Low-temperature, solution-processed molybdenum oxide hole-collection layer for organic photovoltaics†

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Electronic Supporting Information

X-ray Diffraction (XRD)

The pale bluish-white powder obtained from the TGA of $Mo(CO)_3(EtCN)_3$ in nitrogen (maximum temperature: 500 ˚C) was placed on a glass slide using some vacuum grease, and the XRD spectrum was measured in air (Figure S1). The XRD spectrum is consistent solely with that of crystalline MoO_3, with no signs of MoO_2 crystallites or other crystalline species present. In contrast, XRD of the dark brown powder obtained from TGA of $Mo(CO)_3(EtCN)_3$ in dry synthetic air (maximum temperature: 300 ˚C) showed no peaks whatsoever, indicative of an amorphous morphology.

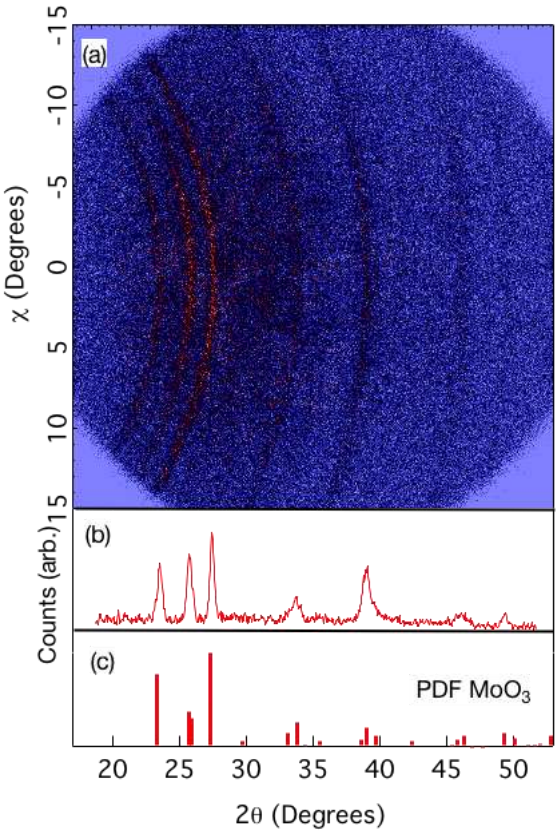

Figure S1. (a) 2D XRD raw image and (b) 2θ obtained by integrating spectrum in χ direction for the sample obtained from TGA of $Mo(CO)_3(EtCN)_3$ in nitrogen, with (c) reference powder diffraction file (PDF) MoO_3 peak positions

Variable angle spectroscopic ellipsometry (VASE)

VASE of MoO_x thin films coated on Eagle 2000 glass was performed to determine the film thickness, along with the optical constants (Figure S2). For MoO_x films deposited using the optimized conditions, with an anneal temperature of 120 ˚C and a 2 minute O_2-plasma treatment, the thickness was determined to be 4.9 nm, which is comparable to that generally used for thermally evaporated MoO_3 HCL layers.[1-3]

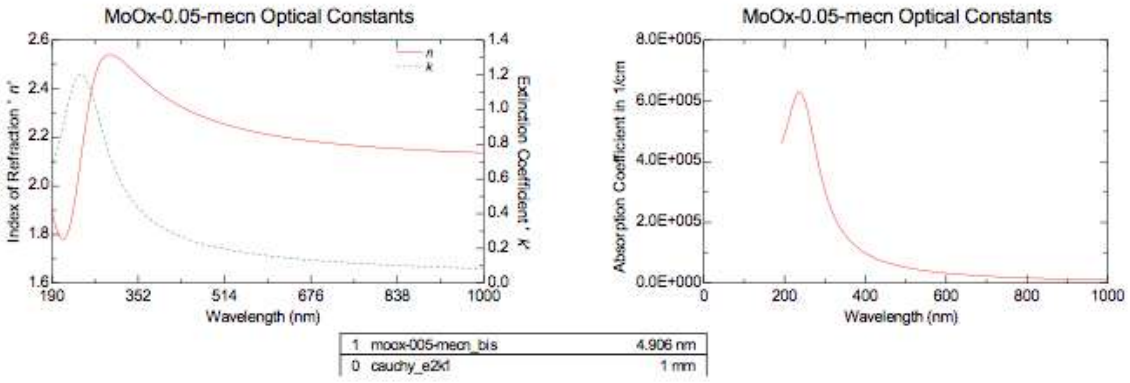

Figure S2. VASE determined optical constants and thickness of MoO_x thin film on Eagle 2000 glass substrate

