Supporting Information for "Electrophoretic deposition of transparent MoS_2/graphene nanosheet composite films as counter electrodes in dye-sensitized solar cells"

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Experimental details

***Synthesis of MoS_2/GNS*:** Graphene oxide (GO) was obtained via the oxidation of natural graphite powder using a modified Hummers method as reported.[S1] For the synthesis of MoS_2/GNS composites, the obtained 1 mg GO suspension was first added into 40 mL deionized water. Then, 0.6 g $Na_2MoO_4 \cdot 2H_2O$ (Acros, 99%) was added into the solution. After ultrasonication and stirring for 30 min, 1.0 M KOH was added to the solution until the pH value was adjusted to 6.5. After that, the mixture and 0.8 g thiourea (Acros, 99%) were dissolved in 80 mL deionized water, and further sonicated for another 30 min before transferred to a 150 mL Teflon-lined autoclave, and then heated in an oven at 240℃ for 24 h. The black precipitates were collected by centrifugation, washed with 1 M HCl (Acros, 99%), deionized water, and ethanol, and dried in a vacuum oven at 80℃ for 24 h. The preparation process of pristine MoS_2 and GNS was similar to that described above, but without the addition of GO and Na_2MoO_4, respectively.

***Preparation of CEs*:** The resultant MoS_2, GNS or MoS_2/GNS powders of 0.25 g were suspended in a 50 mL 1:1 mixture of acetone and ethanol by ultrasonication for 2 h.

For electrophoretic deposition of GNS or MoS_2/GNS on a FTO glass substrate, a cleaned FTO glass substrate (NSG, 13 Ω sq^{-1}) and a Pt sheet (4 cm^2) were used as a working and a counter electrode, respectively. These two electrodes were positioned in parallel with a distance of 0.5 cm apart from each other, and a voltage of 60 V was employed. The information regarding the deposition time verse the corresponding film thickness and transparency of MoS_2/GNS CE is illustrated in Fig. S6. In view of the transparency, the deposition time was set for 1 min in this study. Due to the difficulty in preparing MoS_2 onto FTO glass substrates by electrophoretic deposition, the MoS_2 suspension in the concentration of 5.0 mg mL^{-1} was drop-casted onto the cleaned FTO glass substrates to fabricate the MoS_2 CE. To assess the cell efficiency, sputtered Pt layer (100 nm) on a FTO glass substrate served as a Pt CE in this study.

***Assembly of DSCs*:** TiO_2 nanocrystalline photoanodes were prepared according to our previous report.[S2] The resulting TiO_2 photoanodes were soaked in an ethanol solution containing 0.3 mM N719 dye (Everlight Chemical Industry Co.) for 24 h. The sensitized TiO_2 photoanodes were rinsed with ethanol and subjected to be dried under a cool air flow. Then, the photoanodes were assembled with the various CEs via the thermoplastic hot-melt films (30 μm, Solaronix). The liquid redox electrolyte consisting of 1 M 1, 3-dimethylimidazolium iodide (Merck), 0.15 M iodine (J.T. Baker), 0.5M 4-tertpbutylpyridine (Aldrich), and 0.1 M guanidine thiocyanate (Aldrich) in 3-methoxypropionitrile (Acros) solution was injected into the cells through the pre-drilled holes on the CEs, which were further sealed after electrolyte injection.

***Characterization and measurements*:** The morphology and composition of MoS_2/GNS composite was characterized by using a JSM-7600F field emission

scanning electron microscope (HRTEM), Raman spectroscopy (RENISHAW in Via) and X-ray diffractometer (XRD6000, Shimadzu Corporation, Japan). The transmittance spectrum was performed with an Agilent 8453 UV-Visible diode array spectrophotometer. All electrochemical measurements were carried out using a computer-controlled potentiostate (CHI 6018). Cyclic voltammetragrams (CVs) were conducted with a scan rate of 10 mV s^{-1} in a three-electrode system, in which an as-prepared CE was taken as the working electrode in a three-electrode one-compartment cell, a 4 cm^2 Pt sheet auxiliary electrode and an Pt wire reference electrode in a 3-methoxypropionitrile solution consisting of 50 mM LiI, 10 mM I_2, and 500 mM $LiClO_4$. Electrochemical impedance spectroscopic (EIS) spectra were recorded with two identical electrodes, which were sealed with the aforementioned thermoplastic hot-melt Surlyn leaving an exposed area of 0.64 cm^2. The electrolyte used in the cell tests was also injected into the EIS symmetric cells. The EIS tests were conducted simulating open-circuit conditions within a frequency range of 0.1 Hz–10^5 Hz. A sinusoidal AC voltage signal varying by 5 mV was employed in all cases. The photovoltaic performance of the DSCs was performed using a computer-controlled Keithely 2400 sourcemeter under illumination by a Yamashita Denso YSS-150A solar simulator (AM 1.5, 100 mW·cm^{-2}).

References:

S1. C.-Y. Su, Y. Xu, W. Zhang, J. Zhao, X. Tang, C.-H. Tsai and L.-J. Li, *Chem. Mater.*, 2009, **21**, 5674.

S2. C. Y. Lin, J. Y. Lin, J. L. Lan, T. C. Wei and C. C. Wan, *Electrochem. Solid-State Lett.*, 2010, **13**, D77.

Supplementary Figures

Fig. S1 Photo image of the MoS_2/GNS suspension stored after one week.

