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Efficient recyclable organic solar cells on cellulose nanocrystal substrates with a conducting polymer top electrode deposited by film-transfer lamination

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ABSTRACT

We report on efficient solar cells on recyclable cellulose nanocrystal (CNC) substrates with a new device structure wherein polyethylenimine-modified Ag is used as the bottom electron-collecting electrode and high-conductivity poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS, PH1000) is used as the semitransparent top hole-collecting electrode. The PEDOT:PSS top electrode is deposited by a film-transfer lamination technique. This dry process avoids swelling damage to the CNC substrate, which is observed when PEDOT:PSS is directly spin-coated from an aqueous solution. Solar cells on recyclable CNC substrates exhibit a maximum power conversion efficiency of 4.0% with a large fill factor of 0.64 ± 0.02 when illuminated through the top semitransparent PEDOT:PSS electrode. The performance of solar cells on CNC substrates is comparable to that of reference solar cells on polyethersulfone substrates.

1. Introduction

Organic solar cells represent a cost-effective and an environmentally friendly technology for the generation of renewable energy [1–7]. Over the last decade, the power conversion efficiency (PCE) of organic solar cells has been significantly improved up to values of about 10% [8,9]. Due to the ease of fabrication, organic solar cells have been demonstrated on various kinds of substrates, such as glass, plastic, metal foil and paper substrates. From a life-cycle perspective, substrate materials that can be synthesized from renewable feedstocks at a low-cost are particularly attractive for the realization of a sustainable solar cell technology [10–12]. Paper is considered a promising substrate for organic solar cells, because it is inexpensive, low-weight, flexible and recyclable [13–16]. However, the device performance of solar cells fabricated on paper has been low because of the high surface roughness and porosity of the substrates.

Recently, we demonstrated that polymer solar cells fabricated on cellulose nanocrystal (CNC) substrates, with the structure: CNC/Ag (20 nm)/polyethylenimine ethoxylated (PEIE)/active layer/MoO_3/Ag, are easily recyclable [10]. The active layer in these solar cells was comprised of blends of poly[(4,8-bis-(2-ethylhexyloxy)-benzo[1,2-b:4,5-b′]dithiophene)-2,6-diyl-alt-(4-(2-ethylhexanoyl)-thieno[3,4-b]thiophene)-2,6-diyl]: phenyl-C_{61}-butyric acid methyl ester (PBDTTT-C:PC_{60}BM). Solar cells on CNC substrates yielded a PCE of 2.7%; an unprecedented level of performance for a polymer solar cell fabricated on recyclable substrates derived from renewable feedstocks [10]. However, the efficiency of solar cells with a similar structure, but fabricated on glass/indium–tin oxide (ITO) substrates

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have yielded PCE values of around 6% [1,17]. The lower PCE values displayed by solar cells on CNC substrates were attributed to the low transmittance of the semitransparent Ag (20 nm) bottom electrode. We suggested that the PCE of solar cells fabricated on CNC substrates could reach values comparable to those obtained with devices fabricated on plastic substrates if electrodes with higher transmittance were employed [10].

The conducting polymer poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) has been widely used as a transparent electrode for organic solar cells and organic light-emitting diodes because of its high transmittance values across the visible spectrum and high-conductivity (over 1000 S/cm) [18–25]. However, we have found that direct coating of PEDOT:PSS, which is processed from an aqueous solution, damages the CNC substrates. CNC films can readily be redispersed at room temperature in water, thus leading to easily recyclable solar cell devices [10]. The damage of cellulose substrates by the aqueous processing of PEDOT:PSS leads to poor performance of solar cells [11]. Thus, it is important to find a dry deposition method for PEDOT:PSS electrodes on cellulose substrates to produce efficient recyclable solar cells. Recently, Wang et al. [26] and Gupta et al. [27] reported a deposition process in which a high-conductivity PEDOT:PSS film is first deposited onto a poly(dimethylsiloxane) (PDMS) stamp and then transferred by lamination onto the photoactive layer.

Herein, we report on the demonstration of efficient recyclable solar cells fabricated on CNC substrates using a film-transfer lamination technique to produce a semitransparent PEDOT:PSS top hole-collecting electrode, while using a reflective Ag/polyethylenimine (PEI) bottom electron-collecting electrode (Fig. 1). Polymer solar cells fabricated on CNC substrates with a poly(3-hexylthiophene) (P3HT):indene-C_{60} bisadduct (ICBA) photoactive layer display a high fill factor (*FF*) of 0.64 ± 0.02 and a high average PCE of 3.8 ± 0.2%; a level of performance that is nearly identical to that of solar cells fabricated on polyethersulfone (PES) substrates.

