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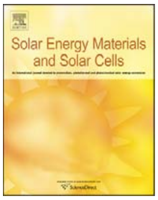

Lengthening the polymer solidification time to improve the performance of polymer/ZnO nanorod hybrid solar cells

Chen-Yu Chou [a], Jing-Shun Huang [a], Chung-Hao Wu [c], Chun-Yu Lee [a], Ching-Fuh Lin [a,b,c,*]

[a] *Institute of Photonics and Optoelectronics, National Taiwan University, Taipei 10617, Taiwan, Republic of China*
[b] *Graduate Institute of Electronics Engineering, National Taiwan University, Taipei 10617, Taiwan, Republic of China*
[c] *Department of Electrical Engineering, National Taiwan University, Taipei 10617, Taiwan, Republic of China*

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ABSTRACT

Lengthening the polymer solidification time in the inverted configuration of polymer/ZnO nanorod hybrid solar cells is studied as a way to improve device performance. As the polymer solidification time is lengthened by lowering the spin-coating rate of the photoactive layer, the photoactive layer becomes thickened, and the polymer chains have enough time to self-organize and effectively infiltrate into ZnO nanorod spacing. While the thickness of the photoactive layer is increased to 400 nm accompanying self-organized polymer, the power conversion efficiency of the device is improved to 3.58% with an enhanced fill factor of 58%. The 400 nm film is composed of the highly ordered polymer and the ZnO nanorod arrays, resulting in light harvesting without decreasing the possibility for charge transport.

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1. Introduction

Solar cells attract great attention owing to the growing need for renewable energy. Polymer solar cells offer the potential for large-scale power generation based on materials that provide flexibility, light weight, low-cost production and low-temperature fabrication [1–6]. The most common and efficient material system thus far for polymer devices is the one consisting of poly (3-hexylthiophene) (P3HT) and (6,6)-phenyl C_{61} butyric acid methyl ester (PCBM) [7,8]. Several groups have developed methods to improve the power conversion efficiency (PCE) of the polymer solar cells [1,7,9]. However, the conventional bulk-heterojunction (BHJ) architecture has limitations in device stability. Exposure of the conventional solar cells to air leads to oxidation of the Al electrode and degradation of the indium–tin oxide (ITO)/ poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) interface because of the acidic nature of PEDOT:PSS [10,11]. Moreover, there exist problems of inherently poor polymer properties, such as the short exciton diffusion length and the relatively low carrier mobility, which limit the usefulness of thick films. A possible approach to overcome the above difficulties is to use inverted configuration with nanostructured semiconductor oxides due to their excellent electron mobility and the long diffusion length [12–18]. The inverted structure has the advantage of improved stability by replacing the low work function metal cathode and PEDOT:PSS, which are both detrimental to device lifetime [10,11]. On the other hand, the environmentally friendly and low-cost ZnO nanorod array is particularly well suited for this application as they can be grown normally to the substrate at low temperature ($<100\,^{\circ}$C) using a hydrothermal method [19]. ZnO nanorod arrays in the polymer solar cells improve charge carrier collection and transport [13–16,20–23]. Therefore, the ZnO nanorods provide the possibility of thick active layer for light harvesting without sacrificing charge transport. However, the poor infiltration of polymer into the spacing between ZnO nanorods causes a low fill factor (FF) of the polymer/ZnO nanorod hybrid solar cells (ITO/ZnO nanorod array/P3HT:PCBM/Ag) [13,14].

In this study, we report the slow-drying process, which lengthens the solidification time and optimizes the thickness of the photoactive layer, to improve the PCE and FF of the inverted structures of the polymer/ZnO nanorod hybrid solar cells. Although Li et al. [7] demonstrated that the efficiency of the polymer solar cells in conventional configuration could be improved by controlling the growth rate of the active layer, there is still no report on using the slow-drying process for inverted polymer/ZnO nanorod hybrid solar cells. Here we report the in-depth investigation on the influences of slow-drying process on the performance of the inverted hybrid solar cells. The slow-drying process not only gives the polymer chains enough time to

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[*] Corresponding author at: Department of Electrical Engineering, National Taiwan University, Taipei 10617, Taiwan, Republic of China.
Tel.: +886 2 33663540; fax: +886 2 23642603.
E-mail address: cflin@cc.ee.ntu.edu.tw (C.-F. Lin).

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self-organize but also helps polymer infiltrate into the ZnO nanorods spacing. The thickened active layer using the slow-drying process on polymer/ZnO nanorod solar cells is discussed. The morphology and the infiltration of the active layer with the lengthened drying time are studied using atomic force microscope (AFM) and field emission scanning electron microscope (FESEM) techniques. The optical properties of the slowly dried active layer films are examined by ultraviolet–visible (UV–vis) absorption spectroscopy. Moreover, combined with the advantages of ZnO nanorod array using solution processing and Ag as an anode allowing the use of the nonvacuum printing technique [24], the slowly dried devices in this study can provide a route for high-efficiency polymer/ZnO nanorod hybrid solar cells devices by all-solution processes.

2. Experimental details

All devices were prepared on ITO-coated glass substrates. A ZnO seed layer (~30 nm) was deposited from a 0.5 mol solution of zinc acetate dihydrate in 2-methoxyethanol by spin-coating as described previously [19]. ZnO nanorod arrays (length~140 nm, density~767.9 rods/μm^2) were subsequently grown from the ZnO seed-coated substrates in an aqueous solution of 50 mM zinc nitrate and 50 mM hexamethylenetetramine at 90 °C for 1 h using the hydrothermal method [19,25].

