tannic acid: A green crosslinker for biopolymer-based food packaging films, *Trends Food Sci. Technol.* 136, 11–23 (2023)
31. A. Pizzi, Tannins: Prospectives and Actual Industrial Applications, *Biomolecules* 9 (8), 344 (2019)
32. A. Barth, Infrared spectroscopy of proteins, *Biochim. Biophys. Acta Bioenerg.* 1767 (9), 1073–1101 (2007)
33. G. Kovtun, D. Casas, T. Cuberes, Influence of Glycerol on the Surface Morphology and Crystallinity of Polyvinyl Alcohol Films, *Polymers* 16 (17), 2421 (2024)

Figures

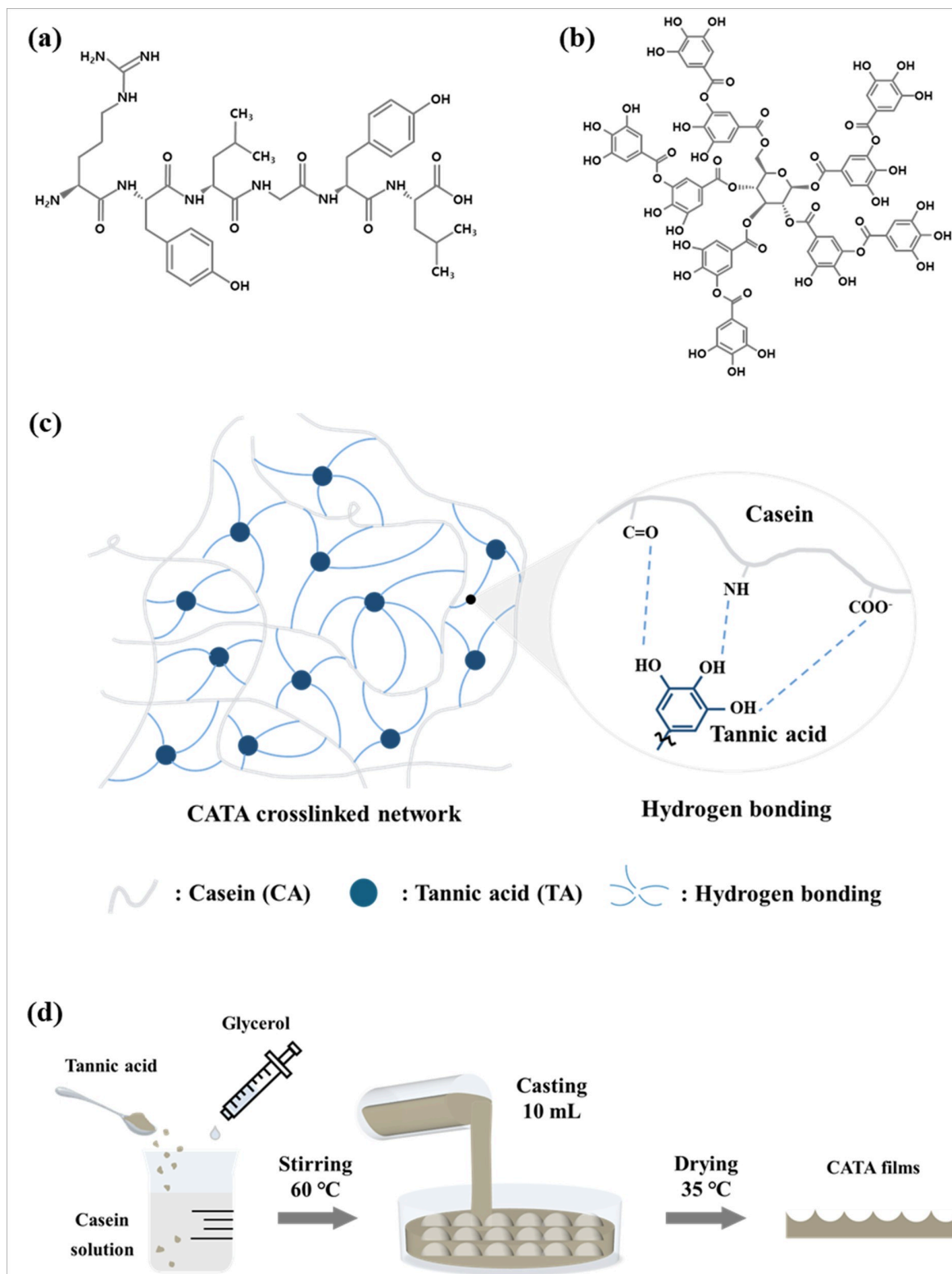


Figure 1

Schematic of (a) molecular structure of CA, (b) molecular structure of TA, (c) TA-induced hydrogen-bonded crosslinked network in the CATA film, and (d) fabrication process of the CATA MLA film

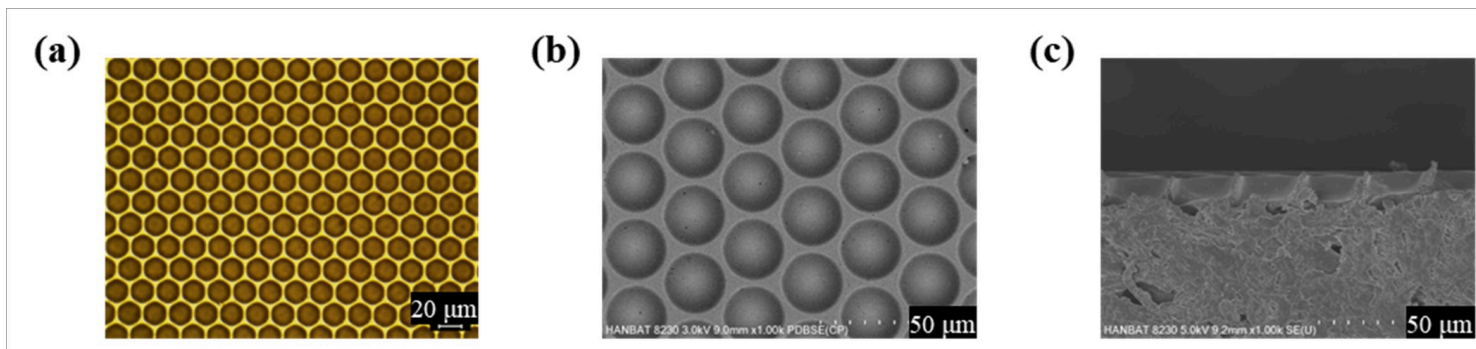


Figure 2

Surface morphology of CATA_0.5 film: (a) optical microscopy (OM) image of the film surface; (b) top-view SEM image; (c) cross-sectional SEM image

