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Inverted polymer solar cells with indium sulfide electron selective layer

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ABSTRACT

The feasibility of indium sulfide thin films as electron selective layer in polymer solar cells has been investigated. Heterojunctions were fabricated using spray deposited indium sulfide and the conjugated polymer MEH-PPV in an inverted configuration. The influence of indium sulfide and polymer layer thickness on the photovoltaic properties of this heterojunction was studied. Optimum layer thickness was found to be 200 nm for indium sulfide and 100 nm for MEH-PPV. Under illumination, a short circuit current density (J_{sc}) of 0.133 mA/cm^2, an open circuit voltage (V_{oc}) of 0.63 V, fill factor (FF) of 44.92% and efficiency (η) of 0.075% have been obtained for this optimized device. The effect of anode work function on the photo-response of the device was also investigated. It was observed that, with silver electrode, the device exhibits enhanced photoactivity.

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1. Introduction

Polymer photovoltaics have emerged as a promising candidate for low priced renewable energy mainly because of the simplicity in its fabrication process. They come handy particularly in low power applications such as in powering cell phones, etc. even though their prospects for large scale power generation cannot be completely ignored [1–3]. In most polymer solar cells, the front electrode is a transparent conducting oxide, such as indium tin oxide (ITO), which functions as a high work function electrode for hole collection. For the efficient operation of the device, the back electrode must be a low work function metal such as calcium, which facilitates smooth collection of electrons [4]. However, these metals are reactive and get easily oxidized in air. This issue is normally addressed by capping the reactive metal with a less reactive one and by encapsulating the device to protect it from environmental effects during operation. On the other hand, the devices can be made more stable by avoiding the need for a reactive low work function top electrode. This can be accomplished by inverting the device geometry such that the holes and electrons generated in the active layer exit the device in the direction opposite to that in a normal device [5]. In this reversed geometry, a hole blocking layer usually an inorganic n-type semiconductor is inserted between the ITO and the active layer so that only electrons are collected by the ITO, whereas the back electrode becomes the hole collecting positive electrode, which can be made from a high work function metal that is more stable in air. Such inverted polymer solar cells with hole blocking layers made of ZnO, TiO_2, CdS, etc. are shown to have efficient photoconversion properties [6–11].

In this work we have used indium sulfide (In_2S_3) as the electron selective layer. In_2S_3 being an n-type compound semiconductor with high electron affinity, wide band gap and excellent photosensitivity should function as efficient electron acceptor/hole blocking layer in polymer photovoltaics. Its potential as buffer layer material in inorganic solar cells has already been demonstrated [12]. It can be easily prepared by cost effective techniques such as chemical spray pyrolysis, electrochemical method, etc. The deposition by chemical means offers the advantage that its properties can be easily tuned by the suitable choice of the precursors and controlling its concentration [13,14]. However there have been only a few reports on its use in polymer heterojunctions. Earlier, there had been a report on In_2S_3/polyaniline and In_2S_3/polypyrrole heterojunctions by Dalas et. al. [15]. Though these heterojunctions exhibited a diode like behavior in the dark, nothing was said about its photoactivity. Recently, Tang et. al. [16] have fabricated and investigated the photovoltaic properties of polymer heterojunction using In_2S_3 nanoparticles and the polymer MEH-PPV. They reported a short circuit current density (J_{sc}) of 15.5 μA/cm^2, open circuit voltage (V_{oc}) of 0.87 V, a fill factor (FF) of 23.9% and an efficiency (η) of 0.02% under monochromatic illumination of 16.7 mW/cm^2 at 500 nm for the device. In the present work, we have characterised In_2S_3 thin films prepared using chemical spray pyrolysis and fabricated a heterojunction by using the polymer MEH-PPV along with this material. Attempt has also been made to optimise thickness of In_2S_3 and MEH-PPV layers so as to obtain better performance of the device.

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2. Experimental details

2.1. Preparation of In_2S_3 thin films

In_2S_3 thin films were deposited on ITO coated glass substrates (thickness 2000 Å, sheet resistance 10 Ω/cm^2) by spraying aqueous solutions of indium chloride ($InCl_3$), and thiourea ($CS(NH_2)_2$), keeping the substrate at 350 ± 5 °C with a spray rate of 2 ml/min. Total volume sprayed was 20 ml. An automated spray unit indigenously fabricated in our lab was used for the film deposition. Parameters like spray rate, substrate temperature, pressure of carrier gas, distance between spray head and substrate can be controlled in this unit. More details related to automated spray unit and CSP technique has been described elsewhere [17,18]. In/S ratio in the films can be adjusted by varying the molar concentrations of respective solutions. We have prepared In_2S_3 thin films using precursor solutions having In/S ratio of 1.2/8. Thickness of the films were measured using the Stylus Profilometer (Dektak-6M) and was found to be approximately 200 nm. Thicker films were prepared by increasing the volume of the spray solution. However attempts to reduce the thickness of the films below 200 nm by reducing the precursor volume resulted in discontinuous films.

2.2. Characterization of In_2S_3 films

Structural analysis of the films was done using X-ray diffraction (XRD) with a Rigaku (D.Max.C) X-ray diffractometer (having CuK_α ($\lambda = 1.5405$ Å) radiation and a Ni filter operated at 30 kV and 20 mA). Optical absorption studies were carried out using a UV–Vis–NIR Spectrophotometer (JASCO V-570 model). Resistivity of the films prepared on glass substrates was measured using the two probe method employing a Keithley Source Measure Unit (SMU, K236).

