

# Poly(3,4-ethylenedioxythiophene):Poly(styrenesulfonate) (PEDOT:PSS)- Molybdenum Oxide Composite Films As Hole Conductor For Efficient Planar Perovskite Solar Cells

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From the Figure S1, it can be seen that the MoO<sub>x</sub> nano-spots can be seen on the ITO substrate.  
Therefore, the deposited MoO<sub>x</sub> is not continuous film.

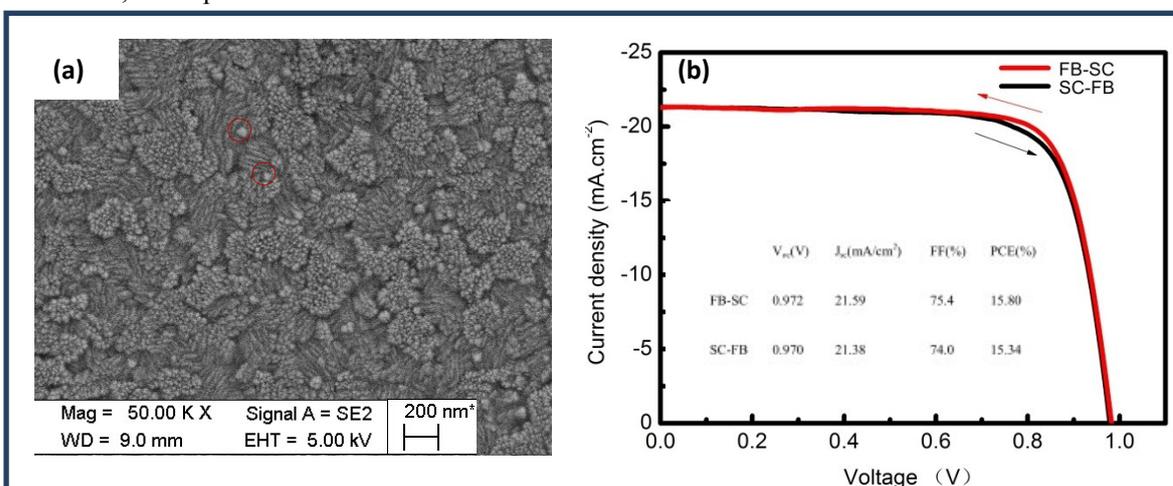


Figure S1 a)SEM image of MoO<sub>x</sub> aqueous solution coating on the ITO, b) obtained from forward bias to short circuit(FB-SC) and from short circuit to forward bias(SC-FB).

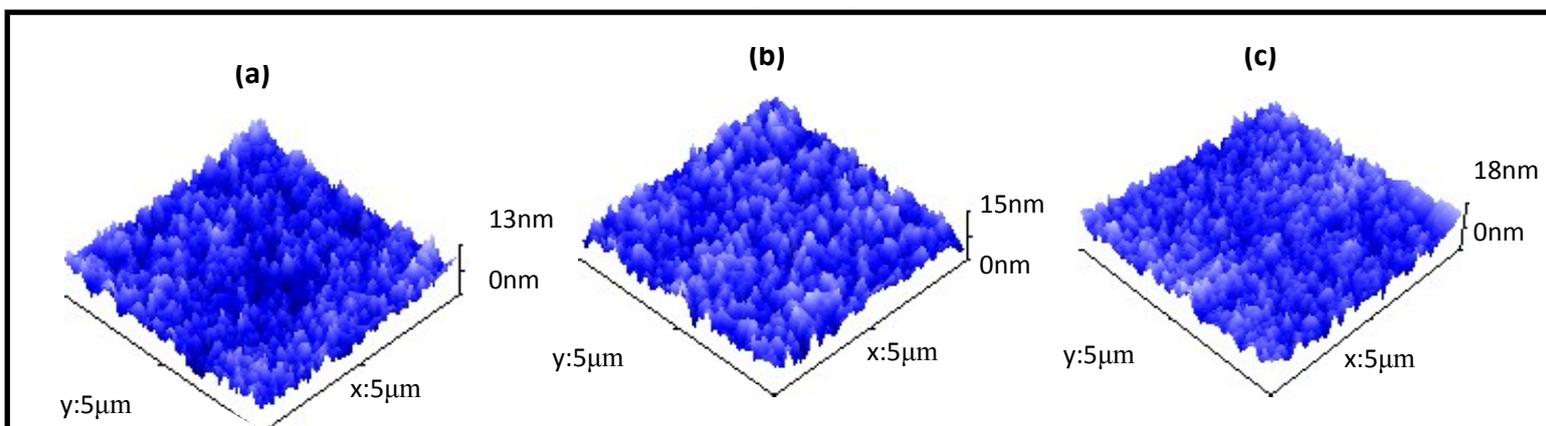


Figure S2 AFM surface morphology(5µm × 5µm) of (a) pristine PEDOT:PSS film,(b)PEDOT:PSS-MoO<sub>x</sub>(1mg/mg) composite film, c) PEDOT:PSS-MoO<sub>x</sub>(1.5mg/mg) composite film.

