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A Printed Metal-oxide-free Cathode for Mechanically-durable and Ultra-flexible Organic Photovoltaics

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Abstract
A printed metal-oxide-free cathode was developed for ultra-flexible organic photovoltaics (OPVs). The champion devices achieved a high power conversion efficiency (PCE) of 9.7% and good cyclic mechanical durability (74% PCE retention after 500 cycles of compression/stretch process). Such excellent properties were realized by robust materials and structure.

1. Introduction

Fabrication of OPVs on ultrathin polymer substrates (1 µm in thickness) will enable great flexibility and ultra-lightweight characteristics[1]. Achieving good mechanical durability is extremely important for such devices to be utilized as flexible power sources in a self-powered system[2]. As a typical photonic device, OPVs need the transparent electrode to permit light enter the device, and the conventional choice is ITO electrode, which is brittle in nature[3]. Besides, ZnO is widely used in OPVs as the electron transport layer (ETL), which is also metal oxide and thereby brittle in nature[4].

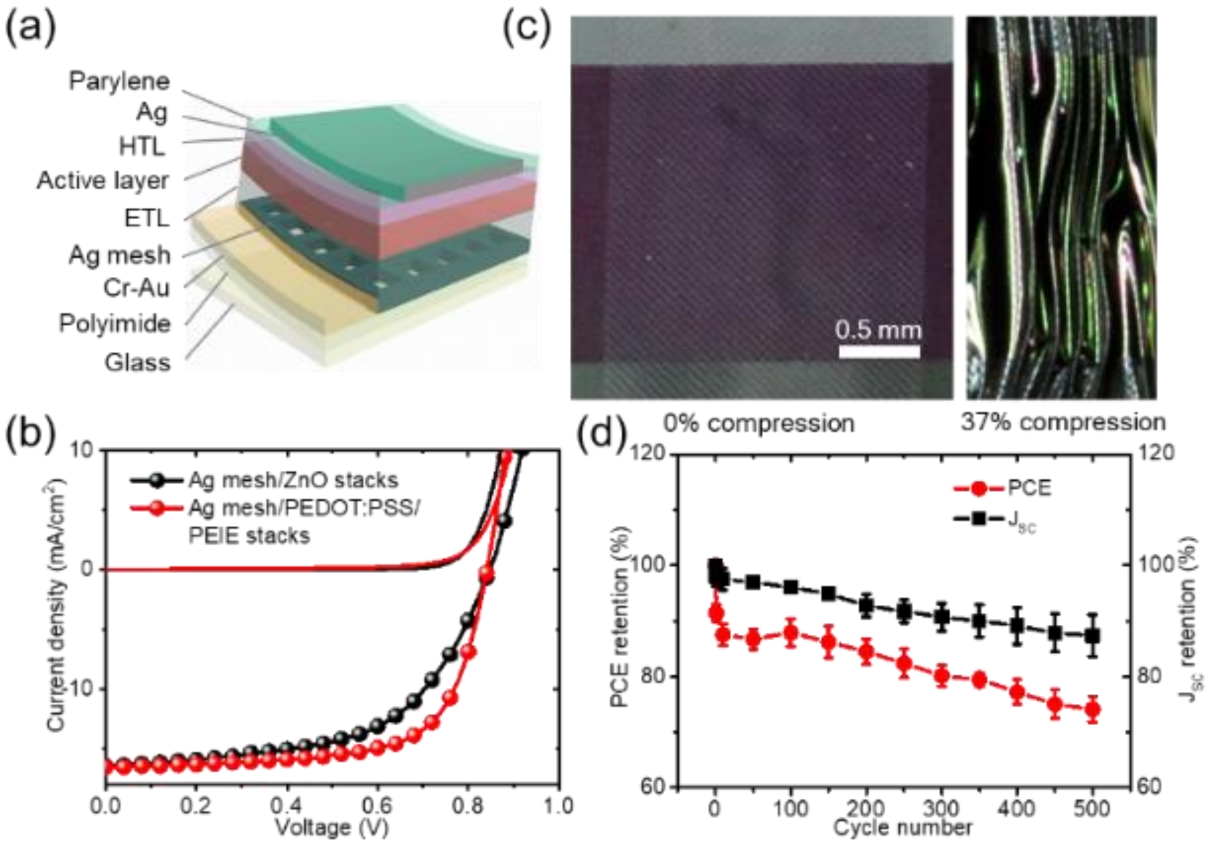

Figure 1. Mechanically durable OPVs based on the metal-oxide-free cathode. (a) A schematic illustration of the structure. (b) A comparison of Current density-voltage curves based on Ag mesh/ZnO and Ag mesh/PEDOT:PSS/PEIE stacks. (c) Optical images of OPVs with and without compression. (d) Cyclic durability with a 37% compression.

Due to the brittle nature of above metal oxides in flexible OPVs' structure, they can generate cracks under deformation, such as bending, twisting, and stretching. These cracks not only decrease the conductivity of the metal oxides, but also they can act as defects to trap light-generated carriers, which would lead to dramatic degradation of OPVs in the energy-harvesting capability. Therefore, the development of a complete stack structure based on robust materials is needed to simultaneously achieve good mechanical durability and high power conversion efficiency (PCE).

2. Results and discussion

Here, we report the development of ultra-flexible OPVs based on a printable metal-oxide-free cathode[5]. The metal-oxide-free cathode in this study is a stacked structure of printed Ag mesh, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), and polyethylenimine ethoxylated (PEIE). Ag mesh (100 nm in thickness) is fabricated on 1-µm-thick transparent polyimide substrates by the reverse-offset printing process[6]. PEDOT:PSS is used to improve the conductivity of Ag mesh in the whole plane direction with a decreased roughness, and PEIE is used to modify the work function. The active layer is a blended film consisting of a thiazolothiazole–naphthobisthiadiazole polymer (PTzNTz) and [6,6]-phenyl C_{71} butyric acid methyl ester ($PC_{71}BM$) and the anode consists MoO_X (7.5 nm) and Ag (100 nm) (**Figure 1a**). The champion device based on the metal-oxide-free structure delivered a maximum PCE of 9.7% (**Figure 1b**), outperforming previously reported flexible OPVs based on ITO-free transparent electrodes[7,8]. More importantly, based on the metal-oxide-free cathode, ultra-flexible OPVs provide excellent mechanical stability, i.e. 74% PCE retention after 500 cycles of stretch/release deformations at 37% compression (**Figure 1c** and **1d**). In contrast, OPVs based on cathodes consisting of ZnO almost totally lose PCE. The metal-oxide-free cathode concept works well in three different active layer systems, indicating that this approach offers a universal technique to achieve good mechanical properties.

3. Conclusion

We designed and tested ultra-flexible OPVs with high PCE and mechanical durability. In addition, we propose an Ag mesh/PEDOT:PSS/PEIE cathode to simultaneously obtain excellent electron collection capability and mechanical robustness. The insight gained from the ultra-flexible metal-oxide-free structure and their application in ultrathin organic photovoltaics will contribute to other mechanically-durable ultrathin photonic devices, and even ultra-flexible systems.

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