Atomic Force Microscopy (AFM)

AFM was performed on cleaned, patterned ITO-coated glass substrates before and after coating with MoO_x following the optimized recipe (Figure S3). The root-mean-squared surface roughness (R_{rms}) values before and after coating, 3.79 nm and 3.67 nm, respectively, are nearly identical, suggesting a smooth and conformal coating that is consistent with an amorphous morphology.

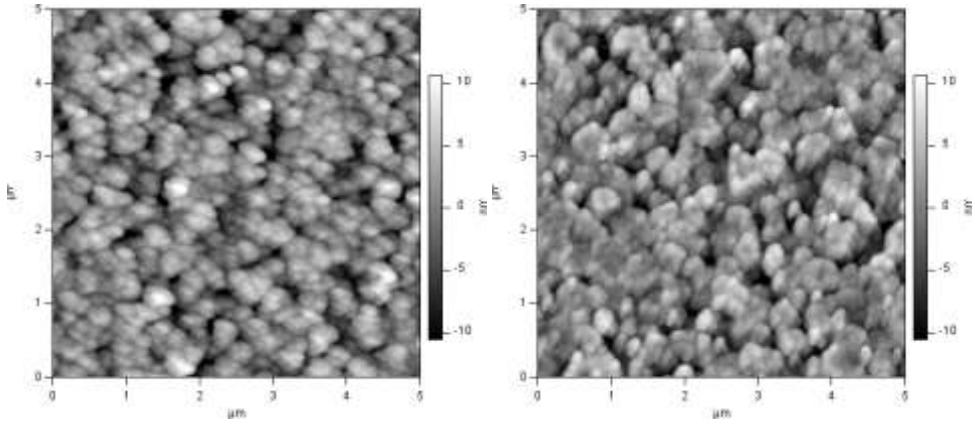

Figure S3. AFM of cleaned patterned ITO-coated glass before (left) and after (right) coating with MoO_x following the optimized recipe

OPV Device Optimization

The MoO_x HCL shows a wide processing window. We examined the effect of MoO_x annealing temperature, from 100 to 140 ˚C, on the performance of the HCL in conventional architecture P3HT:PCBM devices (Figure S4). Within this annealing range, the HCL performance remained identical, with devices yielding comparable power conversion efficiencies. We also explored the effect of different O_2 plasma durations (Figure S5). Varying the duration of the plasma treatment from 0.5-5 min appears to have minimal effect on the device performance. In contrast, varying the solution concentration and spin speed, and thus the film thickness, has a much greater effect on device performance (Figure S6). As MoO_x is quite resistive, the HCL thickness should be minimized to limit resistive losses. The film needs to be thick enough to form continuous films, however, in order to prevent large leakage currents (e.g. Figure S5: 0.05 M, 8000 rpm). The optimal performance was obtained from the 0.05 M solution, spun at 6000 rpm, which yields a thickness of ~5 nm, as determined by VASE, as described above.

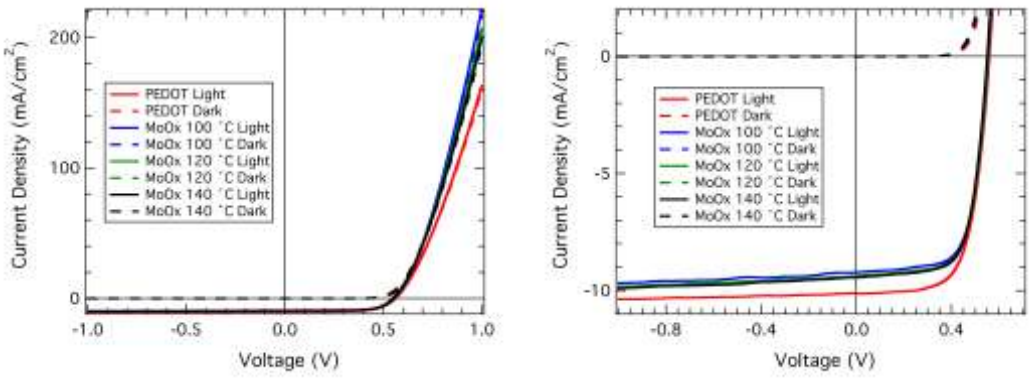

Figure S4. Representative *J-V* traces demonstrating the effect of MoO_x HCL thermal annealing on P3HT:PCBM BHJ device performance

