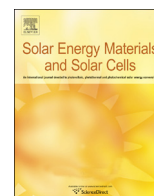




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## Electrochemically deposited nano polyaniline films as hole transporting layers in organic solar cells

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## ABSTRACT

In this study, we prepared organic solar cells based on poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM) with electrochemically deposited nano polyaniline (PANI) buffer layers (either PANI – 0.1 M or PANI – 0.3 M) as hole transporting layers and compared their performances with those of solar cells lacking the buffer layer (i.e., bare ITO) or featuring buffer layers of poly(3,4-ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS) or PEDOT:PSS-covered PANI. The power conversion efficiency of the device featuring PANI/PEDOT:PSS as the buffer layer (2.76%) was greater than those of devices featuring bare ITO (0.75%) or the PANI – 0.1 M (1.33%), PANI – 0.3 M (1.78%), or PEDOT:PSS (2.30%) layers. We suspect that the increased conductivity of the PANI/PEDOT:PSS composite, caused by interactions between the PANI nitrogen atoms and the functional groups of PSS, led to additional doping of PANI. This primary doping effect by PSS toward PANI lowered the series resistance of the PANI/PEDOT:PSS buffer layer and, thereby, increased the photocurrent of the device. As a result, electrochemically deposited PANI buffer layers, with or without PEDOT:PSS, appear to be promising hole transporting layers for organic electronic devices that require different candidate materials (other than PEDOT:PSS conductors) and/or different processing conditions (other than spin-coating).

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## 1. Introduction

Although poly(3,4-ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS) is insoluble in organic solvents, it is often employed as a transparent hole-transporting material in organic cells because of its high conductivity, simple processing, and suitable work function [1]. The use of PEDOT:PSS has several limitations: it degrades under UV illumination, it introduces water into the devices' active layer, and it retains a degree of acidity that can damage indium tin oxide (ITO) at high temperatures, thereby affecting the long-term stability of its devices [2]. Therefore, several highly conductive materials, including carbon nanotubes (CNTs) [3], polyhexylthiophene [4], and polyaniline (PANI) [5–7], have been studied as possible replacements for PEDOT:PSS as the hole injection or transporting layer.

Doping PEDOT with PSS allows it to become soluble in aqueous media, thereby facilitating its deposition onto ITO surfaces through spin-coating—the most common method employed in the fabrication of organic devices. Even though spin-coating is an inexpensive, rapid,

and simple means of depositing liquid materials, it can waste a lot of material, making it less attractive for manufacturing processes.

Several methods for the deposition of hole transporting (e.g., PEDOT:PSS, polyaniline (PANI)) and active layers—including inkjet printing [8], spray deposition [9], roll-to-roll gravure printing [10], brush painting [11], and screen printing [12]—have been developed to enhance the manufacturing process with the goal of mass production of organic electronics. The presence of PANI-coated ITO electrodes can decrease the operating voltage, increase the electroluminescence quantum efficiency, and improved device reliability of organic light emitting diodes (OLED) because they can lower the hole-injection barrier between the anode and the hole-transporting emissive layer [13–21]. Notably, the performance of ITO/PANI-based devices can be similar to that of corresponding ITO/PEDOT:PSS-based devices [20–22].

Recently, electrochemical polymerization was reported as an alternative means of depositing conductive polymers onto ITO surfaces for organic device applications [23–25]. Zhou et al. [23] electrochemically deposited PEDOT:PSS onto an ITO surface during the formation of a device having the configuration ITO/PEDOT:PSS/ZnO:MDMO-PPV/Al and obtained a power conversion efficiency ( $\eta$ ) of 0.33% under AM 1.5 illumination. Mello et al. [24] used

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electrochemical polymerization to deposit sulfonated PANI (SPANI) and then prepared a device having the structure TO/SPANI/poly(3-methylthiophene)/Al that exhibited a value of  $\eta$  of 0.8% under monochromatic irradiation ( $\lambda=580$  nm;  $0.8$  W/m<sup>2</sup>). Qu et al. [25] electrodeposited PANI as the anode buffer layer (thickness: 120 nm) in the device structure ITO/PANI/MDMO-PPV:PCBM/ZnO/Al and obtained a power conversion efficiency of 0.65%.

Although electrochemically deposited PANI films are promising materials for use in organic optoelectronics, the effects of the morphologies of these films on the photovoltaic performance of poly(3-hexylthiophene) (P3HT)/[6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM)-based organic solar cells (OSCs) have seldom been reported. It is well established [25–26] that the surface morphology of a film of an organic material can help or hinder the transport of carriers across various interfaces. In a previous report [26], we demonstrated that a tube-like PANI nanomaterial increased the photovoltaic performance of P3HT:PCBM-based devices when it acted as a hole-transport layer (HTL) between PEDOT:PSS and P3HT:PCBM layers; in other words, the morphology of the hole-transporting material played a key role affecting the photovoltaic properties of that device.

In this study, we prepared OSCs featuring HTLs with various surface morphologies—bare ITO, ITO presenting an electrochemically deposited PANI thin film, and a PANI – 0.3 M thin film spin-coated with PEDOT:PSS—and compared their photovoltaic properties [open-circuit voltage ( $V_{oc}$ ); series resistance ( $R_s$ ); short-circuit current density ( $J_{sc}$ );  $\eta$ ] with respect to their different surface morphologies.