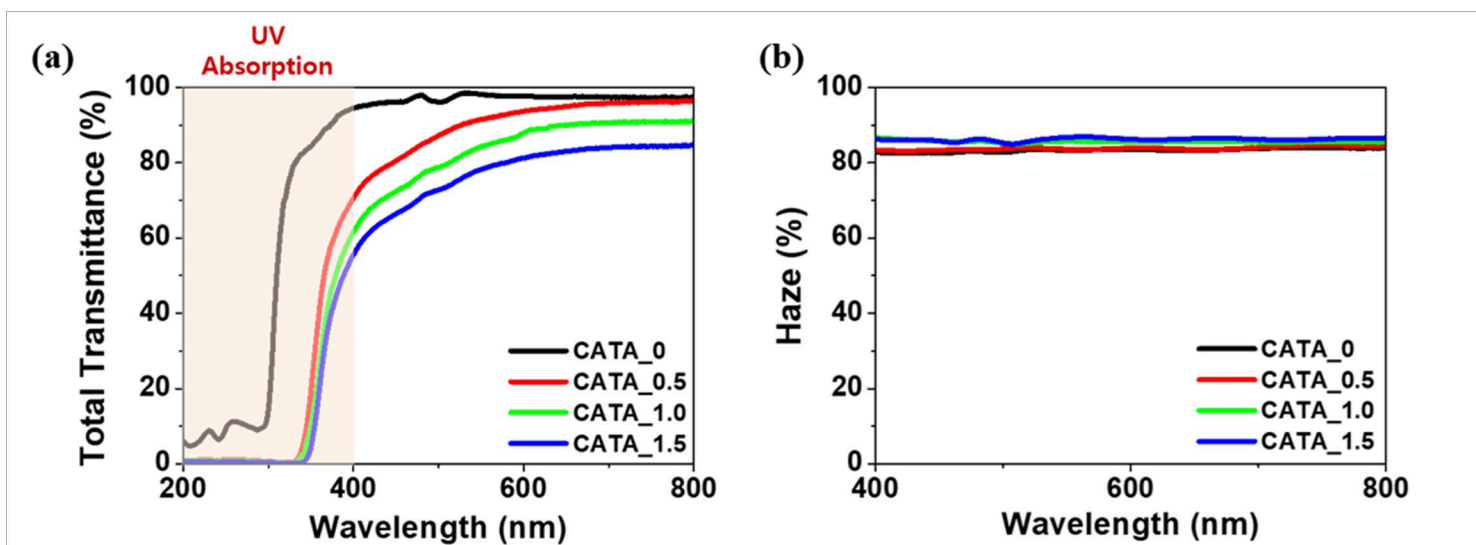


Figure 3

Optical properties of CATA MLA films with different TA concentrations (0–1.5 wt%): (a) total transmittance spectra; (b) haze spectra

