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Brush painting of transparent PEDOT/Ag nanowire/PEDOT multilayer electrodes for flexible organic solar cells

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ABSTRACT

We fabricated highly transparent and flexible poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)/Ag nanowire (NW)/PEDOT:PSS (PAP) multilayer electrodes by continuous brush-painting for used as flexible anode in highly flexible organic solar cells (FOSCs). By combining the flexibility of PEDOT:PSS and low resistivity of Ag NWs, we fabricated highly flexible PAP multilayer electrodes with a low sheet resistance of 13.96 Ω/square and a high diffusive transmittance of 80.48%. Effective embedment of the Ag NW network into the conductive PEDOT:PSS layer led to metallic conductivity and a high diffusive transmittance, which are desirable in transparent anodes for FOSCs. In addition, the PAP electrode showed an invariable resistance ($\Delta R/R_0$) during outer and inner bending testing, due to the high strain failure of both PEDOT:PSS and Ag NW network. The identical current density–voltage behavior of the FOSCs with the brush-painted PAP electrode as the FOSC with a conventional ITO electrode demonstrates that the brush-painted PAP multilayer is a promising alternative to high-cost ITO electrodes to produce cost-efficient FOSCs. Furthermore, we suggest that high-performance, transparent, and flexible electrodes can be fabricated by low-cost brush-painting for paintable FOSCs.

1. Introduction

As cost-efficient and simple energy harvesting devices, flexible organic solar cells (FOSCs) are drawing great attention due to their simple device structure and uncomplicated and fast printing-based fabrication process [1–3]. In addition, the robustness and superior flexibility of FOSCs fabricated on flexible substrates should lead to new application fields such as fabric-attachable energy sources and rollable/stretchable energy sources [1–8]. Furthermore, recent progress of roll-to-roll based printing technologies has made it possible to fabricate FOSCs on roll-type flexible substrates at a low cost [2]. In many important FOSC components, such as substrates, cathode/anode electrodes, organic active layers, and encapsulation films, the flexible transparent electrode is a key component because the light transmittance and carrier extraction, which are directly related to the power conversion efficiency, are mainly affected by the optical and electrical properties of the transparent electrodes. In addition, except for the inorganic anode materials, such as indium tin oxide (ITO) films, all components including the metal cathode and organic layers in FOSCs are very flexible. Therefore, the flexibility of FOSCs is mainly affected by the mechanical properties of transparent electrodes [6,7,9,10]. Until now, amorphous or crystalline ITO films grown by vacuum-based DC or RF sputtering processes have been employed as a main transparent anode or transparent cathode in the fabrication of OSCs or inverted OSCs, even though they are very brittle in terms of substrate bending or rolling [11,12]. In addition, the high costs of indium element and high payback cost of vacuum-based ITO sputtering processes are important drawbacks of using conventional ITO films in cost-efficient FOSCs [13–16]. To address the high costs of indium-based ITO electrodes, extensive research has been devoted to cost-efficient and printable transparent electrodes, such as PEDOT:PSS, carbon nanotubes, graphene, and Ag nanomesh and nanowires (NWs) networks [17–22]. In particular, printable PEDOT:PSS electrodes and a percolation network of Ag NWs have been suggested as high performance flexible and transparent electrodes because both electrodes have a superior flexibility, which is acceptable for FOSCs and can be fabricated by a simple printing process. Recently, as a promising substitute for ITO electrodes, our group reported a simple brush-painted Ag NW network electrode with a sheet resistance of 13.55 Ω/square and a transmittance of 86.27% for FOSCs [23]. However, the Ag NW network film still has critical problems such as its poor adhesion and irregular morphology. Poor adhesion of the Ag NW electrode may cause reliability problems and shortened lifetimes of the FOSCs. In addition, the irregular morphology of the Ag NW can act as a leakage path and decrease the shunt resistance of FOSCs [24–29]. Another promising flexible transparent electrode for FOSCs is a conductive PEDOT:PSS electrode which can be prepared by

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various printing processes [2,5,6]. Although conductive PEDOT:PSS is a printable transparent anode for FOSCs, a further decrease of the sheet resistance is required to meet the demands of an anode in FOSCs [30,31]. Therefore, the development of Ag NW and PEDOT:PSS hybrid transparent electrode to solve the critical problems of individual Ag NW and PEDOT:PSS electrodes is urgently demanded. Recently, Gaynor et al. reported that drop-casted Ag NWs laminated on PEDOT:PSS films had a sheet resistance of 17 Ω/square and a transmittance of 83% [25]. However, the complicated transfer process of drop-casted Ag NWs onto a PEDOT:PSS film still remains as a critical problem of Ag NW and PEDOT:PSS hybrid electrodes. Although Ag NWs and PEDOT:PSS films have been individually investigated as effective ITO replacements for FOSCs, there have been no reports of a PEDOT:PSS/Ag NW/PEDOT:PSS (PAP) multilayer as a hybrid transparent electrode for cost-efficient FOSCs. In particular, PAP multilayer electrodes fabricated by simple and fast brush painting have not been reported to date.

