Iron vacancies and surface modulation of iron disulfides nanoflower as high power/energy density cathode for ultralong-life stable Li storage

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Fig. S1. The high-resolution SEM image of PFDFS@3DHHC nanocomposite.



Fig. S2. The STEM image of the PFDFS@3DHHC nanocomposite and EDX line-scanning mapping along the line.



Fig. S3. The SEM image of the as-prepared pristine DFS nanoparticles.



Fig. S4. (a) The nitrogen adsorption/desorption isotherm and (b) pore size distribution of the PFDFS@3DHHC nanocomposites.



Fig. S5. The Raman spectra of PFDFS@3DHHC and DFS/3DC.



Fig. S6. (a) The XPS spectrum of PFDFS@3DHHC nanocomposite. (b) The high-resolution of C_{1s} spectrum in the PFDFS@3DHHC nanocomposite.



Fig. S7. The CV profiles of the PFDFS@3DHHC and DFS/3DC nanocomposites at higher cut-off voltage of 3.0 V.



Fig. S8. Coulombic efficiencies comparisons of the two electrodes under different cut-off voltages.

As is well known, the Li_2S will be formed in the fully discharging process. According to working principles of lithium sulfur battery, the generated discharged products (Li_2S) is inevitable to convert to polysulfides and then to sulfur in the charging process at high cutoff voltage (3.0 V). After several cycles, the concentration of formed polysulfide intermediators becomes increased in the electrolyte. However, the higher concentration of polysulfides, the notorious shuttling effect becomes more severe. In other words, more soluble polysulfides migrate to the lithium anode and form nonconductive passivation layer on the lithium electrode surface, significantly deteriorating the Coulombic efficiency and the utilization of active materials. That is the reason why the Coulombic efficiency of DFS/3DC (1-3.0V) goes up and down after several cycles.



Fig. S9. The voltage curves of the PFDFS@3DHHC at different rate.



Fig. S10. (a) The SEM image and (b) the EDS spectrum of the PFDFS@3DHHC electrode after cycling.



Fig. S11. The calculated capacitive contribution in the PFDFS@3DHHC electrode at 0.1 mV s⁻¹.