

Supporting Information

Outcoupling-Enhanced Flexible Organic Light-Emitting Diodes on Ameliorated Plastic Substrate with Built-In Indium-Tin-Oxide Free Transparent Electrode

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Fabrication process for ameliorated plastic with embedded Ag network (PET-AN/OC)

The fabrication process of PET-AN/OC is schematically depicted in **Figure S1**. Firstly, the hexagon mask pattern with embedded groove structure was formed on the photoresist (RZJ-390PG, Ruihong Electronic Chemical Co., LTD) on a glass substrate by photolithography with the equipment (iGrapher200, SVG DigitOptics, Co., Ltd), which was developed in NaOH (0.4%) solution for 4 s and dried in the air subsequently. Then, the nickel (Ni) stamp with 150 μm diagonal length, 3 μm line width and 3 μm depth was fabricated from the patterned photoresist mold by electroforming.

For the preparation of PET-AN/OC, a film of UV-curable resin (D10, PhiChem) with a thickness of 10 μm was drop-dispensed on polyethylene terephthalate (PET) substrate. The Ni stamp was then put on the surface of UV curable resin, and spontaneously imprinted at a pressure of 3.0 bar for 30 s under UV light-emitting with an intensity of 1000 mW cm^{-2} at a distance of 1 cm via in-house built roll-to-plane UV nano-imprinting lithography system (SVG DigitOptics, Co., Ltd). Thereafter, the Ni stamp was mechanically peeled off from the patterned UV film, and the surface plasma treatment method was adopted to improve the hydrophilicity of the UV curable resin. Conductive nanostructured Ag ink (particle diameter between 2-10 nm, concentration of 41%, viscosity of 55 cps and surface tension of 30 dyne cm^{-1}) was sequentially drop-dispensed on the patterned UV film, and the hexagonal groove structure was filled by means of scraping using steel blade. The substrate was then sintered at 100 $^{\circ}\text{C}$ for 6 min in the infrared-assisted oven.

The moth-eye nanostructured optical coupling layer on the PET surface was fabricated by UV-assisted nanoimprint lithography with the polydimethylsiloxane

(PDMS) mold under pressure. The fabrication process of the PDMS mold has been described in details in the previous report, which was based on the reactive ion etching of Si substrate with the template of Ag nanoparticles.¹ The PDMS mold was put in conformal contact with the UV resin drop-casted on the PET surface. The imprinting was performed under a constant pressure of 0.8 bar with a UV illumination at a wavelength of 395 nm at 500 mJ cm⁻² for 10 s. Finally, the PET-AN/OC TCE was formed by peeling off the PDMS mold, leading to the moth-eye nanostructures constructed on PET substrates with a period of 200 nm, a groove depth of 300 nm, and the fill factor of 0.6.

Reference:

1. Zhou, L.; Ou, Q. D.; Chen, J. D.; Shen, S.; Tang, J. X.; Li, Y. Q.; Lee, S. T. Light Manipulation for Organic Optoelectronics with Bio-Inspired Moth's Eye Nanostructures. *Sci. Rep.* **2014**, *4*, 4040.

