

Influence of poly3-octylthiophene (P3OT) film thickness and preparation method on photovoltaic performance of hybrid ITO/CdS/P3OT/Au solar cells

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ABSTRACT

Chemically synthesized poly3-octylthiophene (P3OT) solution was spin-coated (SC) or drop-cast (DC) on cadmium sulfide thin film to form a hybrid junction of transparent conductive glass (indium-tin oxide (ITO))/CdS/P3OT/Au. XRD patterns of DC P3OT films show a couple of peaks at 2θ between 8° and 13° . It suggests that a long-range order may exist in the slowly dried DC P3OT films. Optical absorption coefficient spectra of P3OT films exhibit the presence of polaron bands in P3OT films with thickness lower than 700 nm, which indicates that thin P3OT films are susceptible to oxygen doping. The microscopically more ordered DC P3OT films show a coplanar photoconductivity, which was absent in the SC P3OT ones. The effect of P3OT film preparation method and ambient conditions was also notable on the photovoltaic performance of CdS/P3OT-based solar cells. The potential barrier of the CdS/P3OT heterojunctions decreased as the P3OT film thickness reduced from 671 to 106 nm, which was related to the diffusion of oxygen into the polymer film. As the P3OT film thickness increased to more than 1000 nm, the open-circuit voltage (V_{OC}) reached to around 1 V, close to the value suggested by fitting the current–voltage curves with the Schottky barrier diode model. But at the same time, the short-circuit current density (J_{SC}) of the same cells is lowered due to the increasing charge recombination effect inside the polymer material. The simple and economic drop-cast preparation method is promising for polymer film deposition in hybrid solar cell fabrication.

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1. Introduction

Since the first reported conducting polymer-based photovoltaic (PV) cells in the early 1980s, the research works aimed on synthesis and characterization of conjugated macromolecules are continuously documented worldwide with the objective of obtaining, in a near future, cost-effective organic photovoltaic cells. Hybrid organic–inorganic heterojunctions are those solar cells that consist of a p-type semiconductor polymer and an n-type inorganic semiconductor. They combine the characteristics of high electron mobility of the inorganic semiconductor and the mechanical flexibility of the organic one. The geometric structure of a hybrid heterojunction can be mixed (bulk) or bi-layered. In the first case, the two semiconductor materials were blended to form an interpenetrated network, and in the second one, the polymer layer was usually coated on top of the inorganic film. According to numerical simulations [1], as the mixing percentage increases, the exciton dissociation increases and the diffusion

counter-current decreases, resulting in substantially greater short-circuit currents (J_{SC}) but reduced open-circuit voltages (V_{OC}); larger open-circuit voltage is gained for layers and drops to the value of the built-in potential for homogeneous blends. A recent review on hybrid solar cells has resumed different types of organic/inorganic heterojunctions and their photovoltaic performance [2].

Till now most reported hybrid cells are based on a blended structure to seek a higher J_{SC} and consequently higher conversion efficiency, but a simple layered organic/inorganic heterojunction gives a better understanding about structure–property relation in these cells. Examples of layered hybrid solar cells include TiO_2 with poly(2-ethoxy-5-(2-ethyl)hexoxy-phenylene-vinylene) (MEH-PPV) [3–8], TiO_2 with poly3-hexylthiophene (P3HT) or poly3-octylthiophene (P3OT) [7–10], CdS with P3OT [11,12], etc. All these heterojunctions have shown high V_{OC} values under 100 mW/cm^2 illumination, from 0.7 to 1.1 V, and J_{SC} can be more than 1 mA/cm^2 , depending on the cell preparation process. Due to the short exciton diffusion distance in conducting polymers, the polymer film thickness is suggested to keep smaller than 100 nm to reduce the electron–hole recombination effect, and to get as high as possible the value of J_{SC} .

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Another issue related to the use of organic semiconductors in solar cell applications is the material stability. It is reported that oxygen could diffuse through the 265 nm thick MEH-PPV film and affect the bulk of the polymer, leading to an increased trapping for reverse bias [13]. For exposure to water, it is found the strongest effect at the surface of polymer with the metal contact (Al) rather than in the bulk or at the back contact (polymer/indium-tin oxide (ITO) interface) [13]. In the case of poly3-alkylthiophenes (P3AT), it is probed that under increasing pressures of oxygen the carrier concentration as well as the conductivity is increased, but the charge carrier mobility is lowered by the formation of the charge transfer complex between molecular oxygen and P3AT [14]. It is observed that those polymers possessing low ionization potentials and having an amorphous nature, which facilitate diffusion of oxygen into the bulk, will be particularly susceptible to charge transfer complex formation in the presence of oxygen [14]. Although most of the conducting polymer-based PV cells are prepared and tested under inert ambient, it is important to understand how the ambient oxygen affects the PV cell performance and to keep looking for a simple and economic method for the PV cell preparation. In this work, layered P3OT/CdS solar cells were prepared and analyzed under ambient conditions. P3OT film thickness and the polymeric film preparation method (drop-cast (DC) vs. spin-coating (SC)) were experimental parameters. From the technological point of view, the drop-cast method avoids the waste of polymer solution during the device fabrication in comparison with the spin-coating one. The comparison of these two film preparation methods gives us an insight to optimize the cell preparation process and to improve the solar cell efficiency.