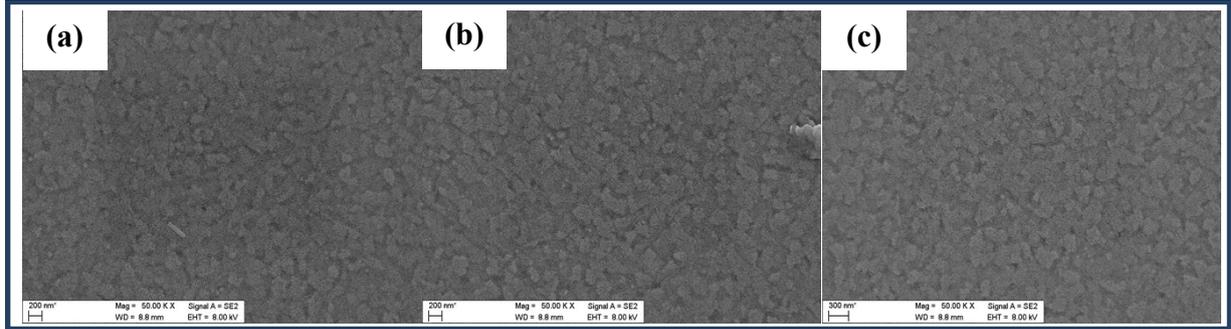


Figure S3 SEM images of PEDOT:PSS with different MoO<sub>x</sub> doping concentration a) 0mg/ml, b) 1mg/ml, c) 1.5mg/ml.

### MoO<sub>x</sub> characteristics

In order to investigate the chemical composition of the MoO<sub>x</sub> thin films using low-temperature solution processing, X-ray photoelectron spectrum (XPS) of the Mo 3d core level of MoO<sub>x</sub> film is shown in Figure 2a. The Mo 3d peak is deconvoluted to obtain the contributions of Mo<sup>5+</sup> (232.0 eV for Mo 3d<sub>5/2</sub>) and Mo<sup>6+</sup> (232.7 eV, 236.0 eV). Mo<sup>6+</sup> and Mo<sup>5+</sup> are found to be the major species present in the MoO<sub>x</sub>. In addition, Mo<sup>5+</sup> species are found to be 18.7 atomic ratios (%). Mo<sup>5+</sup> species are contributed to metal-like electrical properties of MoO<sub>x</sub>, which are beneficial for the collection of photo-generated charge carriers owing to the more favorable band structure and are finally able to increase J<sub>sc</sub>. To investigate the application of MoO<sub>x</sub> in the field of organic-inorganic hybrid solar cells, the ultraviolet photo-electron spectroscopy (UPS) measurements of a pristine PEDOT:PSS film and MoO<sub>x</sub> coated on glass substrate are carried out and the results are shown in Figure S4b. The work function of MoO<sub>x</sub> (5.12 eV) is higher than the pristine PEDOT:PSS (4.99 eV), which indicates the MoO<sub>x</sub> as hole conductor would result in a better energy alignment between ITO and perovskite active layer.

