

## **Electronic Supplementary Information:**

### **Colloidal Synthesis of Greigite Nanoplates with Controlled Lateral Size for Electrochemical Applications**

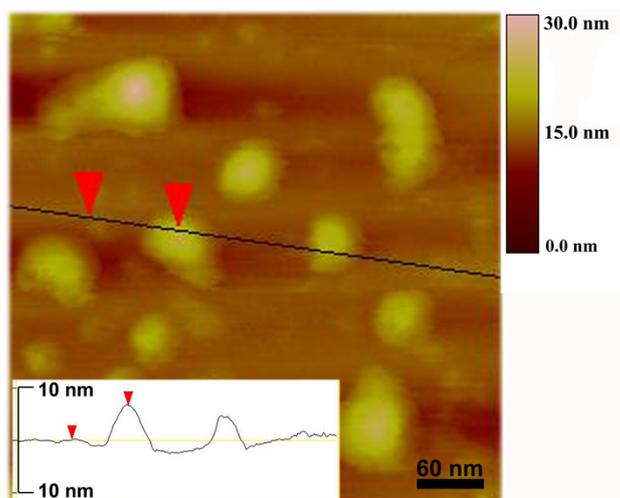
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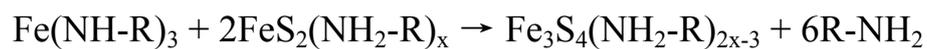
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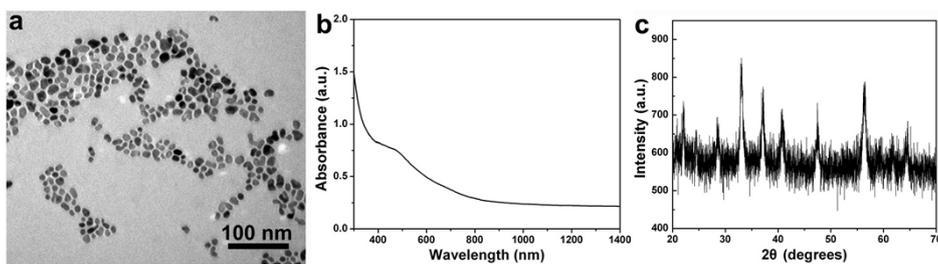
**Figure S1.** AFM observation of the as-synthesized Fe<sub>3</sub>S<sub>4</sub> NPs at the reaction temperature of 220 °C. The average thickness of NPs is 6.5 nm.

**Figure S2.** The proposed mechanism of the reduction of sulfur ion. The reduction reaction is shown below. FeS<sub>2</sub> is composed of one Fe<sup>2+</sup> and two S<sup>-</sup>, and Fe<sub>3</sub>S<sub>4</sub> is composed of one Fe<sup>2+</sup>, two Fe<sup>3+</sup>, and four S<sup>2-</sup>. In the reaction system, both Fe(III) and FeS<sub>2</sub> coordinate with ODA, which possesses weak reducibility. One ODA can provide one electron by β-hydride elimination.

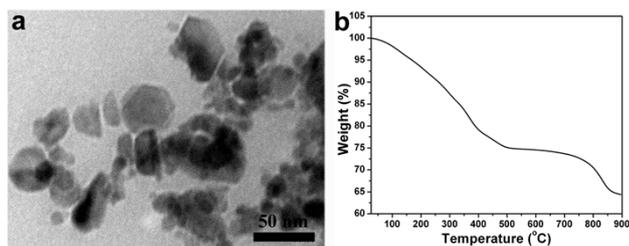


	Fe <sub>3</sub> S <sub>4</sub> NPs	FeS <sub>2</sub> nucleus	Fe-ODA complexes
Fe/S molar ratio	1/1.29	1/1.74	1/5.97

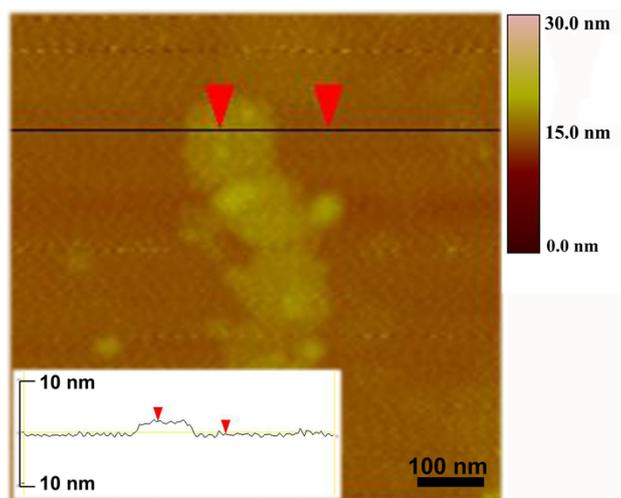
**Table S1.** The Fe/S molar ratio in the products, which is obtained from EDX measurement.



