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# Effect of open-circuit voltage in organic solar cells based on various electron donor materials by inserting molybdenum trioxide anode buffer layer

Jhong-Ciao Ke<sup>a</sup>, Yeong-Her Wang<sup>a</sup>, Kan-Lin Chen<sup>b</sup>, Chien-Jung Huang<sup>c,\*</sup>

<sup>a</sup> Institute of Microelectronics, Department of Electrical Engineering, National Cheng-Kung University, #1 University Rd., Tainan, Taiwan

<sup>b</sup> Department of Electronic Engineering, Fortune Institute of Technology, Kaohsiung, Taiwan

<sup>c</sup> Department of Applied Physics, National University of Kaohsiung, Nanzih, Kaohsiung, Taiwan

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## ABSTRACT

The effect of inserting a molybdenum oxide ( $\text{MoO}_3$ ) anode buffer layer into organic solar cells based on various electron donor materials was studied. The device was composed of an indium tin oxide (ITO)/ $\text{MoO}_3$ /donor material/ $\text{C}_{60}$ /bathocuproine/aluminum structure, and the donor materials were boron subphthalocyanine chloride (SubPc), rubrene, 2-[(7-{4-[N,N-bis(4-methylphenyl)amino]phenyl}-2,1,3-benzothiadiazol-4-yl)methylene]propanedinitrile (DTDCPB) and copper phthalocyanine (CuPc). Inserting a  $\text{MoO}_3$  buffer layer between ITO and donor layer resulted in the efficiency of device improvement from 1.04% to 2.49%, from 0.27% to 1.01%, and from 0.13% to 1.12% for the donor material of SubPc, rubrene, and DTDCPB, respectively. The result was ascribed to a greatly enhancement of open-circuit voltage ( $V_{oc}$ ), which originated from the work function improvement of ITO by covering  $\text{MoO}_3$  layer. However, the function of  $\text{MoO}_3$  was not evident in the device using CuPc as donor material. The interaction between  $\text{MoO}_3$  and CuPc was detected using UV-visible absorption and X-ray photoelectron spectroscopy. The electron transfer between  $\text{MoO}_3$  and CuPc caused the formation of an interface state at the  $\text{MoO}_3$ /CuPc interface, resulting in Fermi-level pinning at the interface. Consequently, the efficiency of the CuPc/ $\text{C}_{60}$  heterojunction device cannot be improved by inserting a  $\text{MoO}_3$  anode buffer layer.

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## 1. Introduction

Organic solar cells (OSCs) have received a considerable amount of attention because of their potential applications, including flexible substrate, lightweight, low-cost, and mass-production [1–5]. Many approaches have been proposed for improving the performance of OSCs, such as inserting a buffer layer between the active layer and the electrode and using various structure configurations and organic materials. Numerous novel organic materials have recently been developed to increase the range of the solar spectrum absorbed by OSCs, which have achieved an efficiency of 10% [6]. High-efficiency OSCs generally require a high open-circuit voltage ( $V_{oc}$ ), which can be achieved by using anode buffer layer. To date, many anode buffer layers have been reported, such as poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) [7,8], molybdenum oxide ( $\text{MoO}_3$ ) [7,9], vanadium oxide ( $\text{V}_2\text{O}_5$ ) [7,10], polyaniline (PANI) [11], tungsten oxide ( $\text{WO}_3$ ) [12], and organic materials [13]. The conventional used anode buffer layers in OSCs to improve the  $V_{oc}$  of device are PEDOT:PSS

and  $\text{MoO}_3$ , respectively. Small-molecule OSCs are generally fabricated using  $\text{MoO}_3$ , and polymer OSCs are generally fabricated using PEDOT:PSS. Both materials are widely used in optoelectronic device applications because they can improve the work function of indium tin oxide (ITO) [14,15], enhancing the  $V_{oc}$  of the devices. In addition, the anode buffer layer of OSCs is designed such that the energy level is matched with the HOMO of the donor to facilitate hole extraction from the donor to the electrode. In theorem, the maximal value of the  $V_{oc}$  is associated with the energy level difference between the highest occupied molecular orbital of the donor ( $\text{HOMO}_D$ ) and the lowest unoccupied molecular orbital of the acceptor ( $\text{LUMO}_A$ ), but in fact the  $V_{oc}$  of a device is determined according to the difference in the work function of the electrodes [16]. However, the variation of  $V_{oc}$  in OSCs cannot be fully included from the above rules, indicating that the origin of  $V_{oc}$  in OSCs needs to be further investigated.