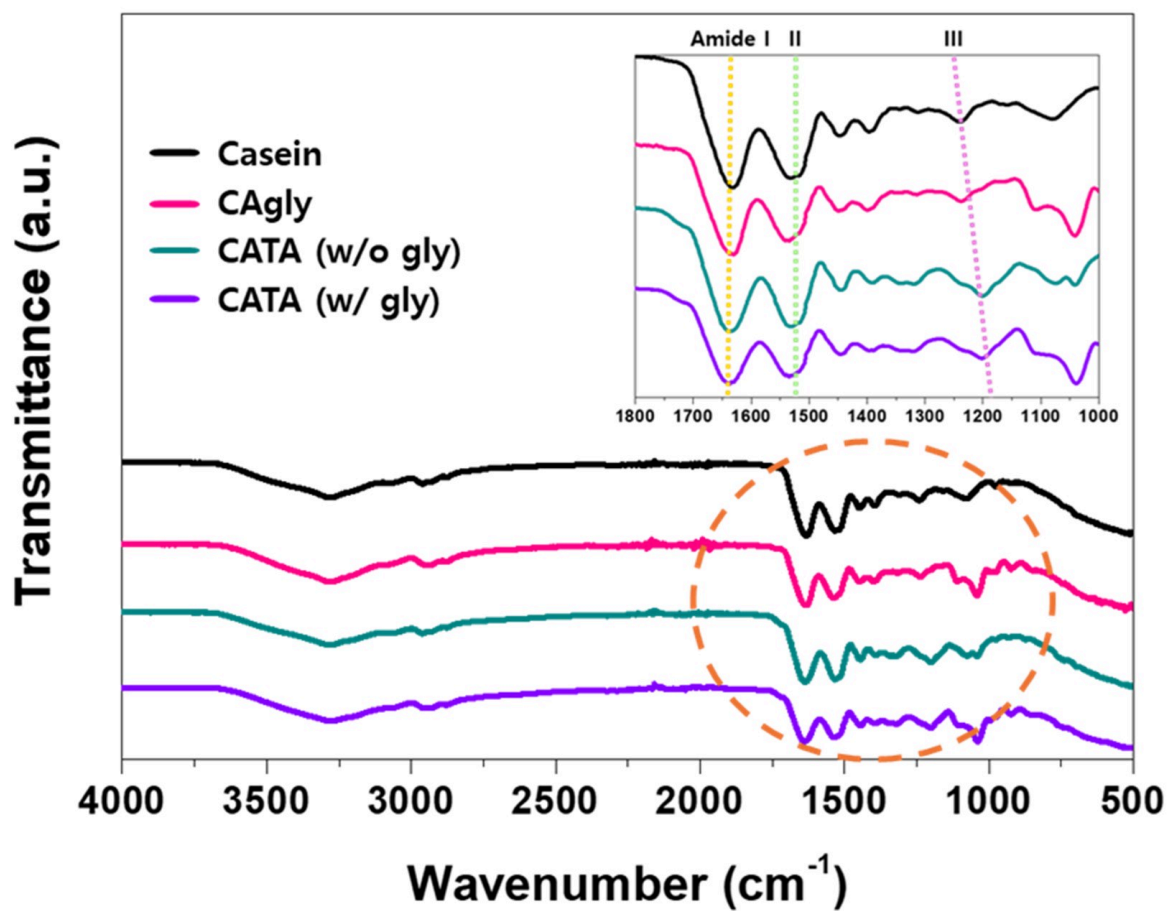


Figure 4

FT-IR spectra of CA-based films with various additives

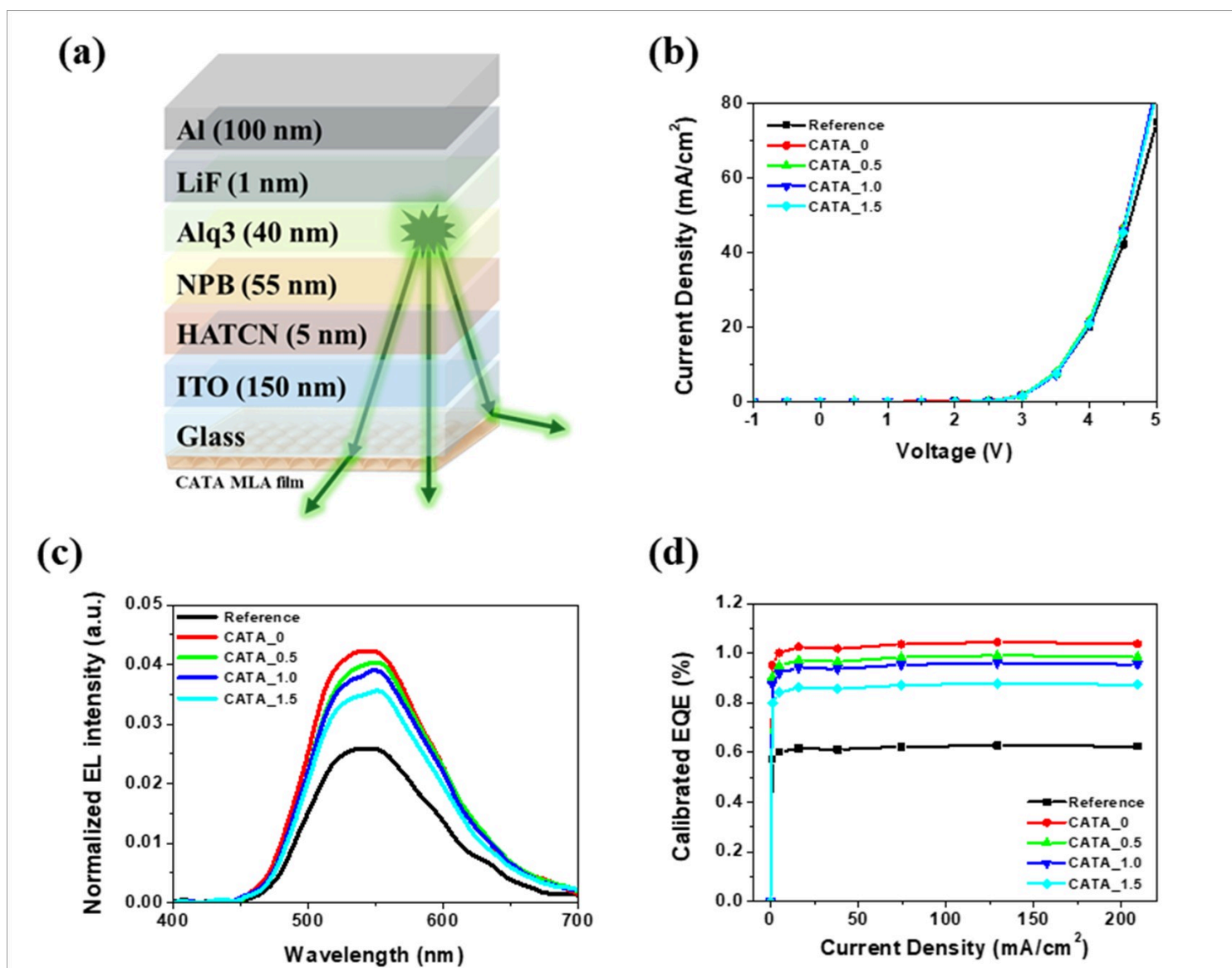


Figure 5

Performance of OLED devices incorporating CATA MLA films: (a) schematic of the OLED device structure with externally attached CATA MLA film; (b) J - V characteristics; (c) EL spectra measured using an integrating sphere; (d) calibrated EQE as a function of J

