Few-layer MoS_2 Flakes as Hole-selective Layer for Solution-processed Hybrid Organic Hydrogen-evolving Photocathodes

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High-efficiency organic photocathodes, based on regioregular poly(3-hexylthiophene) and phenyl-C61-butyric acid methyl ester (rr-P3HT:PCBM) bulk heterojunction sandwiched between charge-selective layers, are emerging as efficient and low-cost devices for solar hydrogen production by water splitting.[1, 2] Nevertheless, stability issues of the materials used as hole-selective layers are hampering the realization of long-lasting photoelectrodes, pointing out the need to investigate novel and stable materials.[3] Here, we propose MoS_2 nano-flakes,[4,5] produced by Li-aided exfoliation of bulk counterpart, as efficient atomic-thick hole-selective layer for rr-P3HT:PCBM-based photocathodes. We carried out a p-type chemical doping to tune on-demand the MoS_2 Fermi levels, i.e. the work function (WF) values, of the MoS_2 films to higher values (from 4.6 eV of the pristine MoS_2 to 5.1 eV), in order to match the highest occupied molecular orbital (HOMO) level of the rr-P3HT, thus easing the hole collection at the electrode.[6] The as-prepared p-doped MoS_2-based photocathodes reached a photocurrent of 1.21 mA cm^{-2} at 0 V *vs.* RHE, a positive onset potential of 0.56 V *vs.* RHE and a power-saved figure of merit of 0.43%, showing a 6.1-fold increase with respect to pristine MoS_2-based photocathodes, under simulated 1 Sun illumination (at AM1.5G and 100 mW cm^{-2} illumination conditions). Operational activity of the photocathodes over time and under 1 Sun illumination revealed a progressive stabilization of the photocurrents at 0.49 mA cm^{-2} at 0 V *vs.* RHE. These results pave the way towards the exploitation of layered crystals as efficiency-boosters for scalable hybrid organic H_2-evolving photoelectrochemical cells.

References

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Figures

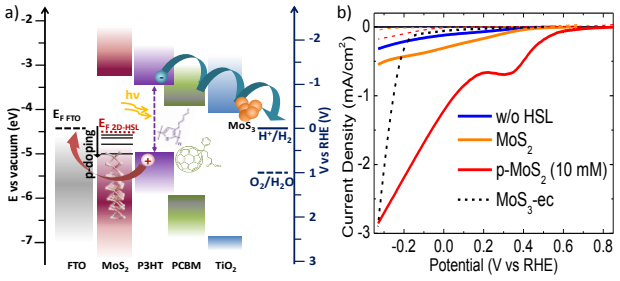

Figure 1: a) Typical materials energy band edges position. b) Photoelectrochemical characterization of the organic photocathodes without HSL (blue lines), with MoS_2 (orange lines) and p-doped MoS_2 (red lines) as HSL and in the dark (short dashed lines). The HER-electrocatalytic activity of MoS_3 (deposited directly on FTO) is also shown (short dashed black line).