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Improvement of transparent silver thin film anodes for organic solar cells with a decreased percolation threshold of silver



Zhizhe Wang, Chunfu Zhang*, Rui Gao, Dazheng Chen, Shi Tang, Jincheng Zhang, Dong Wang, Xiaoli Lu, Yue Hao*

State Key Discipline Laboratory of Wide Band Gap Semiconductor Technology, School of Microelectronics, Xidian University, 2 South Taibai Road, Xi'an, China 710071

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ABSTRACT

Organic solar cells (OSCs) using thermally evaporated Ag thin films as the transparent anode are investigated with poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) films as the active layer. The optimal power conversion efficiency (PCE) of 2.57% is obtained for the device with the 11 nm thick Ag layer, namely the percolation threshold of Ag. Then an interlayer of MoO₃ between the Ag anode and glass substrate is introduced. Two different thicknesses of MoO₃ (2 nm or 10 nm) are chosen here, corresponding to the unclosed or closed MoO₃ layer, respectively. It is observed that the introduction of the MoO₃ interlayer can effectively improve the wetting of Ag on the substrate and reduce the percolation threshold of Ag. When the MoO₃ thickness is 10 nm, since the surface energy of Ag ($\gamma=1.25 \text{ Jm}^{-2}$) is higher than that of MoO₃ ($\gamma=0.06 \text{ Jm}^{-2}$), the Ag–Ag interactions are stronger than the Ag–substrate interactions, weakening the surface-modifying effect. In contrast, since the 2 nm thick unclosed MoO₃ layer may create preferred nucleation sites on the substrate to enhance the lateral growth of Ag film effectively, a very low sheet resistance of $9.32 \Omega/\square$ and a relatively high transmittance are obtained for MoO₃ (2 nm)/Ag (9 nm) anode. The PCE of corresponding OSCs can be improved to 2.71%, comparable to that of ITO-based reference OSCs (PCE of 2.85%). From our results, it is concluded that the performance of the Ag thin film electrode can be improved by decreasing the percolation threshold of Ag with a MoO₃ interlayer (especially when the MoO₃ layer is thin). It may be instructive for the further research of metal thin film electrodes.

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1. Introduction

With the advantages of low cost, light weight, simple process, flexibility and possibility of roll-to-roll mass production, organic solar cells (OSCs) have been in the focus of research interest over the past decade [1–5]. Since the first report of OSCs based on the heterojunction structure by Tang [6], continuous improvement in materials, device architectures and device processing have resulted in considerable advancement in the performance of OSCs [7], particularly in the power conversion efficiency (PCE). Recently, OSCs with certified PCE above 10% have been reported [8], which is approaching the requirement of commercialization.

Owing to its excellent properties in optical transparency and electrical conductivity, indium tin oxide (ITO) sputtered on glass has become the commonly-used transparent anode for OSCs. However, cheap roll-to-roll fabrication of OSCs relies on the

flexible electrodes with low cost, which ITO is unsuitable for. For example, the limited indium source and stringent deposition conditions (usually sputter) leads to significant cost for ITO electrode [9]. Additionally, both the brittleness of ITO and high temperature processes in the deposition of ITO make it incompatible with the flexible substrates in roll-to-roll mass production [10]. Hence, more and more attention has been paid to searching for alternative transparent conductive materials. Indium-free transparent conducting oxides (such as Ga-doped ZnO (GZO) and Al-doped ZnO (AZO)) [11–13], Ag nanowires [10], graphene [9] and carbon nanotubes [14] are successfully employed as transparent electrodes to replace ITO. However, although the cost of GZO and AZO is lower than ITO, the deposition of these electrodes still requires the expensive sputter. Due to the surface roughness of Ag nanowire, graphene and carbon nanotube electrodes, charge injection into or extraction from organic active materials has been limited [9,10,15]. In addition, complex film processing also makes these electrodes unsuitable for application in large-scale devices [9,10,15].

In contrast, a smooth and continuous metal thin film (for example Ag) can be easily deposited by simple thermal evaporation, suitable for

* Corresponding authors. Tel./fax: +86 29 88201660.

E-mail addresses: cfzhang@xidian.edu.cn (C. Zhang), yhao@xidian.edu.cn (Y. Hao).

application in mass production. Moreover, due to their intrinsic flexibility and high conductivity [15], metal thin film electrodes are also suitable for application in OSC roll-to-roll production with the flexible substrates. Thus, more and more attention is attracted to this topic. Single layer electrodes (such as Au [16], Ag [17] and Cu [18]) and multilayer electrodes (such as Cu/Ni [19], MoO₃/Au/MoO₃ [15], MoO₃/Ag/MoO₃ [20], WO₃/Ag/WO₃ [21] and so on) are reported and used in the fabrication of OSCs. However, most of previous work has been mainly focused on the effect of optical interference and the variation in the species of the metal. It should be noted that making the metal layer as thin as possible while maintaining its good optical and electrical properties is of vital importance to improve the performance of metal thin film electrodes no matter they are single layer or multilayer electrodes. However, few studies have been done in the view of how to reduce the metal percolation threshold. That is what we concern about in this paper.