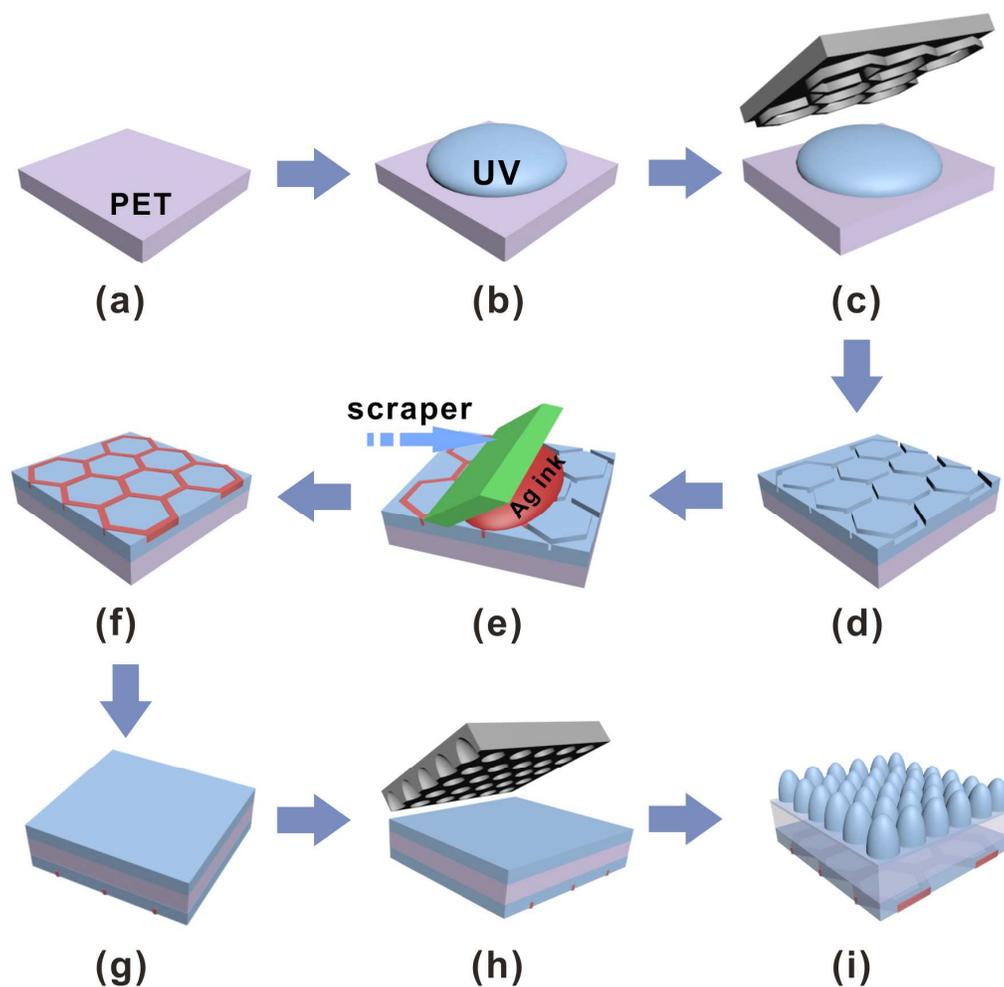


Figure S1. Schematic of the fabrication process for ameliorated plastic with embedded Ag network (PET-AN/OC). (a) The use of polyethylene terephthalate (PET) as the substrate. (b) Dispersion of the UV resin on PET substrate. (c) UV-assisted imprinting by the electroformed Ni mold with a hexagonal pattern. (d) Pattern transfer followed by plasma treating. (e) Filling the Ag ink by the scratching technology. (f) Sintering and cleaning of the embedded Ag networks. (g) Dispersing the UV resin on the other side of the PET substrate. (h) UV-assisted imprinting of the UV resin with the moth-eye nanostructured polydimethylsiloxane (PDMS) mold. (i) Formation of the final PET-AN/OC flexible substrate.

