



Improved performance for polymer solar cells using CTAB-modified MoO₃ as an anode buffer layer



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ABSTRACT

We report a method to prepare molybdenum oxide thin films using the stable cetrinium bromide-modified MoO₃ (CTAB-modified MoO₃) precursor solution. The effect of thermal treatment on the optical, crystalline, morphologic properties and surface wettability is greatly obvious because of the pyrolytic decomposition of CTAB. The highly efficient and stable polymer solar cells (PSCs) have been also fabricated by adopting CTAB-modified MoO₃ nanocomposite films as the novel and universal anode buffer layers (ABLs). The P3HT:ICBA based solar cells with CTAB-modified MoO₃ at 200 °C heat treatment exhibit an best power conversion efficiency (PCE) of 5.80% with long-term stability, and the PTB7:PC₇₁BM based solar cells with CTAB-modified MoO₃ at 200 °C heat treatment show an best PCE of 8.34% with long-term stability, which are higher than that of the corresponding devices with PEDOT:PSS films. The improvement in device performance is mainly due to the agreeable electrical properties and enhanced charge extraction of the CTAB-modified MoO₃ ABLs, and CTAB modification to the MoO₃ surface can effectively passivate its surface traps, suppress the recombination loss of carriers. The surface of CTAB-modified MoO₃ films is also more hydrophobic than that of the PEDOT:PSS. In a word, these indicate the CTAB-modified MoO₃ films may be used as a novel and generally applicable hole transport layer for high-efficiency and ambient stable polymer solar cells.

1. Introduction

Polymer solar cells (PSCs) have attracted much attention due to their potential applications as clean energy sources and the possibility of their large-area fabrication [1–4]. There has been a rapid improvement in the power conversion efficiencies (PCEs) of PSCs, 10% or more of PCE has been achieved and reported because of tremendous advancements being made in the development of efficient conjugated materials including polymers and oligomers [5–8], devices structure [9,10], and electrode interface engineering [5,11,12]. Charge collection at the electrode interfaces is considered to be one of the most crucial factors to create efficient devices. To further increase the efficiency and the stability, a crucial aspect is engineering of the interface between the electrodes and the active layers by using electron and/or hole injection layers [13–16].

PEDOT:PSS has been widely used as ABLs by virtue of its good electrical properties and excellent solution processability, but its acidic and hygroscopic nature are detrimental to the long-term stability of organic solar cells [17,18]. Therefore, the search for new ABLs materials to replace PEDOT:PSS is an important research area. Especially,

solution-processed high work function transition metal oxides such as MoO₃ [19–22], V₂O₅ [23,24], NiO [25,26], and WO₃ [27,28], have been widely investigated and considered as promising alternatives to PEDOT:PSS due to their favorable band structures, and compatibility with large-area solution processing technologies. Devices with such metal oxide ABLs have been reported to show comparable efficiency and increased stability over the ones using PEDOT:PSS [29,30].

Among the above transition metal oxide semiconductors, solution-processed molybdenum oxide has drawn great attention as an interfacial layer for high-performance PSCs owing to its environmental stability, transparency in the range of visible light, non-toxicity and deep-lying electronic states [31–35]. Liu et al. spincoated an acidified aqueous ammonium heptamolybdate (NH₄)₆Mo₇O₂₄·H₂O precursor solution on indium tin oxide (ITO) and heated at 160 °C, to obtain a discontinuous coverage of MoO₃ nanoparticles on the ITO surface [36]. Meyer et al. reported on MoO₃ films spin-coated from a suspension of MoO₃ nanoparticles [37], in which the aggregation of nanoparticles led to excessive film roughness and significantly thicker films were required to prevent leakage in devices. Giroto et al. and Yang et al. developed solution-processed MoO₃ as a hole injection layer using

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the sol-gel technique [38,39]. However, this process requires relatively high temperatures (higher than 250 °C) to crystallize the MoO₃ film, rendering it incompatible with polymer substrates for future roll-to-roll manufacturing. Hammond et al. reported using Mo(CO)₃(EtCN)₃ as a precursor for fabricating continuous MoO_x [40]. However, the obtained MoO_x required O₂ plasma treatment, and the precursor was oxygen-sensitive. It's incompatible with high throughput and scale-up technologies in an ambient atmosphere. Bis (2,4-pentanedionato) molybdenum (VI) dioxide (MoO₂(acac)₂) is another promising precursor for preparing MoO_x [41]. Very recently, Zilberberg et al. and Jasieniak et al. reported sol-gel processed MoO_x from this precursor for PSCs application [42,43]. However, the obtained MoO_x layer must be maintained at ambient conditions for a long time to hydrolyze or age the sol-gel solution for a couple of days. These procedures are time-consuming and incompatible with high throughput and scale-up technologies.

Here, we present a facile synthetic route to prepare the smooth and dense CTAB-modified MoO₃ layers with the good optical properties and proper work function from the CTAB-modified MoO₃ precursor solution. And the CTAB-modified MoO₃ nanocomposite films are used as a novel class of ABLs for improving charge-collection. The CTAB-modified MoO₃ nanocomposite films offer some advantages of (1) facile solution processability (e.g. spin-coating and roll to roll printing); (2) low-cost and environment friendly raw materials; (3) good interfacial characteristics and optoelectronic properties for PSCs. Device performance based on the nanocomposite ABLs is found to be much better than that based on the pure PEDOT:PSS ABLs. And CTAB modification can effectively passivate MoO₃ surface traps, suppress the recombination loss of carriers. By incorporating the CTAB-modified MoO₃ thin films heated at 200 °C as ABLs, the devices with conjugated polymer:fullerene as the bulk heterojunction (BHJ) active layer exhibit greatly improved PCEs and long-term stability. These results indicate the CTAB-modified MoO₃ films can be a generally applicable hole transporting layer for ambient stable and high-efficiency polymer solar cells.

