## Highly-Efficient Flexible Organic Light-Emitting Diodes On ITO-Free Plastic

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Flexible organic light-emitting diodes (OLEDs) are emerging as a leading technology for a variety of wearable intelligent electronics due to their unique capacity to be integrated with soft materials and curvilinear surfaces. Applications include bendable smartphones, foldable touch screens and antennas, paper-like displays, and curved and flexible solid-state lighting devices. To date, the most commonly used transparent conductive electrode (TCE), indium-tin-oxide (ITO), limits the further development of high-performance flexible OLED technology due to its inherent shortcomings such as brittleness, material scarcity, and a low throughput deposition process. It would be highly desirable to replace ITO with a new TCE solution which has low cost and improved bendability for future flexible, rollable or foldable OLED-based applications. Various types of flexible TCEs with attractive electrical and optical properties have been explored as alternative transparent conductors. Particularly, metallic grids, especially printed Ag grids, have provided another route to improving the properties of TCEs. However, further improvements on electrical and optical properties, chemical stability as well as the manufacturing process are required before they can be mass-produced as the promising next-generation TCEs. On the other hand, it is known that light outcoupling efficiency is limited in conventional OLED structures without a light extraction scheme because 70-80% of the emitted light is confined in the substrate and waveguide modes, or lost by the surface plasmon polariton of the metal electrode (SPP mode). To further enhance luminance and efficiency, a light outcoupling structure for liberating the trapped photons must be integrated into the flexible devices.

Here we demonstrate a novel flexible TCE using plastic with embedded Ag networks (PEANs), whose fabrication process is time-saving and cost-effective with the combination of Ag paste scratch technology, nanoimprinting lithography and precise pattern photolithography. The PEAN as a flexible TCE exhibits excellent optical, electrical and mechanical properties, allowing little degradation of electrical properties upon bending. The period of the Ag networks are several orders of magnitude larger than the visible wavelength, which minimizes light diffraction. The PEANs with a period of 150  $\mu$ m possess a transmittance >87% in the wavelength range of 400-800 nm and a sheet resistance < 5  $\Omega/\Box$ , which is superior to the performance of reported TCEs in the scientific literature, such as Ag nanowires, carbon nanotubes, and ITO. In addition to the total transmittance properties, a PEAN with a 150  $\mu$ m period exhibits an average haze value of ~4.3% over the visible spectrum, which is comparable to that of ITO-coated PET (ITO-PET, haze  $\approx 4.9\%$ ) and bare PET substrates (haze  $\approx 4.3\%$ ).

By employing an optimized light outcoupling structure with reduced ohmic losses, flexible green OLEDs yield a power efficiency (PE) > 120 lm/W and current efficiency (CE) >140 cd/A at a brightness of 1000 cd/m<sup>2</sup>, while the flexible white OLEDs exhibit a maximum external quantum efficiency (EQE) of 49% and a record PE of 106 lm/W at 1000 cd/m<sup>2</sup> with angular color stability. The results demonstrated here are much higher than previous reports of flexible white OLEDs in the scientific literature and even competitive to white OLEDs on glass substrates. The flexibilities of OLEDs using PEAN and ITO-PET were tested by repeatedly bending the substrates to a radius of curvature of about 6 mm at a constant current density of 15 mA/cm<sup>2</sup>. The flexible OLEDs using PEAN show stable operation under repeated bends with only a small decrease in efficiency, less than ~19% after 1,000 bending cycles. The device on ITO-PET on the other hand exhibits a quick degradation to failure upon the same repeated bending due to cracking in the brittle ITO and the decrease in conductivity, which demonstrates the superior bending stability of PEAN compared to ITO.

The flexible device presented in this work has the potential to achieve an even higher efficiency if all the device parameters and optical structures are continuously optimized to reduce the energetic and outcoupling losses during electron-photon conversion. The approach demonstrated here opens up the opportunity to other large-scale roll-to-roll fabrication technologies for wearable optoelectronics with high performance, low manufacturing and materials cost.

## Acknowledgment

The authors acknowledge financial support from the National Basic Research Program of China (Grant No. 2014CB932600), the National Natural Science Foundation of China (Grant Nos 91433116, 11474214), Jiangsu Science and Technology Department (No. BK20140053), Collaborative Innovation Center of Suzhou Nano



Science and Technology, Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, and the project of the Priority Academic Program Development (PAPD) of Jiangsu Higher Education Institutions.

Fig. 1. Device structure and performance of flexible white OLEDs. (a) Schematic of flexible white OLED device structure using PEAN as anode. (b) Photograph of a large-area flexible white OLED (50 mm × 50 mm) using PEAN as anode. Inset: the magnified image taken with an optical microscope. (c) Electroluminescence spectra of flexible white OLEDs using PEAN (without and with an outcoupling structure) or ITO-PET as anode. (d) Current density and luminance as a function of driving voltage of flexible OLEDs using various anodes. (e) Power efficiencies as a function of luminance. (f) Angular dependence of light intensity for flexible devices. The dashed line represents the ideal Lambertian emission pattern.

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