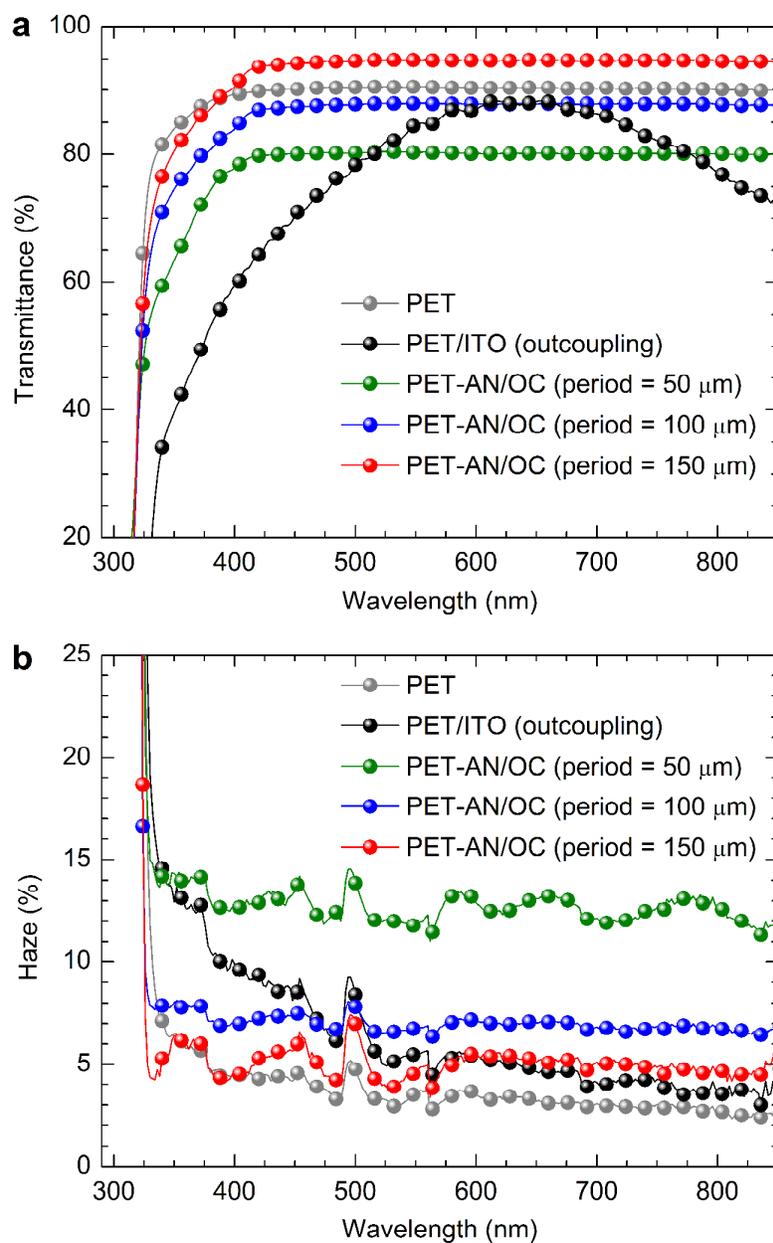


Figure S2. Comparison of optical properties of various PET-AN/OCs with the bare PET and ITO-coated PET (ITO/PET) with a moth-eye antireflection coupling layer. (a) Transmission spectra and (b) haze values of PET-AN/OCs with various hexagonal periods as a function of wavelength. The haze values were obtained by collecting the total transmittance and the specular transmittance with in integrating sphere [haze = (total transmittance - specular transmittance)/total transmittance].

