Supporting Information

Green Route and Ration Design for ZnO-based High-efficiency Photovoltaics

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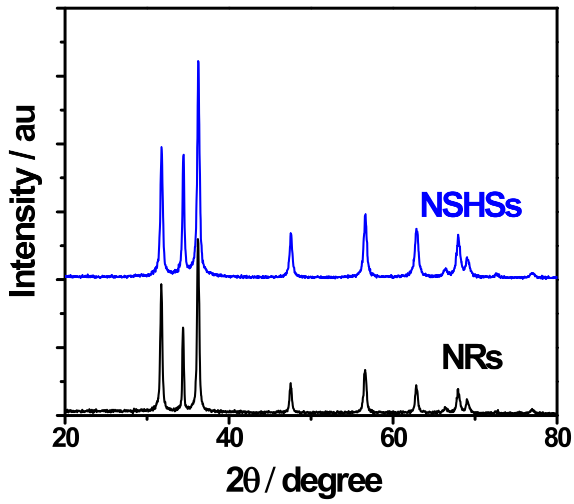

Figure S1. X-ray diffraction (XRD) patterns of the nanorods (NRs) and nanosheets-based hierarchical structures (NSHSs).

Figure S2. PEO-based polymer gel electrolyte at 100 °C, highly viscous.

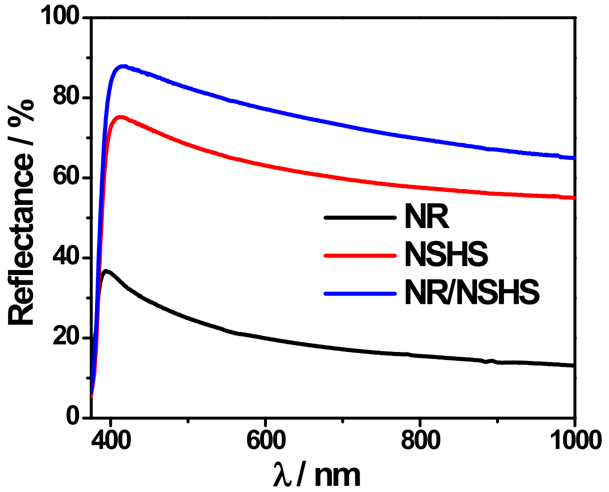

Figure S3. Reflectance spectra of the NR, NSHS, and the composite NR/NSHS photoanodes used for DSCs fabrications in this work

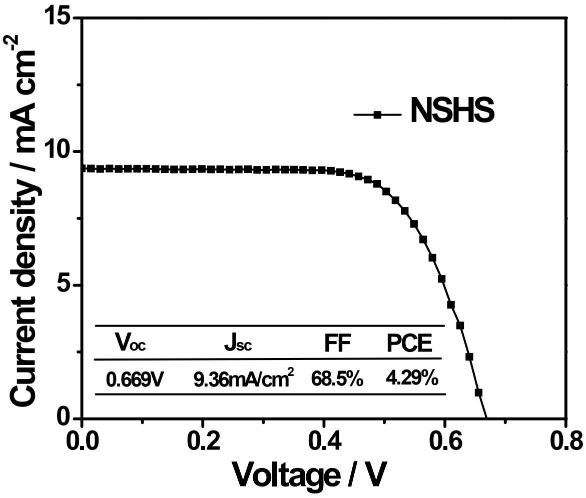

Figure S4. *J-V* curves of the quasi-solid DSCs based NSHS, film thick was ~ 39.2 μm

Electrochemical impedance spectra (EIS) analysis (For review only)