The active layer was fabricated by spin-coating a solution of 1.5 wt% of P3HT and PCBM (1:1 by weight) in dichlorobenzene (DCB) solution on top of the ZnO nanorod arrays. The P3HT with a weight average molecular weight (MW) of 50 000 g mol^{-1} was purchased from Rieke metals, and the PCBM was from Nano-c. The medium molecular weight P3HT was used because either high or low MW has disadvantages. That is, low-MW P3HT has low hole mobility, while high-MW P3HT has solubility problem [26]. We controlled the film drying time and thickness with the spin-coating speed. The spin-coating rate was varied from 400 to 1000 rpm. In addition, in order to provide the effects of slow drying at the same thickness, we performed another experiment. We controlled the drying time at the same thickness by spin-coating at 800 rpm and lowering the solvent evaporation rate. We used the mixed solvent of 3% 1-bromonaphthalene in DCB to lengthen the solvent evaporation time. Due to the high boiling point additive (1-bromonaphthalene), the drying time is lengthened. The blend layers were subsequently dried in air at room temperature. Finally, the Ag (150 nm) top electrode was thermally evaporated under vacuum (~3×10^{-4} Torr). The devices were placed in air without encapsulation. A schematic view of the device geometry is shown in Fig. 1.

All electrical measurements were taken in atmosphere environment. The active area of the devices was defined as 0.1 cm^2 by using a photomask. Current density–voltage (J–V) curves were measured with a Keithley 2400 source meter, under illumination at 100 mW/cm^2 from a 150 W Oriel solar simulator with AM 1.5G filters. The light intensity was calibrated with a calibrated standard solar cell with a KG5 filter that is traced to the National Renewable Energy Laboratory. The cross-sectional morphologies of the BHJ polymer-ZnO hybrid solar cells were examined using a FESEM. The absorption spectra were taken using a Perkin Elmer Lambda 35 UV–vis spectrophotometer.

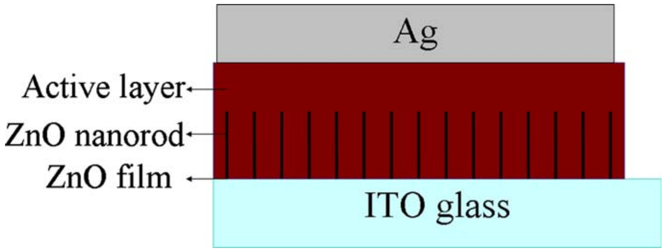

Fig. 1. Schematic view of a device structure of polymer/ZnO nanorod hybrid solar cells.

3. Results and discussion

The slower the spin-coating rate, the longer the drying time (t_d) required for the film to solidify, as detected by the time required for a dramatic change in the color of the film from orange to dark purple when it is transformed from the liquid to the solid phase. We observed that the t_d increases from 5 to 54 min, and the thickness of the active layer increases from 240 to 400 nm while the spin rate decreases from 1000 to 400 rpm. The relationship between t_d and spin-coating rate is shown in Fig. 2. Fig. 3 shows the J–V curves under illumination for the devices with the t_d varying from 54 to 5 min, corresponding to the spin rate varying from 400 to 1000 rpm. Compared to devices with t_d of 4, 12, 26,

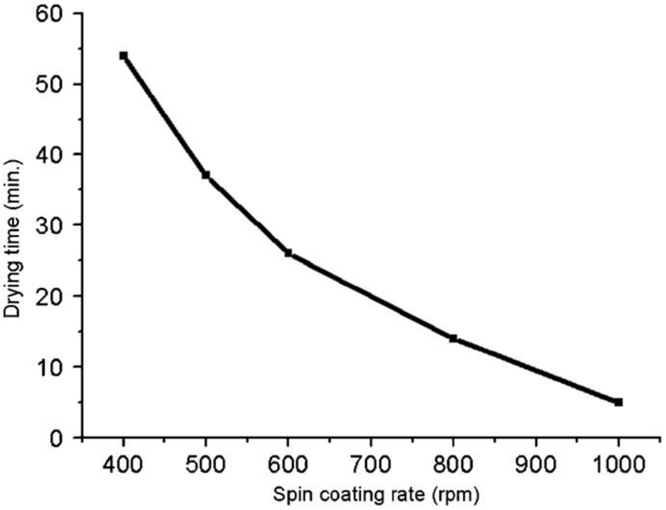

Fig. 2. The drying time (t_d) is plotted as a function of the spin-coating rate.

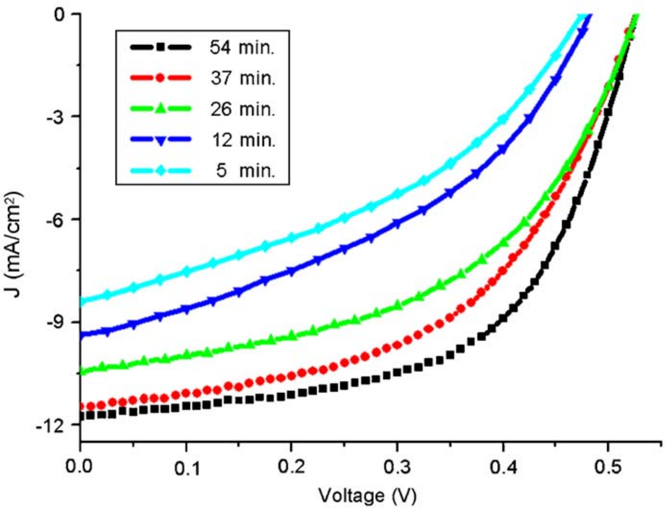

Fig. 3. The J–V curves under illumination of polymer/ZnO nanorod hybrid solar cells with different drying times of 5, 12, 26, 37, and 54 min, respectively.

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