2. Experimental section

Ammonium molybdate (99.98%, ACS reagent), chlorobenzene (99.9%, for HPLC) and CTAB (99%, Product No. H108983) obtained from Sigma-Aldrich Inc. and Aladdin (China) were used as received without further purification. Regioregular electron-donor poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-(2-ethylhexyl) carbonyl] thieno[3,4-b] thiophenediyl]] (PTB7, Product No. LT-S9050), poly(3-hexylthiophene) (P3HT, Product No. LT-S909) and acceptor [6,6]-phenyl C₇₁-butyric acid methyl ester (PC₇₁BM, Product No. LT-S905), indene-C₆₀ bisadduct (ICBA, Product No. LT-S9030) were purchased from Luminescent Technology, Inc. and the PEDOT: PSS (Clevios 4083) solution was purchased from Starck. And all these materials were used as received. The chemical structures of materials used in the PSC devices are shown in Fig. 1(a).

2.1. Preparation of CTAB-MoO₃ nanocomposite films

Solution-derived CTAB-modified MoO₃ nanocomposite films were prepared by a facile sol-gel strategy on indium tin oxide (ITO) substrates (15 mm × 15 mm × 1 mm, CSG Holding Co. Ltd., Shenzhen, China). To prepare CTAB-modified MoO₃ precursor solutions, at first 34.6 mg of ammonium molybdate was firstly dissolved in 20 ml deionized water to obtain the solution A with 30 min continuous stirring at room temperature (RT). Then the CTAB solution was prepared by blending CTAB (18.22 mg) and deionized water (2 ml). The CTAB solution was added dropwise to the solution A under stirring to get the solution B. After adjusting the pH of the solution B to about 3 by the hydrochloric acid solution and followed stirring for 30 min, the sample bottles were maintained at RT for a while to get milky white CTAB-

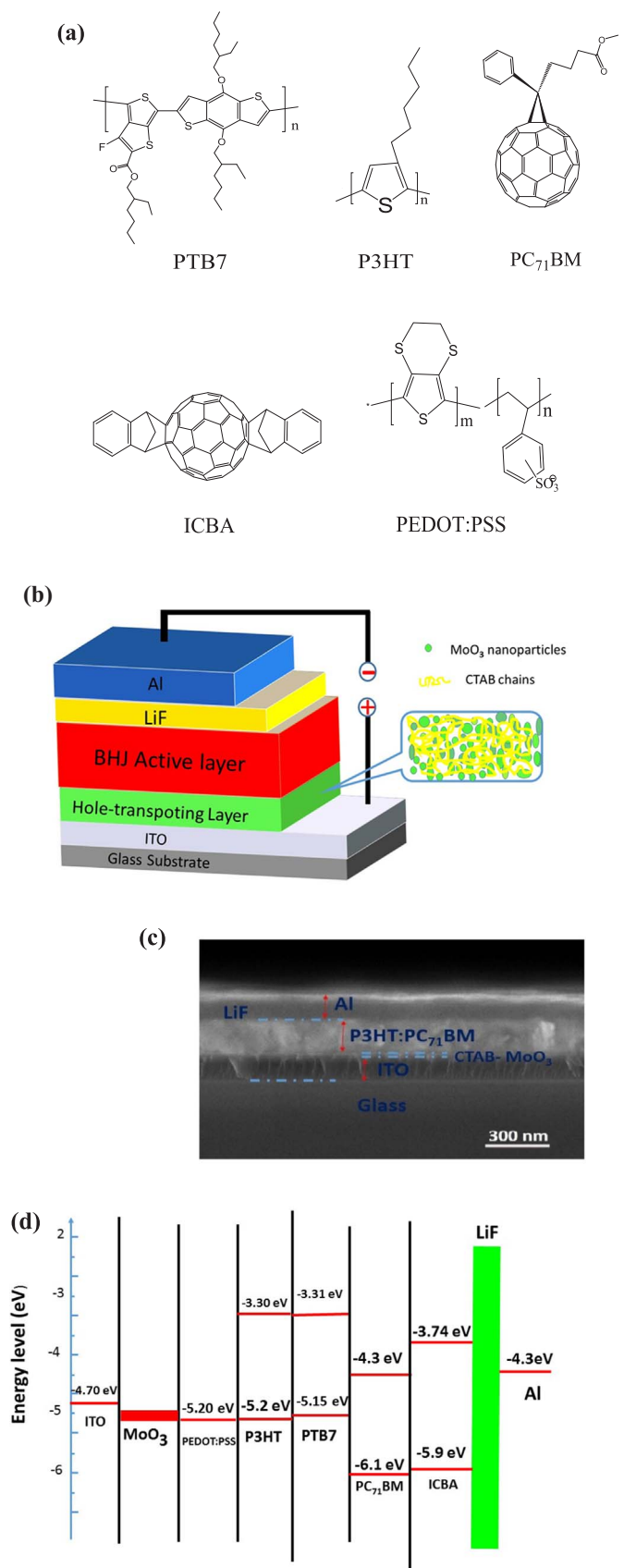


Fig. 1. (a) The molecular structures of PTB7, P3HT, PC₇₁BM, ICBA and PEDOT:PSS, (b) Scheme of the PSC devices structure used in this study, (c) SEM images of the active layers and cross section of the final device, (d) energy level diagram of materials used in the PSC devices [9,44,45].

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