2. Experimental section

2.1. Solar cells fabricated on CNC and PES (Ref. device) substrates with PH1000-L top electrodes

CNC substrates were prepared as described before [10]. CNC and PES (*i*-components Co., Ltd.) substrates were adhered onto polydimethylsiloxane (PDMS)-coated glass. Then, an 80 nm thick Ag film was deposited on half of the area of the CNC substrates through a shadow mask using a thermal evaporation system (SPECTROS, Kurt J. Lesker). Polyethylenimine (PEI, branched, #408727, Sigma–Aldrich) was spin-coated on Ag at 5000 rpm for 1 min in a N_2-filled glove box from a 0.4 wt.% solution in 2-methoxyethanol (#284467, Sigma–Aldrich) and annealed at 100 °C for 10 min. An effective thickness of 10 nm (PEI) was derived through measurement and modeling with spectroscopic ellipsometry (J.A. Woollam Co., M-2000) [1,28]. P3HT (4002-E, Rieke Metals Inc.):ICBA (Luminescence Technology Corp.) (1:1, weight ratio, total 40 mg/ml) was spin-coated at 800 rpm for 30 s in the N_2-filled glove box from a chlorobenzene (#284513, Sigma–Aldrich) solution and annealed on a hot plate at 150 °C for 15 min. The film thickness was about 200 nm as measured by spectroscopic ellipsometry.

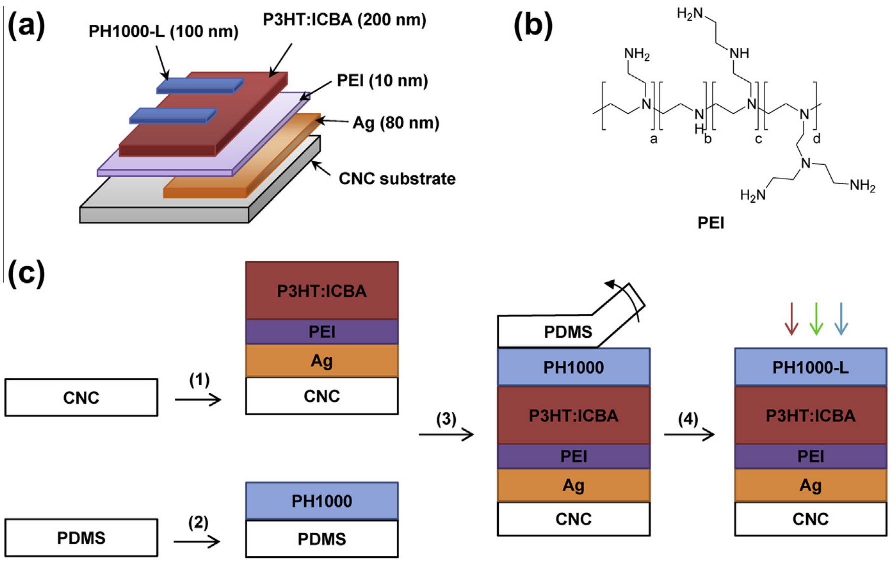

Fig. 1. (a) Device structure of solar cells on CNC substrates: CNC/Ag/PEI/P3HT:ICBA/PH1000–L where PH1000–L indicates the PEDOT:PSS PH1000 prepared by film-transfer lamination as the top electrode; (b) chemical structure of branched polyethylenimine used to lower the work function of Ag; (c) the fabrication procedure of recyclable solar cells on CNC substrates: (1) thermal deposition of Ag and spin coating of PEI and P3HT:ICBA on top of the CNC substrates; thermal annealing applied on PEI and P3HT:ICBA layers after each spin coating; (2) mild O_2-plasma treatment (5 s) on PDMS followed by spin coating of PH1000; (3) PDMS with PH1000 was transferred onto mild plasma treated (1 s) P3HT:ICBA surface facedown with PH1000 contacting the active layer; (4) Peeling-off the PDMS and thermal annealing to cure PH1000–L to finish the device fabrication. Light was illuminated through the top PH1000–L electrode during the photovoltaic performance measurement.