

Support information:

Experimental Section

Preparation of TiO₂ photoelectrodes

A suspension of 3g P-25 powders (Degussa), 60ml ethanol, 0.1ml HNO₃(65%) was put into a rotary evaporator at 80 °C for 6h until the suspension becomes powdery (TiO₂/NO₃⁻). A coating paste was prepared by mixing 0.3g TiO₂/NO₃⁻, 0.12g polyvinylpyrrolidone in 1.5ml ethanol. ITO conductive glass and Ni mesh (MTI corporation) were served as the substrates for the photoanodes. Before use, Ti was coated onto both sides of Ni mesh with a deposition thickness of 500nm by DC Sputter (UniFilm Technology) at a rate of 50 nm/min. The ITO glass and Ti/Ni mesh were pretreated in 50mM TiCl₄ solution at 70 °C for half an hour. The as prepared TiO₂/NO₃⁻ paste was then cast onto ITO glasses (14µm) and Ti/Ni mesh (18µm, both sides), followed by calcination at 500 °C for 2h. The photoanodes were treated again in 50mM TiCl₄ solution at 70 °C for half an hour, followed by calcinations at 500 °C for 30min. After cooling down, TiO₂ on ITO conductive glass was immersed into 0.5mM N719/ethanol solution at room temperature for 24h. TiO₂ on Ti/Ni mesh was immersed into 0.2mM N749 with 20mM deoxycholic acid (DCA) in ethanol at room temperature for 24h. The counter electrodes were prepared by spreading a 0.5mM H₂PtCl₆ ethanol solution to ITO conductive glass, followed by calcinations at 400 °C for 20min. The PTFE separator films were kindly provided by Y-Carbon Inc.

Fabrication of dye sensitized solar cells

The single N719 cells were prepared by placing a Pt counter electrode against a TiO₂/ITO photoanode by using Kapton tapes as spacers. The single N749 cells were prepared by sequentially assembling ITO conductive glasses, N749 sensitized TiO₂ on Ti/Ni mesh, a PTFE

separator and a Pt counter electrode, and sealed with Kapton tapes. Notably, the Ti/Ni mesh was only partially coated with N749 sensitized TiO₂, so that the uncoated part was connected to ITO glasses by indium metal. The hybrid cell was fabricated by sequentially assembling a N719-sensitized TiO₂ on ITO glasses, N749-sensitized TiO₂ on Ti/Ni mesh, a PTFE separator and a Pt counter electrode together, and sealed with Kapton tapes as illustrated in Figure 1. The redox electrolyte used was an ionic liquid containing 0.60 M BMIM-I, 0.03 M I₂, 0.50 M TBP, and 0.10 M GTC in a mixture of acetonitrile and valeronitrile (v/v = 85/15) (No. ES-0004, io.li.tec, Germany). The electrolyte was injected into the cell by a narrow passage left by the Kapton tapes.

Characterizations

The photocurrent density-voltage (J-V) characteristics were measured using a Keithley model 2400 multisource meter. A solar simulator (SoLux Solar Simulator) was used to simulate sunlight for an illumination intensity of 100 mW/cm², calibrated with a Daystar Meter. The surface morphology of samples was examined on a LEO 1530 scanning electron microscopy with an acceleration voltage of 3kV. X-ray diffraction (XRD) analysis was performed on X'Pert PRO diffractometer (PANalytical BV) with Cu K α radiation. Electrochemical impedance spectroscopy was scanned under open voltage by electrochemical workstation (Zahner, Zennium). UV-Vis absorption spectra were measured by a UV-Vis spectrophotometer (SHIMADZU, UV-2600). The IPCE spectra were measured by QE/IPCE measurement kit from Newport Company.