In our work, first, an Ag thin film electrode is employed as an ITO replacement by direct thermal evaporation of Ag on the glass substrate. ITO-free OSCs based on this anode are fabricated, with poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyrac acid methyl ester (PCBM) films as the active layer. The highest PCE of 2.57% is achieved for the device with the 11 nm thick Ag layer (the percolation threshold of Ag in our experiments), under 1 sun AM 1.5 G simulated illumination, comparable to that of ITO-based reference OSCs (PCE of 2.85%). Then an interlayer of MoO₃ between the Ag anode and glass substrate is introduced. Since a closed MoO₃ layer is formed with the approximate thickness of 4 nm [22], two different thicknesses of MoO₃ (2 nm or 10 nm) are investigated here. To our surprise, an interesting phenomenon is observed. The introduction of the MoO₃ interlayer can effectively enhance the lateral growth of Ag thin film and reduce the percolation threshold of Ag. Moreover, this surface-modifying effect is more obvious or stronger for the 2 nm thick MoO₃ layer. With the introduction of the 2 nm thick MoO₃ interlayer, the 9 nm thick Ag layer not only achieves similar conductivity as the 11 nm thick Ag layer but also shows a better optical transparency. Owing to the improvement in the performance of the electrode, the PCE of OSCs based on this MoO₃ (2 nm)/Ag (9 nm) anode can be enhanced to 2.71% (also the highest PCE among all the ITO-free OSCs in this paper). Thus it is concluded that the performance of the Ag thin film electrode can be improved by decreasing the percolation threshold of Ag with a MoO₃ interlayer (especially when the MoO₃ layer is thin). It is instructive for the further research of metal thin film electrodes.

2. Experimental section

2.1. Material and substrate preparation

P3HT was purchased from Rieke Metals Inc., PCBM was purchased from Nano-C Inc., and 1,2-dichlorobenzene and MoO₃ were provided from Aldrich Inc. All the materials were used without any further purification. P3HT and PCBM were dissolved in 1,2-dichlorobenzene with a concentration of 20 mg/mL, respectively. They were mixed in a weight ratio of 1:0.8 and stirred at room temperature for 2 h before use. Glass substrates were cleaned sequentially with detergent (Decon 90, UK), deionized water, acetone and ethanol in an ultrasonic bath for about 15 min.

2.2. Electrode deposition and characterization

Cleaned glass substrates were dried with a nitrogen (N₂) flow and then transferred into a custom-made multichamber ultrahigh vacuum evaporation system. Since 32 samples are fabricated in one processing run, the intentional variation of specific parameters is possible for specific samples while all other parameters are identical. It ensures the validity and reproducibility of our conclusions. The MoO₃/Ag or Ag electrodes were deposited on glass by thermal evaporation at a vacuum pressure $< 5 \times 10^{-4}$ Pa, with an evaporation rate of 0.02 nm/s and 0.1 nm/s for MoO₃ and Ag, respectively. The substrates during deposition were at room temperature. The thicknesses and evaporation rates of MoO₃, Ag and Al were estimated in situ with a calibrated quartz crystal monitor. Since the 2 nm thick MoO₃ layer is very thin, neither smooth nor closed, the thickness given here had to be a nominal value obtained by the monitor, representing the amount of MoO₃ on the sample.

The sheet resistances of these MoO₃/Ag or Ag electrodes were measured by using a four-point probe setup. The spectral transmission and reflectance were recorded by using a UV-vis-NIR spectrophotometer (Lambda 950, Perkin Elmer). Since transmission was measured relative to air, the reflection of the glass substrate was included. The atomic force microscopy (AFM) and scanning electron microscopy (SEM) images were taken by a Bruker Dimension Icon atomic force microscope in tapping mode and a JEOL JSM-6700F field emission scanning electron microscope, respectively.

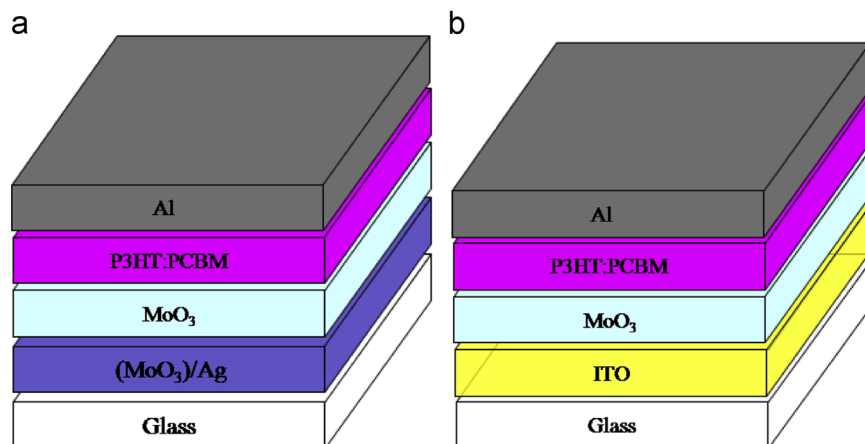


Fig. 1. Stack design of OSCs based on (a) the Ag (or MoO₃/Ag) electrodes and (b) the ITO electrode. Bottom illumination from the glass substrate side is applied for characterization of both sample types.

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