2. Experimental

CdS thin film was deposited by chemical bath deposition [15] on transparent conductive glass substrate (indium-tin oxide, glass with sheet resistance of 8–12 Ω/\square , Delta Technologies). A cadmium salt solution was prepared by mixing 25 mL of 0.1 M $\text{Cd}(\text{NO}_3)_2$, 15 mL of 1 M sodium citrate, 2 mL of concentrated ammonia, 5 mL of 1 M thiourea and 53 mL of distilled water. Clean ITO substrates were immersed in the freshly prepared cadmium solution at 80 °C. After 1 h of deposition, 100 nm thick of CdS film had grown on top of ITO. P3OT was synthesized by oxidative polymerization, as described elsewhere [16]. The molecular weight of the polymer product was 78,100 with a polydispersity of 1.86 and the head-to-tail percentage in the product of about 76% [17]. The black P3OT powder was dissolved in toluene, and the resulting P3OT solution showed an orange-reddish color.

P3OT films were formed by drop-casting or spin-coating the P3OT solution on clean glass substrate or CdS film in air under ambient conditions (22 °C and 35% humidity). For drop-cast P3OT films, the P3OT solution concentration varied from 2.5 to 10 mg/mL (from 0.013 to 0.051 M). A certain volume of P3OT solution was dropped on top of CdS film and dried slowly at 60 °C to form a P3OT layer. For spin-coated P3OT samples, a rotation speed of 2500 rpm was used and the concentration of P3OT solutions varied from 5 to 20 mg/mL (from 0.025 to 0.102 M). After the P3OT film deposition on CdS, the samples were annealed in vacuum (10^{-2} Torr) at 90 °C for 3 h. The thickness of CdS and P3OT films

Table 1
P3OT film thickness as functions of P3OT solution concentration and the film preparation method.

P3OT solution concentration (mg/mL)	2.5	5	10	20
Spin-coated (SC) P3OT film thickness (nm)		106	186	334
Drop-cast (DC) P3OT film thickness (nm)	671	2490	7521	

was measured by Alpha-Step 100 (Tencor). Table 1 shows the relation between the P3OT solution concentration, the film preparation method and the P3OT film thickness. Six ITO/CdS/P3OT/Au heterojunctions were prepared with P3OT films of different thickness as indicated in Table 1: three from spin-coating and three from drop-casting. Gold contacts were evaporated on top of P3OT surface. The P3OT film thickness has been chosen larger than 100 nm to prevent a possible short circuit.

XRD patterns of P3OT films were obtained on Rigaku DMAX-2200 X-ray diffractometer with a low grazing-incidence angle of 0.5°. Optical absorbance spectra of the polymeric films were recorded in the UV–vis region in a Shimadzu spectrophotometer 1601 PC. Photoconductivity of P3OT films was measured under 1 V polarization with coplanar Au electrodes on top of the polymeric films. The electrodes were of about 4 mm length and separated by a distance of about 4 mm. A Tungsten-Halogen lamp of 40 mW/cm² was used for photoconductivity measurements. Current–voltage (*I–V*) curves of ITO/CdS/P3OT/Au heterojunctions were taken under ambient conditions in dark and under illumination with a solar simulator (Oriel). The intensity of the Xenon lamp was adjusted to 100 mW/cm². All electrical characterization of the cells was carried out in air under ambient conditions.

3. Results and discussion

Fig. 1 shows the XRD patterns of DC and SC P3OT films. The three straight lines in the figure represent the diffraction peak position and relative intensity (R.I.) of a monoclinic P3OT at 2θ equal to 37.933° (R.I. = 100), 61.797° (R.I. = 32) and 22.783° (R.I. = 13) from the data base of JCPDS (#49-1977). Because of the low diffraction intensity of the crystalline peaks, it seems that both types of P3OT films were mostly amorphous with a small portion of the crystalline phase. The last one shows a preferential diffraction plane of (311) at $2\theta \sim 23^\circ$. However, a couple of additional small diffraction peaks at $2\theta \sim 8^\circ$ and 12.5° were observed in the DC P3OT film samples. These low-angle diffraction peaks should indicate some kind of long-range order in the molecular arrangement of DC P3OT film, which could be originated from the thermally assisted slow solvent evaporation process in the drop-cast method in comparison with the room-temperature fast dried spin-coated one.

The repercussion of this long-range order is also reflected in the optical absorption coefficient spectra of DC P3OT films. All the

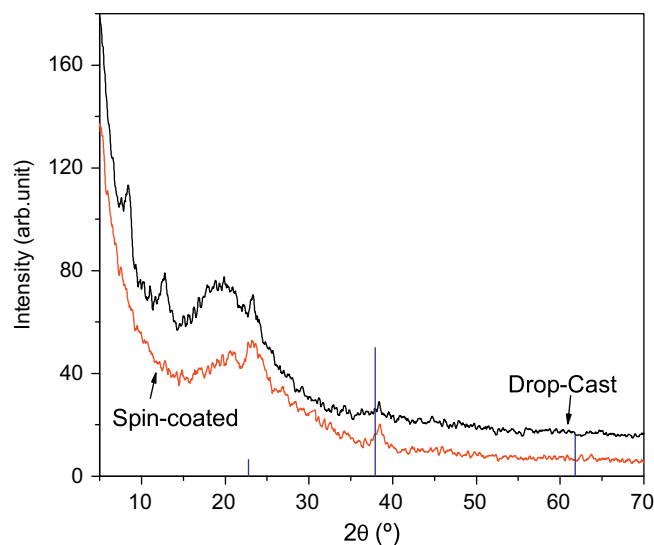


Fig. 1. XRD patterns of drop-cast (DC) and spin-coated (SC) P3OT films.

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