Nano Energy 41 (2017) 681–686

Contents lists available at ScienceDirect

Nano Energy

journal homepage: www.elsevier.com/locate/nanoen

Communication

Very high hole drift mobility in neat and doped molecular thin films for normal and inverted perovskite solar cells

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ARTICLE INFO

Keywords:
Perovskite solar cells
Vacuum depositions
Hole transporting layers
Hole mobilities

ABSTRACT

Hole transporting layers (HTLs) have critical roles in the performance of perovskite solar cells. In this study, films of the vacuum-deposited small molecule of Tris[4-(5-phenylthiophene-2-yl)phenyl]amine (TPTPA) are demonstrated as efficient HTL materials for perovskite solar cells. The horizontal molecular stacking of the TPTPA neat film leads to the exceptionally high hole drift mobility of 7×10^{-3} $cm^2 V^{-1} s^{-1}$, among the highest values observed in the organic molecular neat films. By doping with MoO_3, a clear charge transfer state is formed and the hole mobility further increases to 3.6×10^{-2} $cm^2 V^{-1} s^{-1}$. Both normal- and inverted-structured perovskite solar cells utilizing these HTLs are prepared and optimized; they show high power conversion efficiencies of 17–18%.

1. Introduction

Recently, perovskite photovoltaics have gained great attention owing to their low-temperature operation, low-energy-consumption production and high efficiencies [1]. The device architectures have evolved from mesoporous structures to layer-by-layer thin-film stacks. In thin-film type perovskite solar cells, the perovskite active layer is sandwiched between the hole transporting layer (HTL) and electron transporting layer (ETL). Because the first perovskite solar cell used an ETL near the substrate, the cathode/ETL/perovskite/HTL/anode structure thus is called the normal structure, while the anode/HTL/perovskite/ETL/cathode counterpart is called the inverted architecture. Both normal and inverted perovskite solar cells require the HTL and ETL to promote their highest performance potentials. Extensive efforts have been made in the development of good inorganic and organic ETL and HTL materials to pair with perovskite active layers [2–4]. Fullerene derivatives [5–7], TiO_2 [8–10], and SnO_2 [11–13] are popular ETLs for high-performance perovskite solar cells. Meanwhile, inorganic NiO [14–16], small-molecule 2,2′,7,7′-Tetrakis-(N,N-di-4-methoxyphenylamino)-9,9′-spirobifluorene (Spiro-OMeTAD) [17–19], polymeric Poly(4-butylphenyl-diphenyl-amine) (poly-TPD) [20,21], and Poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) [22–24] are currently among the most common HTLs nowadays. To cooperate with high-mobility perovskite active layers, HTLs and ETLs should also possess high mobilities. For some organic thin films with low hole drift mobilities, such as Spiro-OMeTAD (1×10^{-5} $cm^2 V^{-1} s^{-1}$) [17,25], conductive doping with Li salts and oxygen doping are usually utilized to increase their hole mobilities [26,27].

In this study, we report a new vacuum-deposited thin film of the small molecule Tris[4-(5-phenylthiophen-2-yl)phenyl]amine (TPTPA) as an efficient HTL material for perovskite solar cells. TPTPA has previously been used in small-molecule organic solar cells as active layers and HTLs [28–31]. The TPTPA neat films showed the high intrinsic hole drift mobility of $\sim 7 \times 10^{-3}$ $cm^2 V^{-1} s^{-1}$. This is attributed to the highly horizontally oriented molecules and transition dipoles, which facilitate more compact hole transporting moieties and shorter hole hopping distances. By doping with MoO_3, the preferable horizontal molecular orientation was disturbed; however, a clear charge-transfer state was formed and the hole mobility further increased to 3.6×10^{-2} $cm^2 V^{-1} s^{-1}$. The neat and doped TPTPA thin films were then utilized in perovskite solar cells with both normal and inverted device structures. In the inverted device, thin TPTPA neat films of 10 nm were used and the devices showed the promising power conversion efficiency (PCE) of 17.1% with an open-circuit voltage (V_{OC}) of 1.07 V, short-circuit current density (J_{SC}) of 22.70 mA cm^{-2}, and fill factor (FF) of 70.4%. In the normal-architecture device, thicker HTLs are usually necessary to prevent the direct contact between the rough perovskite active layer and metal cathode. We found that the very high-mobility MoO_3 doped TPTPA thin films permitted high stabilized PCEs reaching 18.0%.

