
Supporting Information

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Figure S1. Pyrolysis synthesis product when the S source was removed under otherwise the same reaction conditions. (A) Scanning electron microscope (SEM) image reveals a similar sponge-like three-dimensional (3D) nanochain network; (B) X-ray diffraction pattern indicates a magnetite phase, which is a result from the reaction of the Fe nanosponge with the air forming Fe$_3$O$_4$. 
Figure S2. BET surface area analysis of the as-prepared FeS$_2$ molten-nanoparticles (A-B) and nanochain networks (C-D). (A) and (C) N$_2$ absorption and desorption isotherm curve; (B) and (D) Pore diameter distribution curve.

Figure S3. Preparation of FeS$_2$ thin film and bulk conductivity test. (A) Photographs of FeS$_2$ (nanochain network) isopropanol (IPA) nano-ink and as-deposited FeS$_2$ thin film on FTO glass; (B) Schematic illustration of bulk resistance test for FeS$_2$ thin film on glass; (C) Linear voltammetry curves of pristine and compressed FeS$_2$ thin films on glass, and compression post-treatment significantly improved the inter-particles connection and conductivity.
Figure S3A represents typical digital photos of FeS$_2$ IPA nano-ink and the as-deposited thin films. To investigate the effect of compression post-treatment, bulk conductivity measurements were conducted using pristine and compressed FeS$_2$ thin films on glass substrates. As shown in Figure S3B, two contacting Ag electrodes were deposited on FeS$_2$ thin films, linear voltammetry curves were recorded. As can be seen from Figure S3C, the resistance of compressed thin films is two orders of magnitude lower than that of the pristine ones. To eliminate the possible contact resistance problem, we further tested the bulk conductivity using Resistivity/Hall Measurement System (HL5500PC, Bio-Rad). Similarly, the testing result shows that the conductivity of the compressed FeS$_2$-thin film (~0.5 Ω cm) is two orders of magnitude higher than pristine ones (~30 Ω cm). In general, high quality FeS$_2$ nanochain thin film can be fabricated by drop-casting combined with compression post-treatment method.

Figure S4. (A) Cyclic voltammograms (CV) curves of FeS$_2$-chain-CEs prepared by multiple-coating process, it was found that 2 coating processes are sufficient for obtaining superior catalytic activity; (B) Solar cell power conversion efficiency as a function of film thickness; (C) Typical cross-section SEM image of the best-performing FeS$_2$-chain-CEs.

As shown in Figure S4A and S4B, the counter electrode needs enough amount of FeS$_2$ (2 cycles of coating, 4 μm) to achieve a superior catalytic performance and solar cell performance (PCE:
~7%). However, too thick a FeS$_2$-chain-CEs film will cause DSSC performance to decline presumably due to charge transport limitation. Figure S4C presents a typical cross-section SEM image of the best-performing FeS$_2$-chain-CE with a thickness of about 4 µm.