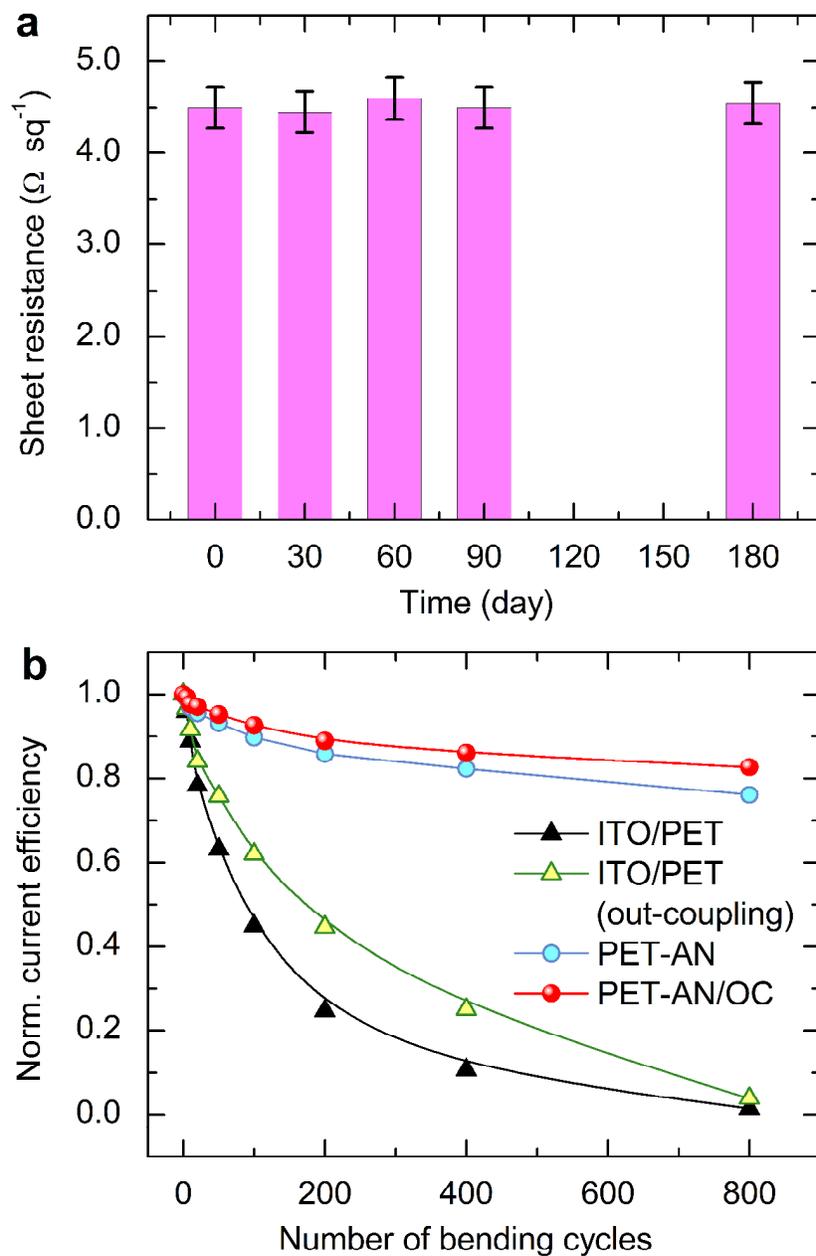


Figure S3. Comparison of the storage and mechanical stabilities of PET-AN/OCs. (a) The storage stability of PET-AN/OCs exposed to the air without any protection. (b) Normalized current efficiencies of green FOLEDs using various substrates are compared during the repeated bending, where the tests were conducted by repeatedly bending the substrates to a radius of curvature of about 6 mm at a constant current density of 15 mA cm^{-2} .

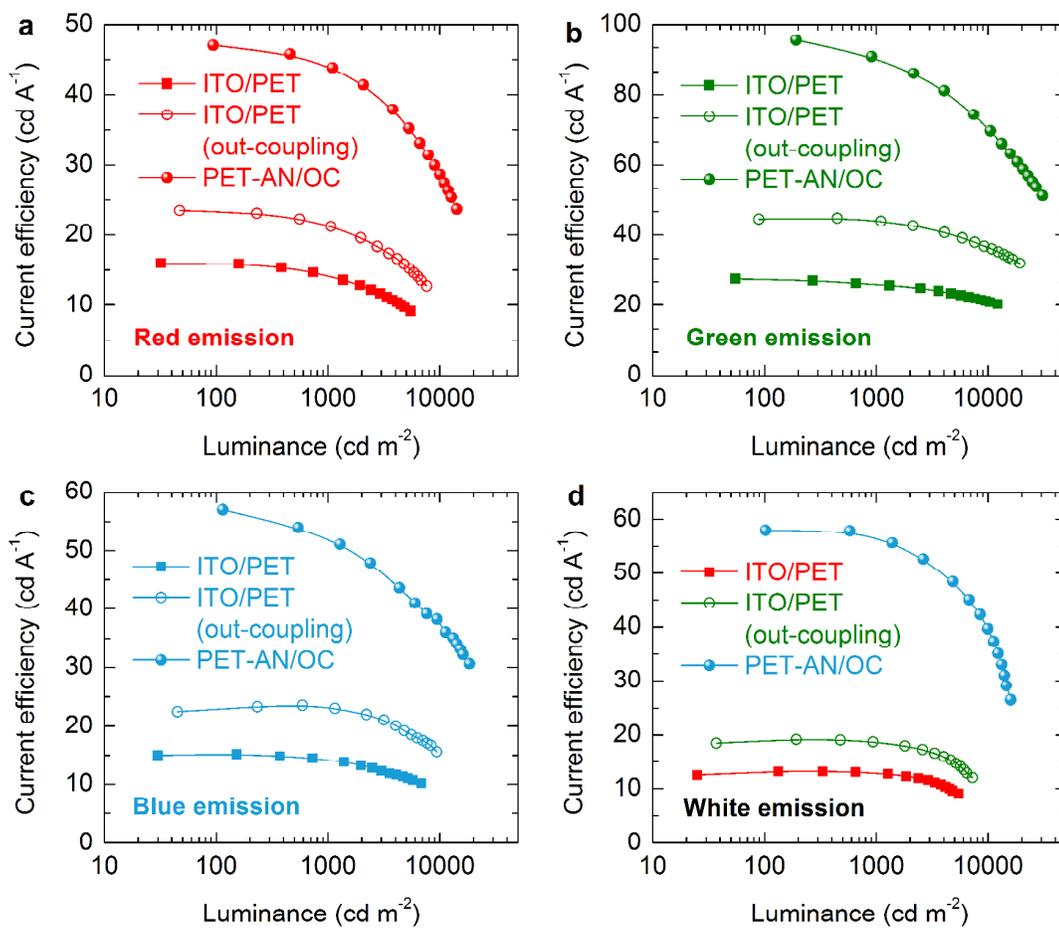


Figure S4. Current efficiencies as a function of luminance for FOLEDs using various plastic substrates with (a) red, (b) green, (c) blue, and (d) white emissions.