Supporting Information

Dual-Functional ZnO Nanorod Aggregates as Scattering Layer in the Photoanode for Dye-Sensitized Solar Cells

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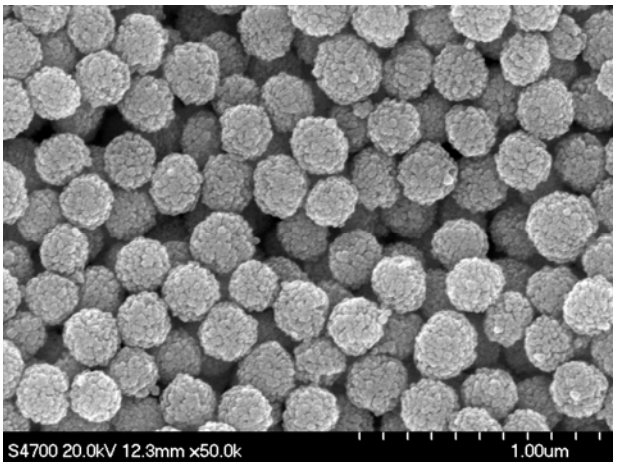

Fig. S1 FESEM image of MA-ZnO.

Experimental details

ZnO film Preparation: Fluorine-doped tin oxide-coated glass (FTO, Hartford, 14 Ω/sq, 80% transmittance) plates were utilized as the substrates. Before fabricating ZnO film, the FTO substrates were ultrasonically cleaned sequentially in HCl, acetone, ethanol and water each for 15 min and dried at the atmosphere of Ar. All the ZnO colloids including NC-ZnO, MA-ZnO and NA-ZnO were dispersed in ethanol and sealed for storing at room temperature, respectively. A few drops of the NC-ZnO colloids were firstly dipped onto the FTO substrates to construct the NC-ZnO underlayer film by means of drop-coat method following by calcination at 350 ^{o}C for 60 min to evaporate any residual organic solvent from ZnO surface. For fabricating a bilayer-structured NN-ZnO and NM-ZnO films, the as-prepared NA-ZnO and MA-ZnO particles were respectively coated on the above-mentioned NC-ZnO underlayer films and then heating at 350 ^{o}C for 60 min. Simultaneously, NC-ZnO

monolayer films were prepared under identical experimental condition as described for the bilayer-structured ZnO film. In this work, the thickness of films was controlled by the concentration of ZnO in ethanol. For comparison, the thickness of single NC-ZnO film was also experimentally controlled to be identical to that of NN-ZnO film and NM-ZnO film. For dye adsorption, the resulting ZnO-based films were heated at 70 ^{o}C for 30 min and immersed in a 0.5 mM ethanolic solution of N3 for approximately 30min at room temperature. The sensitized ZnO-based films were rinsed with ethanol to remove excess dye remaining on the ZnO surface and then dried in air.

ZnO cell fabrication: The Pt-coated thin films on the FTO, served as counter electrodes, were prepared by depositing 0.35 mM H_2PtCl_6 solution on the FTO substrates, followed by annealing at 400 ^{o}C for 15 min in air. Pt electrodes were placed over the dye-adsorbed ZnO electrodes and the edges of the solar cells were sealed with ~ 2 mm wide stripes of sealing sheet. Then hot-pressing methods were performed at 80 ^{o}C for 1~2 min for cell assembly. Finally, the electrolyte solution, consisted of 0.3 M LiI, 0.06 M I_2, 0.5 M *t*-BPy and 1.0 M DMPII in acetonitrile, was injected into the cell from the edges. The active area of the resulting cell exposed in light was approximately 0.25 cm^2 (0.5 cm × 0.5 cm).