## 2. Experimental

Aniline (ANI) was doubly distilled prior to use and stored at 5 °C. Electrochemical experiments are performed in a three-electrode cell; patterned ITO glass, a platinum wire, and Ag/AgCl

functioned as the working electrode, counter electrode, and reference electrode, respectively. Cyclic voltammetry (CV) was performed using an AUTO-LAB apparatus. The PANI – 0.1 M and PANI – 0.3 M films were obtained after electro-polymerization of aqueous solutions containing 0.1 and 0.3 M ANI monomer, respectively, and 1 M H<sub>2</sub>SO<sub>4</sub>, applying a sequential linear potential scan rate of 0.01 V/s between –0.2 and +0.9 V versus the Ag/AgCl electrode. The PANI/PEDOT:PSS film was obtained by spin-coating a PEDOT:PSS (AI 4083) solution onto the ITO/PANI – 0.3 M surface and then drying at 150 °C for 20 min. Field emission scanning electron microscopy (FE-SEM) images of water- or hexane-diluted dispersions dried on cover glasses were recorded using a Hitachi FE-2000 apparatus. The surface morphologies of the films were characterized using a Ben-Yuan CSPM4000 scanning probe microscope and an atomic force microscope operated in the tapping mode. The film thickness was measured using a Dektak 6M stylus profilometer. Optical spectra were recorded using a UV–vis spectrophotometer; the work functions of the films were measured using a Riken Keiki AC-2 surface analyzer photoelectron spectrometer. The current density–voltage ( $J$ – $V$ ) characteristics were measured using a Keithley 2400 source meter while illuminating the devices with white light (100 mW/cm<sup>2</sup>) from a halogen lamp. A single-crystalline silicon solar cell was used as a reference cell to confirm the stability of the light source; the mismatch factor was not taken into account.

## 3. Results and discussion

### 3.1. Surface morphologies of devices

Smooth surface morphologies are required for anodes used in organic optoelectronic devices because “spikes” can cause breakdown and/or shorting, thereby affecting the performance [27].

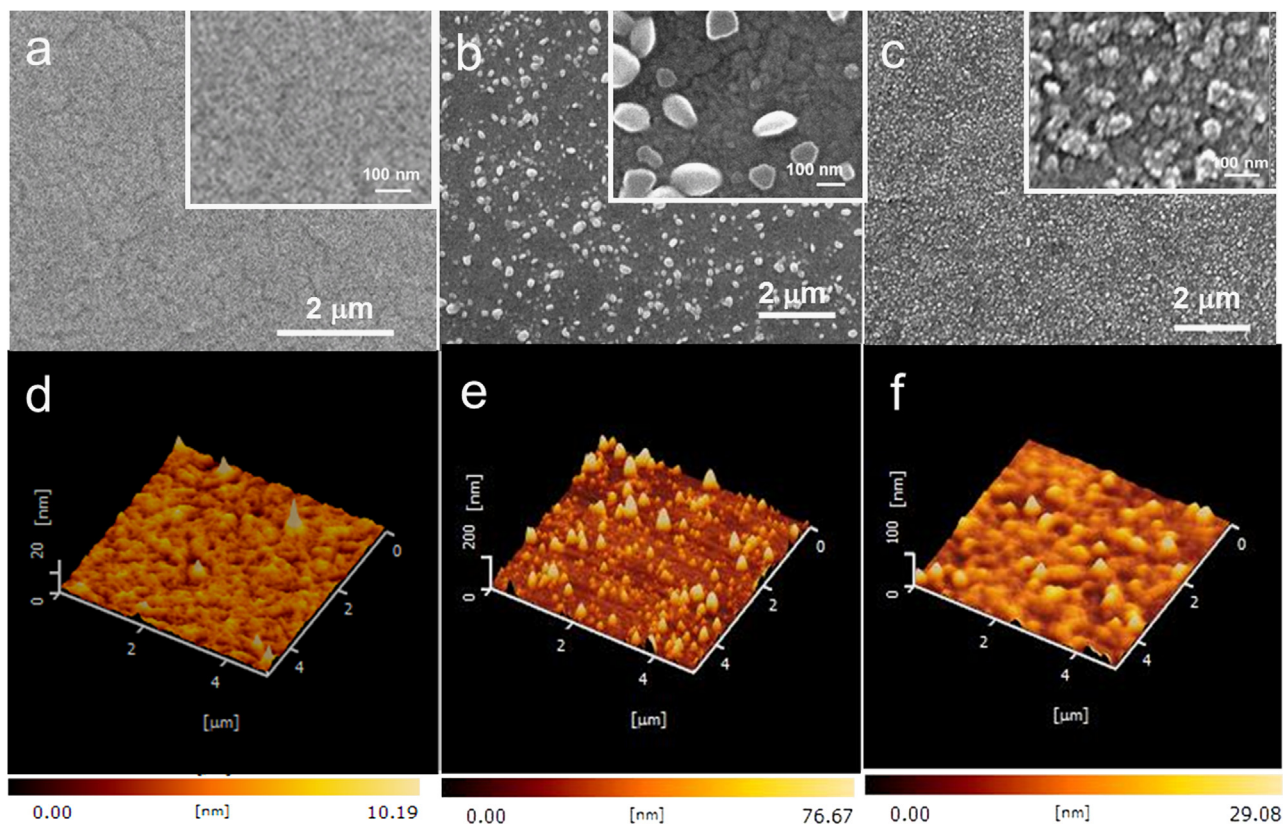


Fig. 1. (a)–(c) SEM and (d)–(f) AFM images of (a, d) ITO, (b, e) PANI – 0.1 M, and (c, f) PANI – 0.3 M films.

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