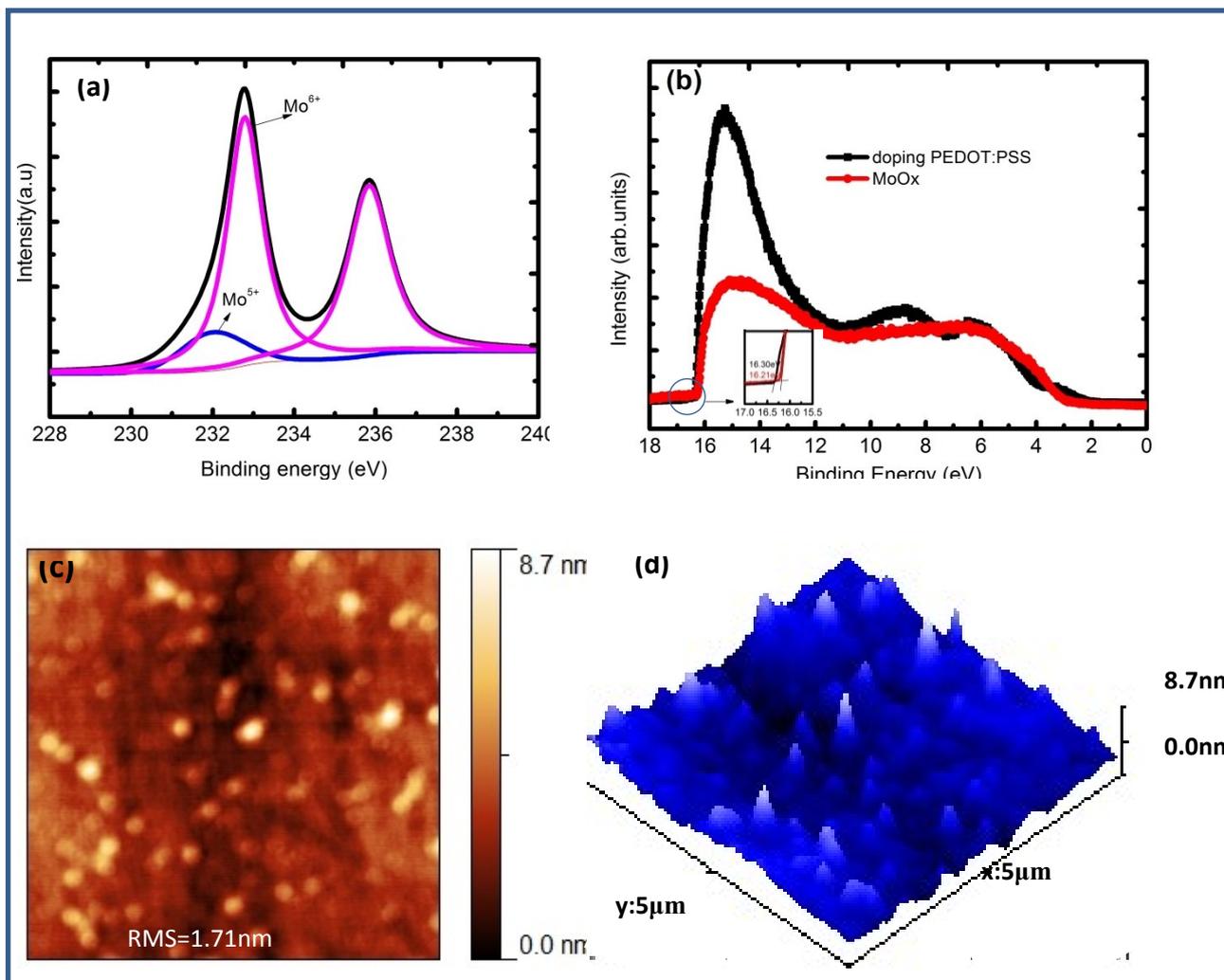


Figure.S4 a) XPS profiles for the MoO<sub>x</sub> layer. b) Their corresponding UPS spectra. c) AFM morphology (5 μm × 5 μm) of the MoO<sub>x</sub> layer. d) Three-dimensional AFM surface topographical image of the MoO<sub>x</sub> layer.

Figure S4c shows the surface topographical image of MoO<sub>x</sub> aqueous solution coated on glass by atomic force microscopy (AFM). The surface roughness values of the root mean square (RMS) are 1.71 nm for the whole scanned area (Figure 4Sc and d), the nano-scale spots form with heights ranging from 4 to 8.7 nm. This result indicates that the MoO<sub>x</sub> is presented as the nanoparticles and is not the continuous film on glass,

which is consistent with the SEM image in Supporting Information Figure S1. This is also the reason that the MoO<sub>x</sub> could not completely replace for the PEDOT:PSS as the hole conductor in PSCs although the MoO<sub>x</sub> has a higher work function than PEDOT:PSS. This is presumably caused by shorting associated with pinholes on the MoO<sub>x</sub> layer. However, based on a consideration that the MoO<sub>x</sub> could ensure the better growth of high quality perovskite films, and high work function which can provide sufficient charge dissociation and extraction, a perfect hole interfacial film for PSCs is expected by mixing PEDOT:PSS-MoO<sub>x</sub>.