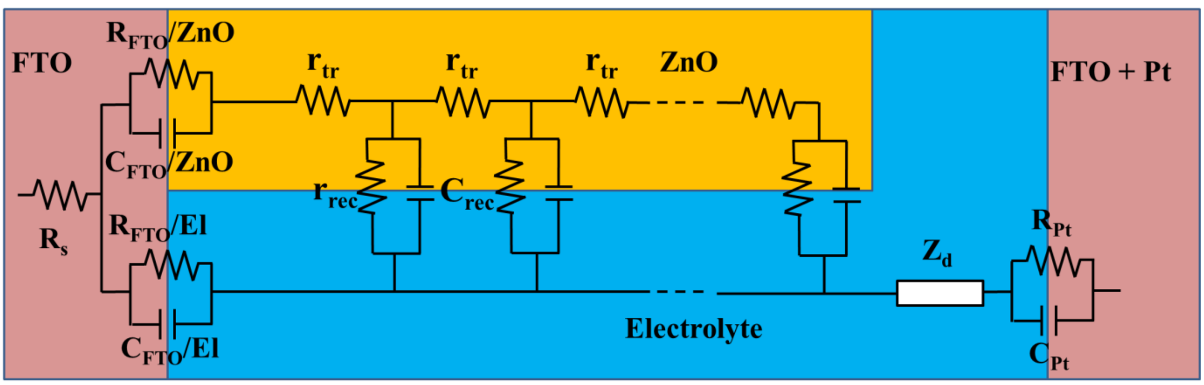

Figure S5. The transmission line model of DSCs, where the orange and blue parts represent the two channels of ZnO and electrolyte, respectively. r_{tr} represents the electron transport resistance in ZnO film; r_{rec} is the charge-transfer resistance between electrons in the ZnO film and I_3^- in the electrolyte, C_{rec} is the corresponding capacitance; Z_d is the Warburg element showing the Nernst diffusion of I_3^- in the electrolyte; R_{Pt} and C_{Pt} are the charge-transfer resistance and double-layer capacitance at the counter electrode, respectively; $R_{FTO/El}$ and $C_{FTO/El}$ are the charge-transfer resistance and the corresponding double-layer capacitance at the exposed FTO-electrolyte interface, respectively; $R_{FTO/ZnO}$ and $C_{FTO/ZnO}$ are the resistance and the capacitance at the TCO-ZnO contact, respectively; R_s is the series resistance, including the sheet resistance of the FTO glass and the contact resistance of the cell.

Upon small perturbation from external AC, the impedance Z of the photoanode can be expressed as Eq. 1:

$$Z = \left(\frac{R_{tr} R_{rec}}{1 + i\omega / \omega_k} \right)^{1/2} \coth\left[\left(\omega_k / \omega_d \right)^{1/2} \left(1 + i\omega / \omega_k \right)^{1/2} \right] \quad (1)$$

$$Z = \frac{1}{3} R_{tr} + \frac{R_{rec}}{1 + i\omega / \omega_k} \quad (2)$$

$$Z' = R_{tr} \left(i\omega / \omega_d \right)^{1/2} \quad (3)$$

where R_{tr}, R_{rec}, ω_k, and ω_d represent resistance of the electron transport in ZnO, the charge-recombination resistance at the ZnO/electrolyte interface, rate constant for recombination, and characteristic angular frequency for electron diffusion in ZnO, respectively. When the electrons are collected efficiently, Eq. 1 switches its expression to Eqs. 2 and 3 at lower and and higher frequencies, which correspond to the resistance of the charge recombination and electron transport, respectively.

Reference

[1] J. Bisquert, *J. Phys. Chem. B* **2002**, *106*, 325.

[2] Q. Wang, S. Ito, M. Grätzel, F. Fabregat-Santiago, I. Mora-Seró, J. Bisquert, T. Bessho, H. Imai, *J. Phys. Chem. B* **2006**, *110*, 25210.

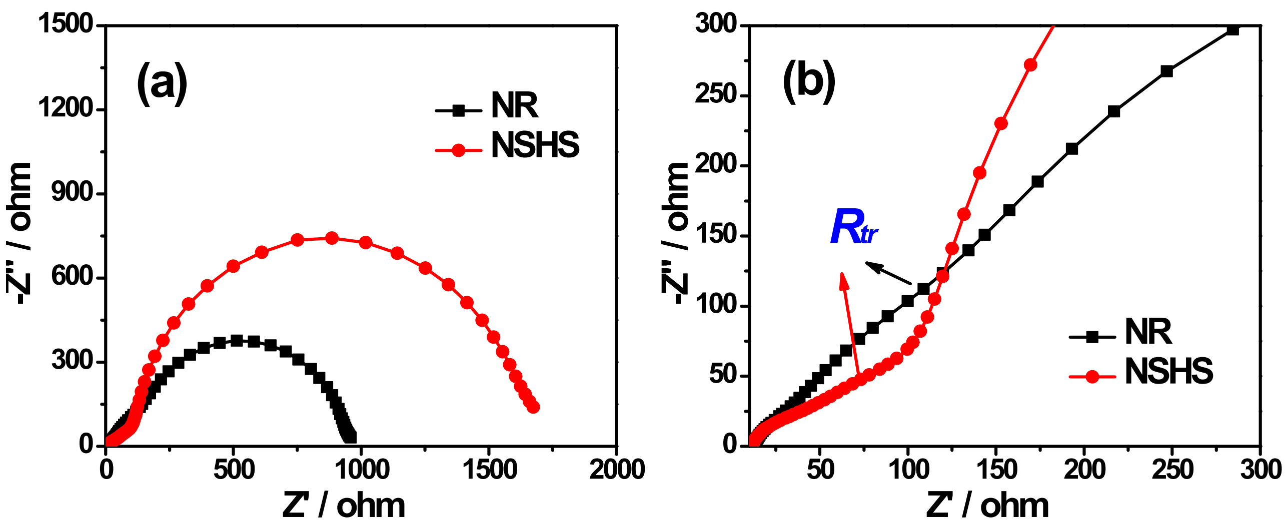

Figure S6. (a) Nyquist plots of the quasi-solid DSCs based on NR and NSHS under a bias of -0.56V, thicknesses of these two films were ~ 15 μm; (b) the enlarged parts of the high frequency region. According to the transmission line model and above EIS analysis, electron transport resistance (R_{tr}) within the NSHS photoanode was obviously smaller than that within NR.