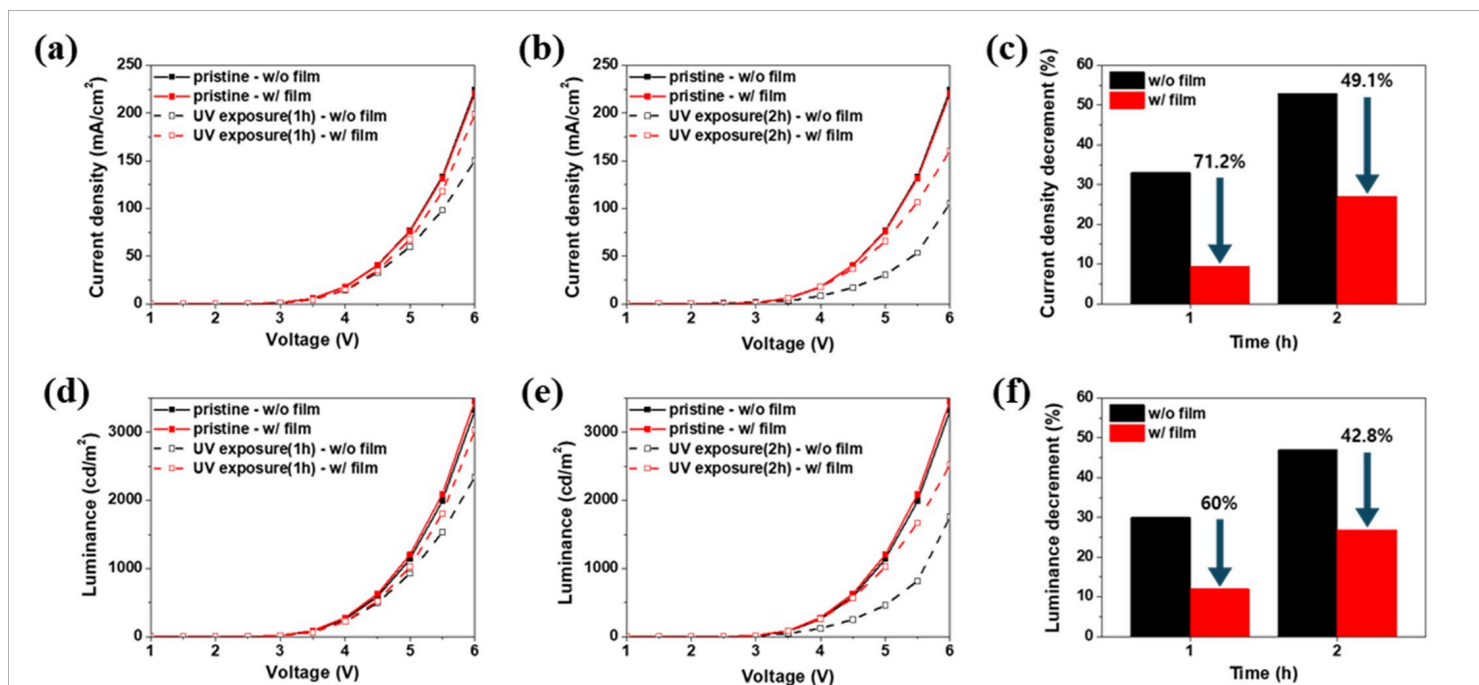


Figure 6

UV-induced degradation behavior of OLED devices with and without CATA MLA films: (a,b) J – V characteristics after UV irradiation for 1 and 2 h, respectively; (c) percentage decrease in J ; (d,e) L – V characteristics under the same conditions as in (a,b); (f) percentage decrease in L