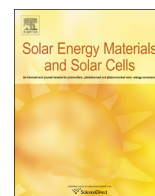




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Understanding the role of thickness and morphology of the constituent layers on the performance of inverted organic solar cells

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ABSTRACT

Inverted bulk heterojunction organic solar cells have generated significant interest in organic photovoltaic research due to much improved life times. Efficiency maximization for a given polymer–acceptor blend requires control of thickness and morphology of various constituent layers. In this manuscript, we present our results on the thickness variation of various constituent layers, viz. ZnO (electron transport layer), blend layer of poly(3-hexyl thiophene) and [6,6]-phenyl-C61 butyric acid methyl ester (active layer) and MoO₃ (hole extraction layer) vis-à-vis effect of morphology of layers and optical field distribution on the electrical performance of inverted organic solar cells. The peak performance of the devices occurs for 30 nm thick ZnO films, 100 nm thick active layer and 10 nm of MoO₃ layers with an average power conversion efficiency of approximately 3.2% and a fill factor of 61%. The maximization of the device performance upon thickness variation of device constituents appears to result from the combined effects of morphology and the optical electric field distribution in the active layer.

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1. Introduction

Organic solar cells (OSC) have emerged as a promising photovoltaic (PV) technology because of the possibility of low cost production and flexible devices [1]. The use of environmentally safer organic semiconductors and processing technologies such as thermal evaporation (small molecules) and spin coating, printing (conjugated polymers), allows them to compete with Si and other PV technologies. Polymer based OSCs have additional potential advantage of being fabricated at a large scale via roll to roll processing to provide high throughput of devices [2–4]. The most common strategy to maximize the efficiency of OSC devices is to mix the donor and acceptor materials in an optimum composition leading to a bulk heterojunction (BHJ) device. In BHJ, this inter-mixed state is bi-continuous and its interpenetrating network maximizes the area of the donor–acceptor interface to provide a large number of exciton dissociation sites [5]. In a normal structured OSC device, the active layer (ActL) of donor–acceptor blend is sandwiched between the indium tin oxide or ITO (anode) coated on the glass substrate and a low work function metal, for example, Al acting as cathode with device structure as ITO/PEDOT:

PSS/ActL/Ca/Al where poly (3,4-ethylenedioxythiophene):poly (styrenesulfonate) (PEDOT:PSS) is used as a hole transport layer (HTL) on the ITO anode side. The most commonly used blend consisting of poly(3-hexylthiophene) and [6,6]-phenyl-C61 butyric acid methyl ester (P3HT/PCBM) routinely yields device efficiencies between 3 and 4%.

While development of new donor and acceptor materials has resulted in higher device efficiencies with %PCE reported over 7% for donor–acceptor blend combination of PTB7/PC71BM (Poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]]/phenyl C61 butyric acid methyl ester) in normal device structure [6], one major impediment for OSC devices is their poor environmental stability resulting in fast device degradation and hence short life times, often limited to a few hundred hours. There are various mechanisms proposed for such behavior such as (i) presence of less air stable, Al electrode causing its oxidation, in turn, enhancing the series resistance of the device, (ii) the diffusion of oxygen and moisture via the electrode pin holes and grain boundaries and (iii) presence of corrosive and acidic hole transport layer PEDOT:PSS which causes the erosion of ITO in an acidic medium and subsequent In (indium) diffusion toward ActL in the OSC device and leading to poor device life times [7–9].

One of the suggested way to improve the life times of OSC devices is to use an inverted device structure in the form glass/ITO/ETL/ActL/HTL/Ag or Au (ETL: electron transport layer,

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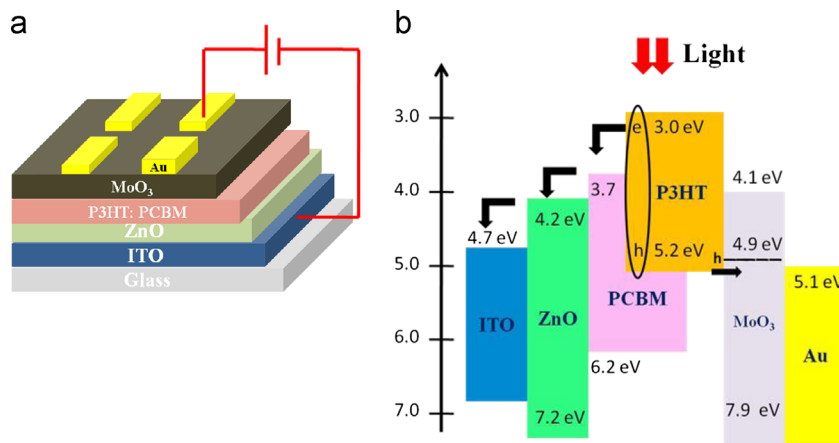


Fig. 1. (a) Schematic of an inverted OSC device. (b) Energy band diagram showing energy levels of ZnO and MoO₃ as cathode and anode buffer layers respectively w.r.t. ActL energy levels. Arrows show the flow of photo generated electrons (e) and holes (h) toward cathode and anode.