In this study, the  $\text{MoO}_3$  anode buffer layer was applied to small-molecule OSCs fabricated using various donor materials, and the performance in devices was analyzed especially in the variation of  $V_{oc}$ . Although the efficiency of devices is low due to the double-heterojunction structure, it can avoid disorder of bulk heterojunction structure and provide precise and stable parameters of device, which is helpful for the fundamental research. The  $V_{oc}$  of devices

\* Corresponding author. Tel.: +886 7 5919475; fax: +886 7 5919357.

E-mail address: [chien@nuk.edu.tw](mailto:chien@nuk.edu.tw) (C.-J. Huang).

using boron subphthalocyanine chloride (SubPc), rubrene, and 2-[(7-{4-[N,N-bis(4-methylphenyl)amino]phenyl}-2,1,3-benzothiadiazol-4-yl)methylene]propanedinitrile (DTDCBP) as a donor was greatly improved through insertion of MoO<sub>3</sub> layer. The result was attributed to the fact that the MoO<sub>3</sub> can improve the work function of ITO, and it was found that the carrier recombination have a great influence on the variation of V<sub>oc</sub>. A detail analysis of relation between the work function and the V<sub>oc</sub> was shown in the article. In contrast, the V<sub>oc</sub> of the copper phthalocyanine (CuPc)/C<sub>60</sub> heterojunction device was approximately the same irrespective of whether a MoO<sub>3</sub> buffer layer was inserted in the device. The function of MoO<sub>3</sub> was not evident in the CuPc/C<sub>60</sub> heterojunction device, which might result from the Fermi-level pinning. To elucidate this phenomenon, UV–visible (UV–vis) absorption and X-ray photoelectron spectroscopy (XPS) of CuPc and SubPc were performed to determine whether the V<sub>oc</sub> of a device is associated with the interaction between the donors and the MoO<sub>3</sub>. In this article, a detailed investigation of origin of V<sub>oc</sub> in OSCs is presented.

## 2. Experimental

The energy level diagrams of materials used in this study and the device structure were shown in Fig. 1. SubPc (99%, Lumintec), rubrene (99.5%, e-Ray), DTDCBP (97%, Aldrich), and CuPc (99%, Aldrich) were used as electron donors, respectively. C<sub>60</sub> (99.95%, Aldrich) and bathocuproine (99.5%, Aldrich) were used as an electron acceptor and exciton blocking layer, respectively. All of the materials were used without performing further sublimation. The OSCs were fabricated on glass substrates precoated with ITO. The ITO–glass substrates (AimCore Technology), which featured a sheet resistance of 7 Ω/sq, were sequentially cleaned by performing ultrasonic treatment in acetone, methanol, and deionized water for 5 min each and dried with a nitrogen blow. All organic materials in the OSCs and aluminum (Al) were deposited by vacuum thermal evaporation below a pressure of 4.8 × 10<sup>−6</sup> Torr. The deposition rate of the organic materials was approximately 0.02–0.04 nm/s, and Al (used as a cathode) was deposited through a shadow mask, yielding an active area of 6 mm<sup>2</sup> at a deposition

rate of 0.2–0.3 nm/s. The deposition rate and film thickness were monitored using a quartz crystal oscillator.