In this work, we combined Ag NWs with PEDOT:PSS films prepared by brush painting as a hybrid multilayer electrode for cost-efficient FOSCs. By alternative brushing of PEDOT:PSS and Ag NWs, we fabricated highly transparent and flexible PAP multilayer electrodes with a low sheet resistance of 13.96 Ω/square, high transmittance of 83.17%, and superior flexibility. The electrical, optical, and surface properties of the brush-painted PAP hybrid electrodes were investigated as a function of the number of repeated brush painting cycles of the Ag NW network, which was embedded into the PEDOT:PSS layer. In addition, the performances of FOSCs fabricated on PAP hybrid electrodes were compared as a function of the number of Ag NW brush painting cycles. The similar current density–voltage curve of the FOSC with a PAP electrode to the FOSC with an ITO electrode demonstrated that transparent PAP multilayers fabricated by brush painting can be a promising transparent electrode in cost-efficient FOSCs as a substitute for vacuum process-based high-cost ITO anode.

2. Experimental

The Ag NW and PEDOT:PSS hybrid multilayer films were fabricated by simple brush painting at room temperature under atmospheric ambient. The brush painting technique was recently suggested as a simple coating method for cost-efficient FOSCs to enhance the ordering of the polymer induced by the shear stress of the brush. Recently, we also suggested that Ag NW network film could be coated by a simple brush painting process [23]. This simple brush painting process was employed to prepare the PAP multilayer on a flexible PET substrate. Fig. 1(a) shows a schematic of the multi-step brush painting process used to fabricate a

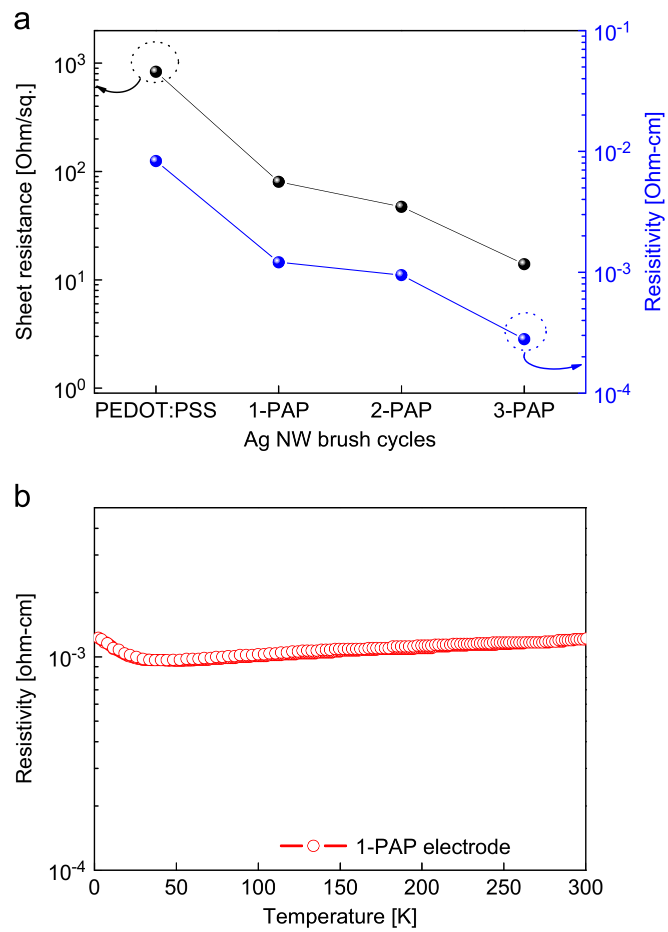

Fig. 2. (a) Sheet resistance and resistivity of brush-painted 1-, 2-, and 3-PAP electrodes as well as a single PEDOT:PSS layer. (b) Temperature dependence of the 3-PAP multilayer from 5 to 300 K.

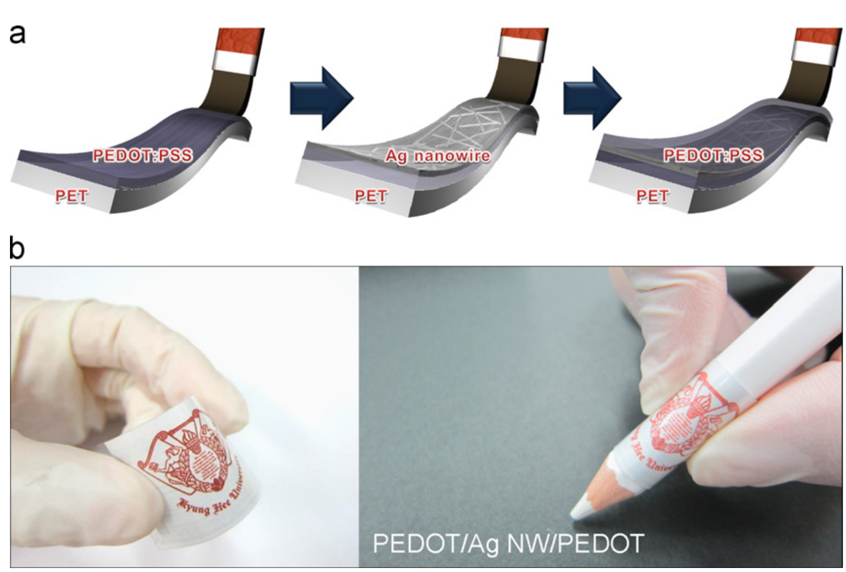

Fig. 1. (a) Fabrication process of the PEDOT:PSS/Ag NW/PEDOT:PSS multilayer using simple brush painting. (b) Picture of a flexible PAP electrode painted on a flexible PET substrate. The brush-painted PAP electrode possesses a high transparency and flexibility.