ActL: active layer) where ITO electrode is used for electron collection (cathode) while the top electrode is made of a high work function metal such as Ag or Au serving as anode for hole collection. Consequently, the charge transport in this device structure is opposite to that in the normal device structure. This layer stacking sequence is believed to provide superior ambient stability of the device in comparison to the devices using Al electrodes and PEDOT:PSS as HTL. Such an inverted configuration uses various n-type compounds with appropriate band gap and energy levels as electron transport layers (ETL) on top of ITO/Glass with examples being ZnO, TiO₂, Cs₂CO₃, CsF₂, zinc tin oxide (ZTO), and a-IZO (indium zinc oxide) [10–16]. These ETLs provide an energy barrier to hole flow toward ITO, leading to electrons only collection on the ITO side. In a similar fashion, HTL can be made of oxides such as CuO_x, WO₃, PEDOT:PSS, NiO, V₂O₅, and MoO₃ [17–20] which are believed to block the flow of electrons and excitons toward the high work function metal electrode i.e. anode (e.g. Au or Ag) on the top side. Peak device efficiency reported with such a device structure is approximately 7.4% based on a blend of donor polydithienogermole–thienopyrrolodione (PDTG–TPD) polymer and acceptor [6,6]-phenyl-C₇₁-butyric acid methyl ester (PC₇₁BM) [21].

Due to constraints imposed by high exciton binding energy and complexities of charge separation and their subsequent transport in OSC devices, photovoltaic characteristics are highly sensitive to the thickness and morphology of the constituent layers. While there are many reports elucidating the role of thickness and morphology of the active layer on OSC device performance [22–26], there are limited studies on similar aspects of ETL and HTL layers [7,27–30]. This aspect is especially crucial for the inverted OSC devices because ETL layers on ITO are typically made of high melting and resistive compounds, such as oxides e.g. ZnO. The processing of these materials can result in a variety of morphologies and hence, it is vital to examine the role of thickness and morphology of these layers to maximize the carrier transport across these layers to the respective electrodes. The objective of this work is to study the effect of thickness and morphology of all three layers, i.e. ZnO as ETL, MoO₃ as HTL and the ActL to maximize the performance of inverted organic solar devices. Our work shows that a particular thickness of these layers results in a peak device efficiency, which is governed by the morphological changes, light absorption in the device and optical electric field profile in the active layer.

2. Experimental and modeling details

The different constituent layers used in the inverted OSC device configuration are shown in Fig. 1(a). For realizing it, 99.9% pure

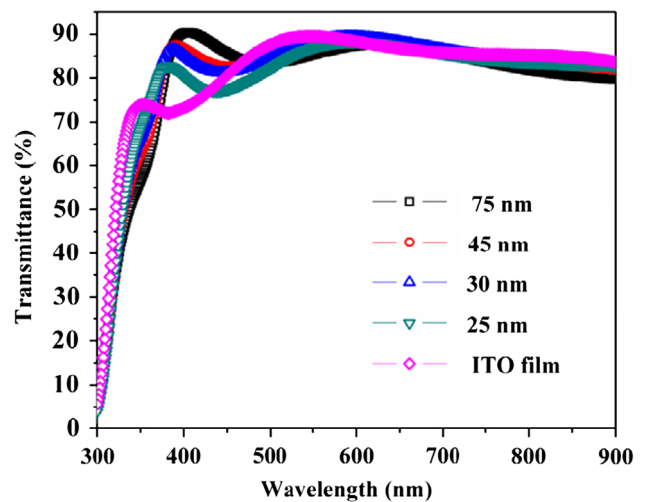


Fig. 2. UV–visible transmission spectra of ITO/ZnO films as a function of ZnO thickness.

regioregular poly 3-hexylthiophene (P3HT) and [6,6] phenyl C₆₁ butyric acid methyl ester (PCBM) were procured from Sigma Aldrich and Nano-C, respectively, while zinc acetate di hydrate (C₄H₁₀O₆Zn), 2-methoxy ethanol (C₃H₈O₂) and ethanolamine (C₂H₇NO) were procured from Sigma Aldrich. Organic solar cells were fabricated over patterned 120 nm thick ITO coated glass substrates having sheet resistance of approximately 10 Ω/□. Prior to device fabrication, the substrates were ultrasonicated for 10 min in a boiling soap solution, followed by ultrasonication in DI water, acetone, isopropyl alcohol respectively and drying with N₂ followed by exposure to 20 min of UV ozonization.

First, ZnO layers were spin coated directly on to the cleaned substrates from a solution prepared after mixing zinc acetate di hydrate (C₄H₁₀O₆Zn), 2-methoxy ethanol (C₃H₈O₂) and ethanolamine (C₂H₇NO) yielding a solution of 0.45 M concentration. Spin coated films were subsequently annealed at 250 °C for 10 min in air, yielding thicknesses of approximately 75, 45, 30 and 25 nm. For ActL fabrication, 10 mg poly 3-hexylthiophene (P3HT) and 10 mg [6,6] phenyl C₆₁ butyric acid methyl ester (PCBM) were mixed in 1 ml chlorobenzene followed by stirring for 12 h at room temperature. This solution was spin coated on ZnO coated substrates to yield thicknesses of 100, 82 and 70 nm. Subsequently these ActL coated substrates were transferred to a N₂ ambient where these were annealed at 130 °C for 30 min on a hot plate. These substrates were then transferred into the vacuum chamber

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