The current density–voltage (J–V) characteristics for OSCs were measured using a power source meter (Keithley 2400) under an illumination of 100 mW/cm<sup>2</sup> produced by an AM1.5 G sun simulator (Oriel 96000 150 W Xe lamp, Newport). The light intensity was calibrated using a reference solar cell and meter (Oriel 91150, Newport). The OSCs were encapsulated before executing the measurement of J–V characteristics to ensure the reliability of parameters. The ITO substrates were cleaned by standard procedure and then transferred to the vacuum chamber to deposit MoO<sub>3</sub> layer. After thermal deposition, the sample of ITO/MoO<sub>3</sub> was exposed in the air shortly before introduced into an ultra high vacuum chamber for ultraviolet photoelectron spectroscopy (UPS) measurement (Sigma Probe, Thermo VG-Scientific). The work function of ITO and ITO/MoO<sub>3</sub> were performed by using monochromatized He I (21.2 eV) discharge lamp. The spectrum was taken for −5 V sample bias to separate the sample and the secondary edge for the analyzer. For atomic force microscope (AFM) measurement (XE-70, Park Systems), the surface morphologies were operated in non-contact mode in air. The absorption spectra of the films were measured using a UV–Vis–near-infrared spectrophotometer (UV3900, Hitachi) in the 300–800-nm wavelength range. The external quantum efficiency (EQE) measurements were carried out by using Solar Cell QE/IPCE measurement system (QE3000, TEO). The EQE spectra were performed with a Stanford SR830 lock-in amplifier under monochromatic illumination at a chopping frequency of 185 Hz. The J<sub>sc</sub> of devices derived from the integrated EQE spectrum are within few percent compared with the J<sub>sc</sub> value measured under simulated AM 1.5 G solar illumination at 100 mW/cm<sup>2</sup>. The films of CuPc, SubPc, CuPc:MoO<sub>3</sub> and SubPc:MoO<sub>3</sub> were deposited on the glass substrates for the XPS measurement. The interface electron states of films were measured and analyzed using XPS instrument (JEOL, JAMP-9500F Auger Electron Spectroscopy) with Mg Kα radiation (1253.6 eV) as X-ray source.

## 3. Results and discussion

Figure 2(a) showed the UPS spectra of ITO and ITO/MoO<sub>3</sub>, respectively. The MoO<sub>3</sub> deposited on ITO cause a 0.65 eV energy shift of secondary electron cutoff towards to lower binding energy, implying that the work function increased 0.65 eV by depositing MoO<sub>3</sub> layer on ITO substrate [17]. The estimate work function of ITO and ITO/MoO<sub>3</sub> are 4.75 and 5.4 eV, respectively, which is consistent with literature [18]. Fig. 2(b) showed the three dimension AFM images of ITO and ITO/MoO<sub>3</sub>, respectively. The roughness of ITO decreased from 3.555 ± 0.045 nm to 2.735 ± 0.085 nm by covering the MoO<sub>3</sub> layer. As a result, the function of MoO<sub>3</sub> not only increased the work function but improved the surface roughness of ITO. Figs. 3(a) and (b) show the J–V characteristics of devices fabricated using various donor materials with and without a MoO<sub>3</sub> buffer layer, respectively. Table 1 listed the cell performance parameters of the devices. Inserting the MoO<sub>3</sub> buffer layer between ITO and donor layer resulted in a greatly enhancement of the V<sub>oc</sub>. The V<sub>oc</sub> of devices using SubPc, rubrene and DTDCBP as donor was separately increased from 0.41 to 1.03 V, from 0.22 to 0.68 V and from 0.14 to 0.66 V corresponding to the efficiency improved from 1.04% to 2.49%, from 0.27% to 1.01% and from 0.13% to 1.12%. The V<sub>oc</sub> enhancement was attributed to the work function of ITO improved by covering MoO<sub>3</sub> buffer layer (from 4.75 to 5.4 eV), increasing the difference of work function between two electrodes, and leading to improvement of the built-in electric field (E<sub>bi</sub>) in device. The large E<sub>bi</sub> of device can increase the collection efficiency of free carriers and decrease the recombination probability. Thus, the shunt resistance (R<sub>sh</sub>) of device

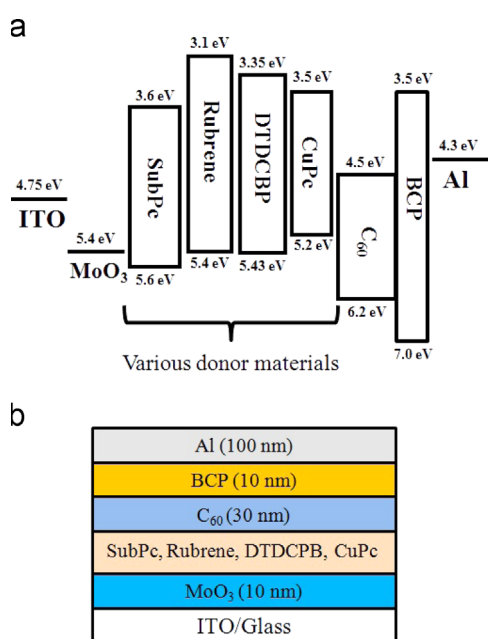


Fig. 1. (a) The schematic energy-level diagrams of different materials and (b) structure diagram of OSCs in this study.

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