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The effect of ITO/Mo/MoO₃ anode multilayer film on efficient hole extraction in MEH–PPV/ZnO NP hybrid solar cells

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Abstract

Acidic PEDOT:PSS anode buffer layers (ABLs) are widely used for improving efficiency in organic-based solar cells. Recently stable P-type metal oxides, such as NiO, MoO₃, and V₂O₅, have been applied for replacing the commonly used acidic PEDOT:PSS ABL because of the difficulty in coating PEDOT:PSS solutions onto electrodes or polymer active layers. This highlights the importance of the fabrication and characterization of various ABLs that affect the efficiency of hole extraction and the blocking of electron backflows. We studied MoO₃ ABLs formed by the rapid thermal annealing (RTA) of molybdenum (Mo) in oxygen atmosphere. In particular, we propose the use of an indium tin oxide (ITO)/Mo/MoO₃ anode multilayer for efficient hole extraction in bulk heterojunction (BHJ)-structure organic-based solar cells. The formation of Mo and MoO₃ thin films was controlled by the RTA time with a fixed annealing temperature of 350 °C. We obtained a two-fold improvement in the short circuit current (J_{sc}) of the device with the ITO/Mo/MoO₃ anode multilayer film (AMF), compared to a device with a PEDOT:PSS ABL.

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Keywords: Buffer layer; Hybrid solar cells; MoO3; ZnO nanoparticles; RTA

1. Introduction

Organic solar cells have been intensively studied for their low manufacturing cost, processing expediency, and potential application to flexible devices. Recently, inorganic materials were used in attempts to combine them with organic solar cells. For efficient charge carrier separation from charge carrier generation in the light absorption layers, the electrical properties of the inorganic materials and the control of the surface morphology between the polymer and the inorganic materials are important [1,2]. Not only inorganic materials such as ZnO [3], CdSe [4,5], and TiO₂ [6] but also their structures, such as thin films [7,8], nanorods [9,10], nanoparticles (NPs) [11], and nanofibers [12], have been widely investigated to improve the efficiency. In particular an active layer structure of polymer and ZnO NPs is regarded as the most promising candidate

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configuration, because of its similarities to organic bulk heterojunction (BHJ) solar cells [13,14].

In these BHJ structures, anode buffer layers (ABLs) are very important because they affect the efficiency of hole extraction and the blocking of the electron backflow [15]. An acidic PEDOT: PSS ABL is commonly used because of its applicability in solution processes and its appropriate electrical and optical properties [16,17]. Nevertheless it is difficult to coat acidic PEDOT:PSS ABLs uniformly onto the anode or polymer active layer, because of their hydrophilic and unstable properties [18]. For these reasons, stable p-type metal oxides such as CuO₂ [19], MoO₃ [20,21], and V₂O₅ [22] have been used in attempts to replace the commonly used acidic PEDOT:PSS ABL. MoO₃ thin film is most widely used in organic solar cells, because of its good stability and appropriate electrical and optical properties. Therefore, the fabrication and characterization of a diverse array of ABLs are of great importance in organic-based solar cells.

We have systematically studied MoO₃ ABLs fabricated by rapid thermal annealing (RTA) for MEH–PPV/ZnO NP hybrid

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solar cells. Particularly, we introduced indium tin oxide (ITO)/ Mo/MoO₃ anode multilayer films (AMFs) to improve the hole extraction in organic-based solar cells. The structural properties of the ITO/Mo/MoO₃ AMFs were studied by scanning electron microscopy (SEM) and high-resolution X-ray diffraction (HRXRD). The optical and electrical properties were also studied using a UV–vis spectrophotometer and a Hall measurement system. The photovoltaic properties of MEH–PPV/ZnO NPs hybrid solar cells with the ITO/Mo/MoO₃ AMFs were compared with those of devices without an anode multilayer.

2. Experimental method

MEH–PPV polymer powder, ZnO NPs (in ethanol 40 wt%), aluminum (Al) wire, and all other chemicals were purchased from Sigma-Aldrich Co. Ltd. ITO-deposited soda–lime glass substrates were sequentially cleaned ultrasonically in acetone, isopropyl alcohol, and deionized water, and dried in nitrogen gas. Cleaned glass/ITO substrates were loaded into an RF magnetron sputter chamber and treated with plasma under 30 W RF power for 60 s. After plasma treatment the sample was moved to a DC sputter chamber by a transferring system while maintaining a vacuum, followed by the deposition of a molybdenum (Mo) thin film with an argon pressure of 1 Pa. The DC magnetron sputter chamber was equipped with an 8-in. target, and a pressure of 3×10^{-6} Pa was maintained to prevent the target from oxidizing. The deposited glass/ITO/Mo sample was annealed in an RTA processor with various RTA conditions. During oxidation of the Mo layer the chamber was purged with 9 standard liters per minute of oxygen, and the annealing temperature was set to 350 $^{\circ}$ C. The duration was changed from 60 s to 120 s.

In order to fabricate hybrid solar cells with the ITO/Mo/MoO₃ AMF, a mixed solution of MEH–PPV (5 mg/mL):ZnO NPs (0.5 wt% to MEH–PPV) was coated with a spinning speed of 2000 rpm on the ITO/Mo/MoO₃ AMF. For annealing the mixed solution layer, coated glass/ITO/Mo/MoO₃/MEH–PPV:ZnO samples were moved to the RTA processor and annealed with nitrogen gas at an annealing temperature of 105 °C for 10 min. Finally, a 100 nm Al cathode was deposited with a patterned shadow mask on the active layer at a pressure of 4×10^{-6} Pa.

SEM was performed on a Hitachi S-4700 scanning electron microscope. The crystal structure of the AMFs was characterized using the HRXRD technique with high-quality Cu K α radiation (λ =1.541 Å), with a 2 θ scan range between 20° and 90°. Transmittance and absorption spectra were measured on a Cary 500 scan UV–vis–NIR spectrophotometer. The electrical properties were measured by a Hall measurement system (Ecopia, HMS-5000, Chandler Heights, USA) and a four-point probe. The energy conversion efficiencies were measured under 100-mW/ cm² illumination from an AM1.5 solar simulator.

3. Results and discussions

To confirm the formation of MoO₃ by increasing the annealing temperature, SEM micrographs were examined.

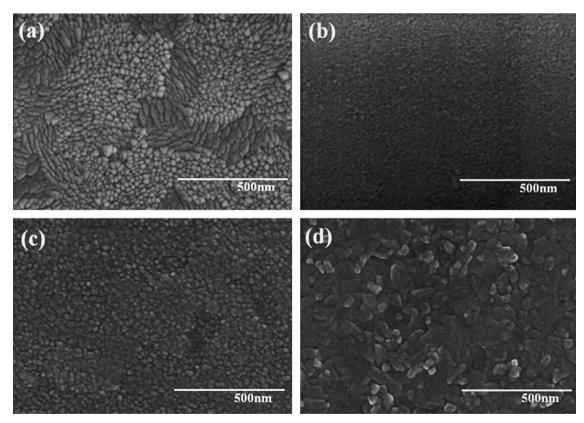


Fig. 1. SEM images of various multilayer films as a function of RTA conditions: (a) ITO anode, (b) 20-nm Mo deposited on ITO, (c) annealed ITO/Mo at 350 °C for 60 s, and (d) annealed ITO/Mo at 350 °C for 120 s.

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