Characterization: The FESEM images were recorded on JSM 6701-F microscopes, respectively. An ASAP 2010 surface area analyzer was employed to determine BET data of ZnO-based samples. The amount of adsorbed dye was determined by desorbing the dye into a 1.0 M NaOH solution (50:50, V/V) and by absorption

measurement of the desorbed-dye solution using the adsorption peak intensity of N3 around 510 nm by a UV-vis spectrophotometer (UV 2501 spectrometer, Shimadzu). The photoelectrochemical measurements of DSSCs was performed using electrochemical station (CHI660C, ShangHai) under simulated AM 1.5 sunlight illumination with 100 $mWcm^{-2}$ light output. A 1000 W xenon lamp (Thermo Oriel, America) served as the light source. The EIS tests were carried out applied a bias of the open-circuit voltage of the ZnO-based cells with ac amplitude of 10 mV and a frequency ranging from 10^{-1} to 10^{5} Hz under 100 $mWcm^{-2}$ illumination. Monochromatic light in the range of 400-800 nm was obtained by using a series of filters and the incident photo to current conversion efficiency (IPCE) measurement was performed on a Keithley Model 2000 Source Meter. For transient absorption measurements (LP920，Edinburgh instruments Ltd.), the third harmonic (355 nm) of Nd:YAG laser was employed for excitation. The probe light from a pulsed xenon arc lamp was passed through various optical elements, samples, and a monochromator before being detected by a fast photomultiplier tube and recorded on a Tektronix TDS 3012B digital signal analyzer.

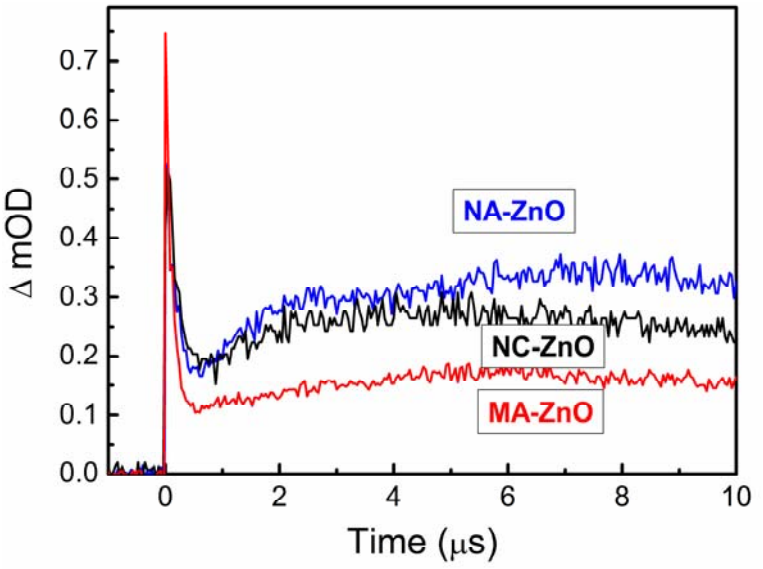

Fig. S2a Transient absorption decay kinetics of the NC-ZnO, MA-ZnO and NA-ZnO (excitation at 355 nm) collected at 550 nm.

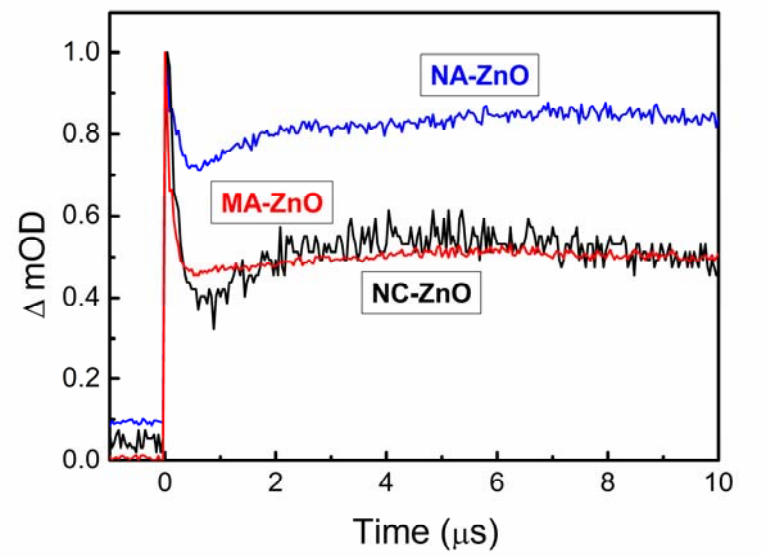

Fig. S2b Normalized decay kinetics of the NC-ZnO, MA-ZnO and NA-ZnO at time zero.