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http://dx.doi.org/10.1016/j.nanoen.2017.10.026
Received 12 August 2017; Received in revised form 26 September 2017; Accepted 9 October 2017
Available online 13 October 2017
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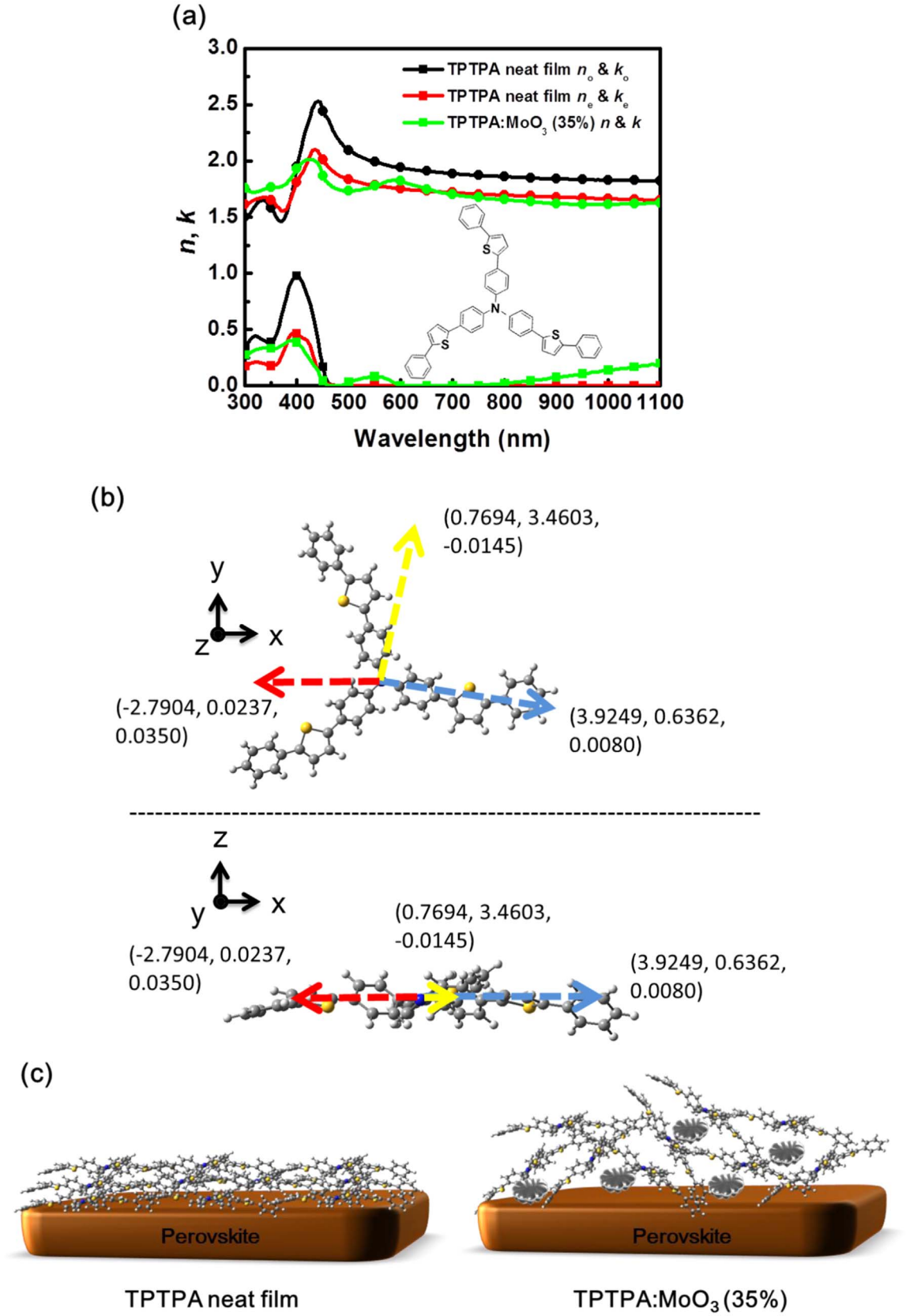

Fig. 1. (a) Optical constants of the TPTPA neat film and the TPTPA:MoO_3 (35%) thin film. Inset: molecular structure of TPTPA. (b) DFT simulations of TPTPA molecule with three highest-amplitude transition dipole moments. (c) Schematics of TPTPA and TPTPA:MoO_3 (35%) deposited on the perovskite.

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