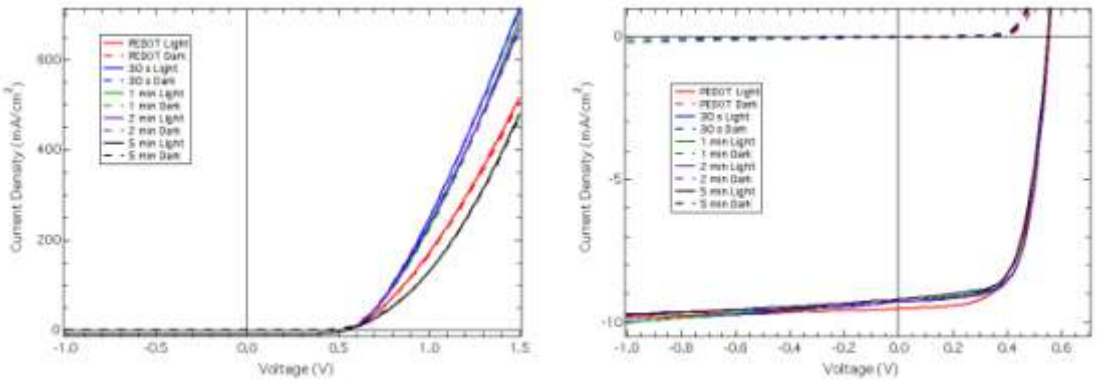

Figure S5. Representative *J-V* traces demonstrating the effect of MoO_x HCL O_2-plasma duration on P3HT:PCBM BHJ device performance

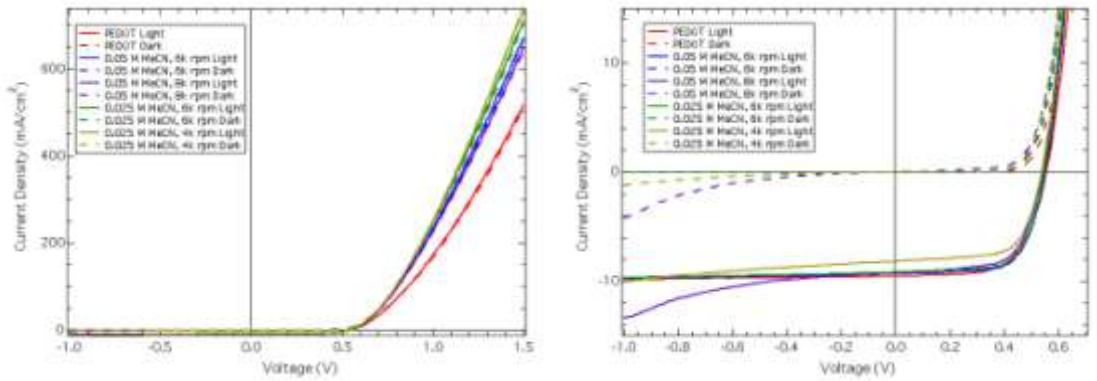

Figure S6. Representative *J-V* traces demonstrating the effect of $Mo(CO)_3(EtCN)_3$ solution concentration and spin speed on P3HT:PCBM BHJ device performance

References

1. V. Shrotriya, G. Li, Y. Yao, C.-W. Chu and Y. Yang, *Appl. Phys. Lett.*, 2006, 88, 073508.
2. F. Liu, S. Shao, X. Guo, Y. Zhao and Z. Xie, *Sol. Energy Mater. Sol. Cells*, 2010, 94, 842-845.
3. Y. Sun, C. J. Takacs, S. R. Cowan, J. H. Seo, X. Gong, A. Roy and A. J. Heeger, *Adv. Mater. (Weinheim, Ger.)*, 2011, 23, 2226-2230.