



Inverted P3HT:PCBM organic solar cells on low carbon steel substrates

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Abstract

Solar cell device fabrication on opaque metallic substrates is limited by the transmission of light from top transparent electrode. Moreover, conductivity of the substrates requires insulation for cell integration in a module. In this manuscript, we report fabrication of inverted ITO-free organic solar cells on opaque steel substrates without use of any lithography with power conversion efficiencies of ca. 1.35%. The device structure was Steel/Insulator/Al/ZnO/P3HT:PC₆₀BM/MoO₃/Au. The insulated substrates were first planarized using a thin poly(methylmethacrylate) or PMMA layer to minimize the substrate roughness and to enable the fabrication of subsequent layers with minimum roughness. We investigated the effect of thickness of various layers to maximize the performance of these devices with major emphasis on improving the transmission of top electrode to maximize the light absorption in the active layer blend. For best photovoltaic characteristics of the devices, thickness determination of thin transparent conducting layer of gold was carried out by conducting transmission, conductivity and morphological studies. Solution processed ZnO on Aluminum showed ridge like structure which provides larger interfacial area for the collection of electrons. Photoluminescence measurements on Active layer/MoO₃ emphasize the importance of insertion of sufficiently thin MoO₃ layer in the device in terms of efficient exciton dissociation.

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1. Introduction

Organic solar cells (OSCs) based on polymer-fullerene blend bulk heterojunctions have attracted considerable attention in recent years owing to their potential for flexible, lightweight, and low-cost applications of solar energy conversion (Hoppe and Sariciftci, 2004; Mayer et al., 2007 and Søndergaard et al., 2012). One of the most established organic polymers for organic solar cells is based on regioregular poly 3-hexylthiophene (P3HT) and [6,6]

phenyl C₆₁butyric acid methylester (PCBM), yielding devices with efficiency approaching 5% (Ma et al., 2005). Although significant progress have been made towards developing polymer-fullerene based solar cells on glass substrates with power conversion efficiencies approaching 8% (He et al., 2011b) for bulk heterojunction devices and 11–12% (You et al., 2013, <http://www.heliatek.com/>) for multi junction solar cells, P3HT:PCBM blend still remains a workhorse system for various studies.

Whilst Indium Tin Oxide (ITO) coated glass substrates are extensively used for the fabrication of solar cell devices, the potential of organic solar cells in flexible devices have led to exploration of flexible substrates such as ITO coated Polyethyleneterephthalate(PET)/Polyethylene Naphthalate

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(PEN). The devices fabricated on these substrates using P3HT:PCBM have yielded efficiencies approaching 4.2% (Wang et al., 2010) with high flexibility. Among other flexible substrates, low-carbon steel substrates have potential for incorporation of such devices with many advantages such as flexibility, low cost, high strength and compatibility with roll-to-roll processing for high volume production over other substrates. A key advantage of steel substrates over PET and PEN substrates is their excellent barrier properties towards oxygen and water vapor transmission rates. Low-carbon steel is used as rooftop material for housing, industrial and commercial building throughout the world and these roof surfaces can potentially be integrated with OSCs.

