Catalyst-Free Amorphous Silicon-Based Tandem Thin Film

Photocathode with High Photovoltage for Solar Water Splitting

Bofei Liu^{1, 2}, Zhonghua Jin^{1, 3}, Lisha Bai^{1, 2}, Junhui Liang¹, Qixing Zhang¹, Caichi Liu^{1, 3},

Ying Zhao^{1, 2}, and Xiaodan Zhang^{1, 2*}

¹ Institute of Photo Electronics thin Film Devices and Technology of Nankai University, Key Laboratory of

Photoelectronic Thin Film Devices and Technology, Tianjin 300071, P. R. China

²Collaborative Innovation Center of Chemical Science and Engineering (Tianjin), Tianjin 300072, People's

Republic of China

³Institute of Semiconductor Materials, Hebei University of Technology, Tianjin 300130, China



SUPPORTING INFORMATION



Catalytically active a-MoS₃ catalysts were deposited with a scalable wet chemical synthesis to act as hydrogen evolution catalysts in the proposed a-Si/a-SiGe tandem photocathode. To characterize the PEC catalytic activity of the a-MoS₃ catalysts in an overall water-splitting system, a wired water-splitting device comprising of four μ c-Si:H single-junction solar cells in series was deployed in a three-electrode configuration with a Pt counter

electrode and the immersed catalyst electrodes acting as the working electrodes. The four μ c-Si:H single-junction solar cells (V_{oc} =0.507 V) in series can provide an enough V_{oc} of 2.2 V for water splitting. A-MoS₃ catalysts were fabricated on a conductive FTO substrate. The *J*-*E* characteristics of the FTO supported a-MoS₃ and the referenced bare FTO and Pt electrodes under illumination are presented in Figure S2.



Figure S2. *J*-*E* characteristics of the FTO, FTO supported a-MoS₃, and Pt catalysts measured with a serial water-splitting device.

Obviously, the addition of a-MoS₃ catalysts on the FTO substrate can positively shift the onset potential E_{on} from 1.43 V to 2.0 V, indicating that large overpotential losses on the FTO/electrolyte can be partially compensated for. Although the photocurrents at the positive potentials for the FTO supported a-MoS₃ electrode are still lower than that of the Pt electrode, a nearly similar E_{on} value can be achieved for our a-MoS₃ catalysts in comparison to the Pt electrode.



Figure S3. EIS Nyquist plots of unprotected, p-a-SiC:H protected, and a-MoS₃ catalyzed unprotected a-Si/a-Si tandem photocathodes.



Figure S4. *J-t* curve of the unprotected a-Si/a-SiGe tandem photocathode measured at 0 V vs. RHE.