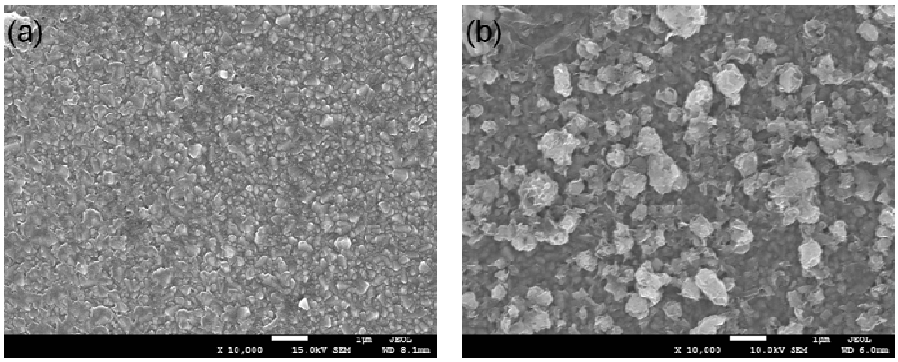

Fig. S2 (a) before and (b) after the electrophoretic deposition of MoS_2/GNS on a FTO substrate

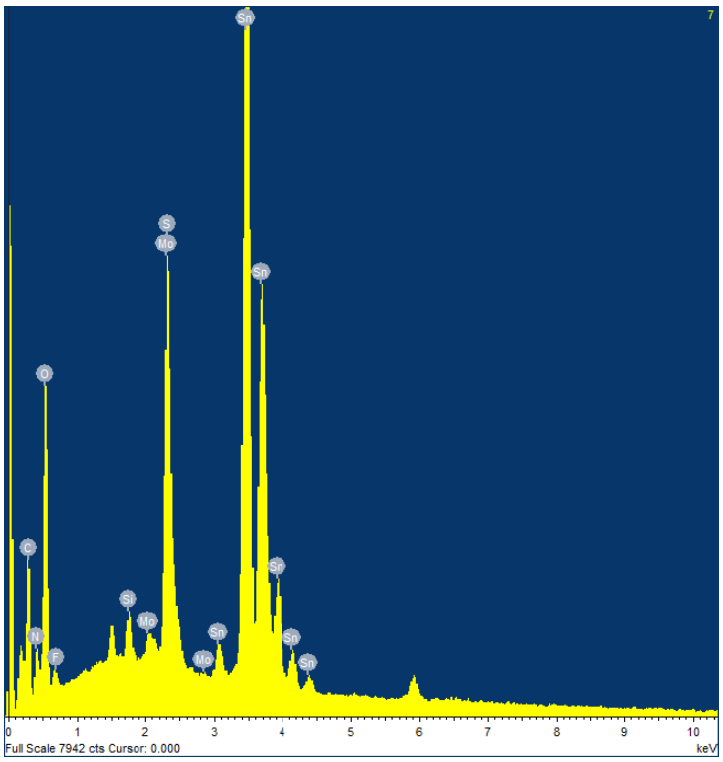

Fig. S3 X-ray energy dispersive spectroscopy of MoS_2/GNS CE.

Fig. 4S The photo image of the opaque MoS_2/graphene flake composite CE in Ref. 20.

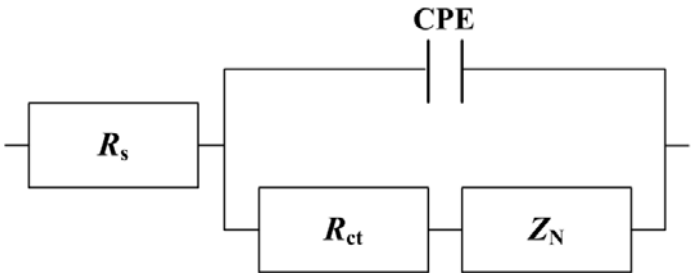

Fig. S5 Equivalent circuit used for fitting the EIS results of the symmetric cells.

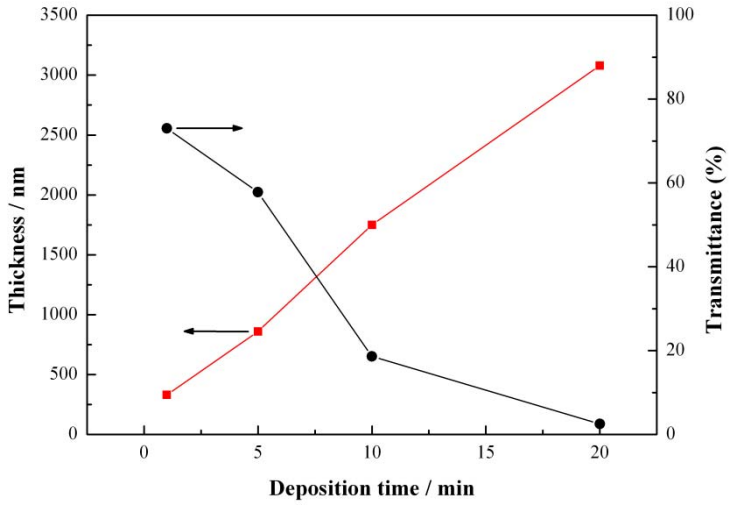

Fig. S6 The relationship between the deposition time and the corresponding film thickness and transparency of MoS_2/GNS CE. The transparency was measured at the wavelength of 550 nm. Each result was the average of three different MoS_2/GNS CEs.