Supporting Information

Turning Commercial Transition-Metal Oxides into Efficient Electrocatalysts *via* Facile Hydrogen Treatment

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Samples synthesis

The three commercial oxides (C-Fe₂O₃, C-In₂O₃ and C-SnO₂, Sinopharm) were carefully ground in advance, which was carried out in a ball mill using wet grinding method (using ethanol as solvent, 24 h, under rotation speed of 300 rpm). Then, the obtained powder of the three commercial oxides were annealed in hydrogen atmosphere (1 bar, 50 sccm H₂ flow) at 400 \degree for 60 min in a tube furnace, respectively. Finally, hydrogen-treated samples were synthesized.

Electrodes preparation¹

The procedure of preparing the TiO₂ photoanode, and the hydrogen-treated samples (H-Fe₂O₃, H-In₂O₃ and H-SnO₂) were described in detail as follow. Firstly, the blocking layer was deposited on the fluorine doped SnO₂ (FTO) conducting glass (NSG, 8 Ω /square) by dipping each FTO substrate in 40 mM TiCl₄ solution for 30 min at 70 °C. Then a 12 µm-thick layer of 20 nm-sized TiO₂ particles was loaded on the FTO by screen printer technique. After sintering at 125 °C, the obtained layer was coated with a 4-µm thick scattering layer of 200 nm-sized TiO₂ particles (HEPTACHROMA, DHS-NanoT200) followed by sintering at 500 °C. Then another TiCl4 treatment was carried in 40 mM TiCl4 for 30 min at 70 °C followed by sintering 500 $^{\circ}$ C for 30 min. After cooling to 80 $^{\circ}$ C, the whole TiO₂ films were immersed in a 5×10^{-4} M solution of N719 dye (Solaronix SA, Switzerland) in acetonitrile/tert-butyl alcohol (V/V=1/1) for 24 h and the working photoanodes were obtained. The H-Fe₂O₃, H-In₂O₃ and H-SnO₂ CEs were prepared on the FTO conductive glass by the doctor-blading method. The H-Fe₂O₃ powder mixed with 2% was hydroxypropylmethylcellulose (HPMC) in deionized water and absolute ethanol (V/V=5/3) solution to form a homogeneous paste. The FTO glass coated with H-Fe₂O₃ was then sintered in argon (1 bar, 50 sccm Ar flow) in a tube furnace at 450 °C for 30 min. The H-In₂O₃ and H-SnO₂ CEs electrode was prepared by the same procedure. The Pt-electrode was prepared by drop-casting 0.5 mM H₂PtCl₆/ethanol

solution on the clean FTO conductive glass. Subsequently, the FTO glass coated with Pt was sintered in a muffle furnace at 450 $\,^{\circ}$ C for 30 min and the CEs were obtained.

Fabrication of DSCs²

DSCs were assembled together with the dye-sensitized TiO₂ electrode and the CE by a 25- μ m thick hot-melt film (Surlyn 1702, DuPont) and sealed up by heating. The cell internal space was filled with typical liquid electrolytes using a vacuum pump. The liquid electrolyte was composed of 0.60 M 1-butyl-3-methylimidazolium iodide, 0.03 M I₂, 0.50 M 4-tert-butyl pyridine, and 0.10 M guanidinium thiocyanate with acetonitrile as the solvent. The assembled DSCs were used for the photocurrent-voltage test with an active area of 0.25 cm².

Materials characterization

The morphology and structure of the samples were characterized by field emission scanning electron microscopy (FESEM, HITACHI S4800). The crystal structure of the samples was determined by X-ray diffraction (XRD, Bruker D8 Advanced Diffractometer, Cu K α radiation, 40 kV). The chemical states of the elements in two samples were measured by X-ray photoelectron spectroscopy (XPS, Kratos Axis Ultra DLD), and the binding energy of the C 1s peak at 284.8 eV was taken as an internal standard. The current-voltage tests of DSCs were performed under one sun condition using a solar light simulator (Oriel, 91160, AM 1.5 globe). The power of the simulated light was calibrated to 100 mW cm⁻² using a Newport Oriel PV reference cell system (model 91150 V). The EIS experiments were measured with full cells in the dark by using an electrochemical workstation (Parstat 2273, Princeton). The frequency range of EIS experiments was from 100 mHz to 1 MHz with an AC modulation signal of 10 mV and bias DC voltage of 0.60 V. The curves were fitted by the Zview software. Cyclic voltammetry (CV) was conducted in a three-electrode system in an acetonitrile solution of 0.1 M LiClO4, 10 mM LiI, and 1 mM I₂ at a scan

rate of 20 mV s⁻¹ by using a BAS 100 B/W electrochemical analyzer. Pt served as a CE and the Ag/Ag⁺ couple

References:

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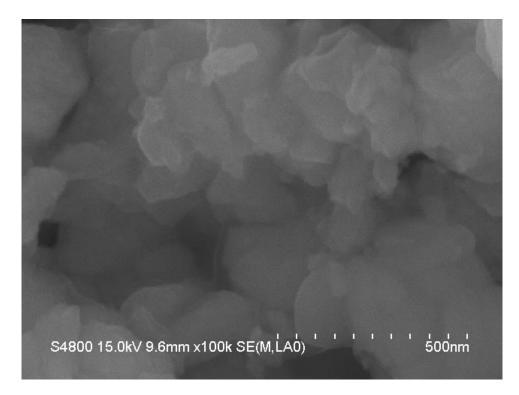


Fig. S1 SEM image of the untreated commercial Fe₂O₃ (C-Fe₂O₃).