Although solar cell devices on steel substrates are well established in case of Silicon (Si) and copper indium gallium selenide (CIGS) systems (Schropp and Rath, 1999; Lee et al., 2013; Wuerz et al., 2009; Dhere et al., 2002) there are very few reports of organic solar cells fabricated on steel substrates (Galagan et al., 2011; Gupta et al., 2013a, 2014). However, opaqueness of steel poses a few challenges which include fabrication of an appropriate device architecture such that maximum amount of light enters the device from top. Among transparent electrodes, ITO is a promising candidate and has been used on top of active layer (ActL) (Chen et al., 2008; Bailey-Salzman et al., 2006; Schmidt et al., 2009; Ahlswede et al., 2007) in organic photovoltaic devices. However, since ITO cannot be evaporated easily and hence has to be sputtered, sputtering may damage the ActL underneath. Also, due to concerns on possible scarcity of Indium and incompatibility of ITO for flexible substrates because of its brittleness, use of several other transparent electrodes have been reported (Søndergaard et al., 2012), for example, poly(3,4-ethylene dioxothiophene):poly(styrenesulfonate) (PEDOT:PSS) with metal grids (Zou et al., 2010; Tvingstedt and OlleInganäs, 2007; He et al., 2011a; Galagan et al., 2011), thin metal films (Wilken et al., 2012) and oxide/metal/oxide structures (Zadsar et al., 2012; Shen et al., 2012), carbon nanotubes (CNT) (Wang et al., 2009a; Rowell et al., 2006), graphene (Wang et al., 2009b; Park et al., 2010), and metal nanowires (Lee et al., 2008; Yang et al., 2011) on glass and plastic substrates. However, many of these methods are unlikely to achieve desired characteristics when a transparent electrode has to be deposited on top of device, as is the case with opaque steel substrates. Hence the top electrode, whether as anode or cathode, must be transparent for sufficient amount of light to reach the ActL. Among a few earlier works on steel substrates, Galagan et al. used steel itself as electrode for solar cell fabrication with a stack of thin metal layers as transparent electrode but with very thick buffer and ActL in order to reduce the deleterious effect of steel roughness (Galagan et al., 2012). In another study, Gupta et al. used highly conductive PEDOT:PSS with metal grids as transparent electrode (Gupta et al., 2013a) using stamp transfer technique with thermally evaporated metal grids as transparent

electrode and obtained an efficiency of 3.25% with device area 0.09 cm^2 and 0.16 cm^2 , where they used about 190 nm thick ActL in the bulk heterojunction devices. The challenges in fabricating a device on steel substrates are two fold: to minimize the substrate roughness by using a planarizing layer so that smooth interfaces can be formed and to maximize the transmission of light from top with an electrode as transparent as possible. Also, it is important to fabricate a commercially attractive OPV device on steel substrate to make the device without many variations in the process and by using simple processing techniques. In this study, we demonstrate a simple process to fabricate an organic solar cell device on opaque insulated steel substrate with a thin PMMA layer to planarize the rough substrate and by incorporating a thin metal layer deposited by thermal evaporation, as transparent electrode with environmentally stable ZnO and MoO₃ buffer layers and a reflecting Aluminum (Al) back electrode.

2. Experimental details

We have used a polymer coated low-carbon steel substrate for device fabrication with device structure shown in Fig. 1 having 0.16 cm^2 area pixels. Low-carbon steel substrates coated with a proprietary polymer based on polyimide family were provided by Tata Steel. The coated steel substrates were cleaned with soap solution followed by ultrasonication with DI water, Acetone and Isopropyl alcohol for 5 min each respectively and drying with nitrogen. Roughness of these substrates was very high compared to typically used ITO electrodes and hence another dielectric Poly(methylmethacrylate) polymer (PMMA) is spin coated on top to achieve a smoother surface. A 100 nm thin PMMA film was spin coated on polyimide coated steel substrate using a 2.4 wt% PMMA solution dissolved in n-butylacetate. The film was annealed at 180 °C on hot plate for 30 min in normal environment. Subsequently, 99.999% pure Al (Sigma Aldrich) was thermally evaporated to grow a 100 nm thin Al film using a metal mask at a pressure of 5×10^{-6} mbar. 0.4 M ZnO solution was prepared by dissolving Zinc acetate dihydrate (C₄H₁₀O₆ Zn) in 2-Methoxyethanol (C₃H₈O₂) with tiny addition of ethanolamine (C₂H₇NO) as stabilizer. The solution was then spin coated on Al layer followed by annealing on hot plate at 150 °C for 30 min in air before transferring to a nitrogen filled glove box. P3HT:PCBM blend solution was prepared by mixing regioregular poly(3-hexylthiophene) (RR-P3HT – 30 mg/ml) and phenyl-C₆₁-butyric acid methyl ester (PCBM) (30 mg/ml), procured from Sigma Aldrich and Nano-C, respectively, in chlorobenzene. The solution was then spin coated on cleaned ZnO coated substrates at 1500 rpm for 1 min followed by annealing at 130 °C for 20 min in nitrogen ambient to get 100 nm film. MoO₃ (Sigma Aldrich, 99.98% purity) layer, acting as a hole transport layer was thermally evaporated at a pressure of 5×10^{-6} mbar on top of the ActL. Finally, 99.999% pure gold (Alfa Aesar)

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