2.3. Device fabrication

Heterojunctions were prepared by spin coating a solution of poly(2-methoxy-5-[2′-ethyl hexyloxy]-1,4-phenylene vinylene) (MEH-PPV) in chlorobenzene (5 mg/ml), on top of the In_2S_3 film deposited on ITO substrates. MEH-PPV was synthesized as described elsewere [19]. Layer thickness of MEH-PPV was approximately 100 nm and this was increased by increasing the concentration of the solution used for spin coating. For electrical contact at the top, silver electrodes of 12 mm^2 area were vacuum evaporated (at a pressure of 6×10^{-6} Torr) using a shadow mask on top of the polymer layer. Except for electrode deposition, all the other deposition and characterization process were carried out under ambient air atmosphere. The device structure is depicted in Fig. 1.

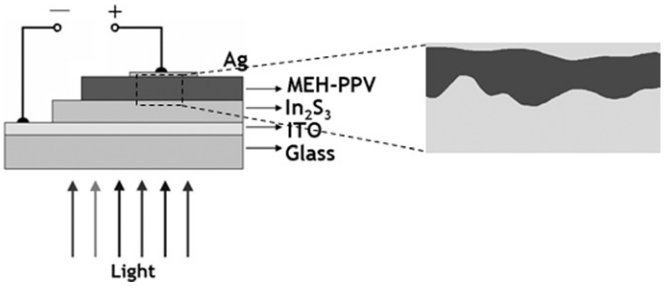

Fig. 1. Structure of In_2S_3/MEH-PPV heterojunction. Magnified portion shows the schematic of the interface between In_2S_3 and MEH-PPV.

The cross sectional SEM images of the bilayer was taken using a Hitachi SU6600 variable pressure field emission scanning electron microscope (FESEM). The *J–V* characteristics of the device were measured using the Keithley SMU, K236 and Metric's interactive characterization software (ICS). The device was illuminated using a tungsten halogen lamp, with an intensity of 50 mW/cm^2, on the substrate surface. An infrared filter, along with a water jacket, was used to ensure that there was no heating of the device during measurement.

3. Results and discussion

3.1. Structural, optical and electrical properties of In_2S_3 films

3.1.1. XRD analysis

Fig. 2 shows the XRD pattern of In_2S_3 thin films over ITO substrates. Well defined peaks corresponding to the [1 0 3], [1 0 9], [2 2 0], [3 0 9] and [4 0 0] planes of β-In_2S_3 could be clearly observed. The *d*-values coincided with that of β-In_2S_3 in standard JCPDS data card (25-390). It should be noted that the preferential orientation is along the [2 2 0] plane, except for the 200 nm film, for which it is along the [1 0 9] plane. The grain size was calculated from the Debye–Scherrer formula, $D = 0.9\lambda/\beta \cos\theta$, where D the grain size, λ the wavelength of the CuK_α line and β the full width at half maximum of the diffraction peak. The obtained grain size is given in Table 1.

3.1.2. Optical and electrical properties

Optical absorption spectra of In_2S_3 thin films for different thicknesses are presented in Fig. 3a. As can be seen, there is a shift in the absorption edge towards the higher energy side of the spectrum with the decrease in thickness of In_2S_3. Correspondingly, the band gap calculated from the $(\alpha h\nu)^2$ versus $h\nu$ plot (Fig. 3b) shows an increase from 2.58 to 2.75 eV, as the thickness decreases from 550 to 200 nm.

Resistivity and photosensitivity of the films were of the order of $10^5\ \Omega$ cm and 10^2, respectively. Photosensitivity (S) is calculated using the formula, $S = (I_L - I_D)/I_D$, where I_L and I_D are the photocurrent and dark current, respectively.

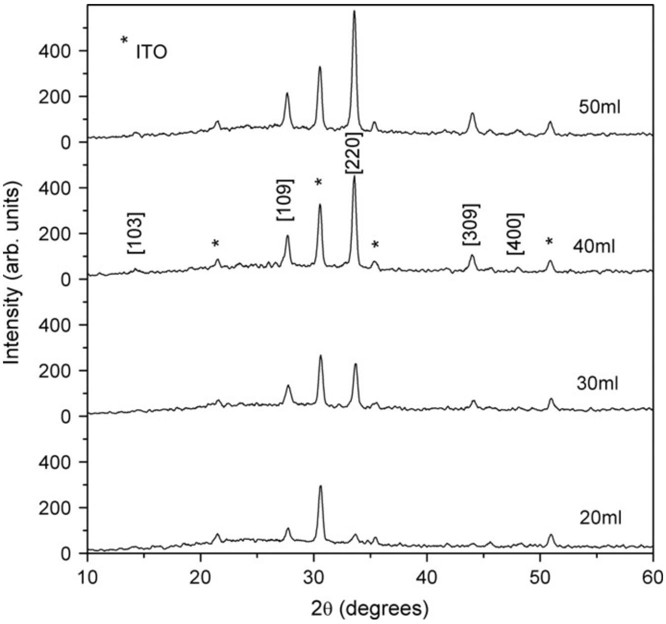

Fig. 2. X-ray diffraction pattern of In_2S_3 thin films over ITO substrates.

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