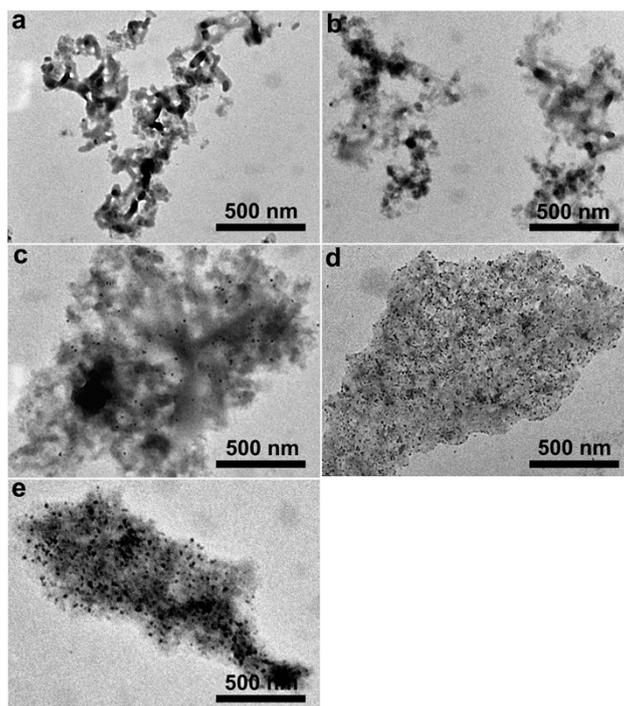
**Figure S3.** TEM image (a), UV-vis absorption spectrum (b), and XRD pattern (c) of the as-synthesized nanocrystals. The Fe source is  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ . The nanocrystals are pure phase  $\text{FeS}_2$ .



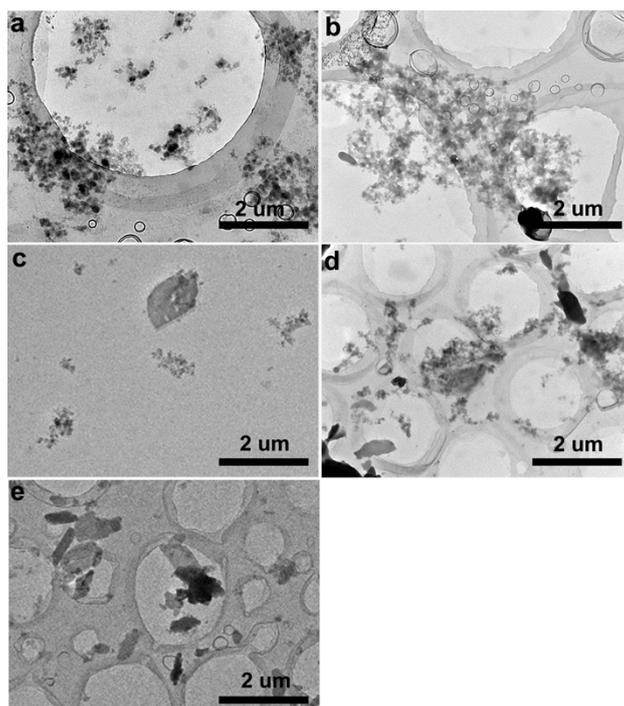
**Figure S4.** (a) TEM image of the  $\text{Fe}_3\text{S}_4$  NPs after annealing at 400 °C for 2 h under Ar atmosphere. (b) Thermogravimetric analysis profile of  $\text{Fe}_3\text{S}_4$  NPs heated in  $\text{N}_2$  from room temperature to 900 °C.



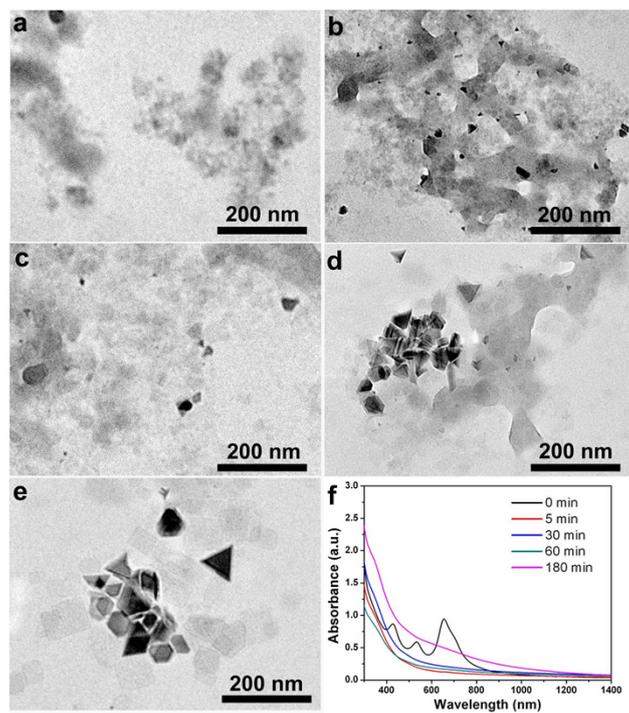
**Figure S5.** AFM observation of the as-synthesized Fe<sub>3</sub>S<sub>4</sub> NPs at the reaction temperature of 200 °C. The average thickness of NPs is 2.5 nm.



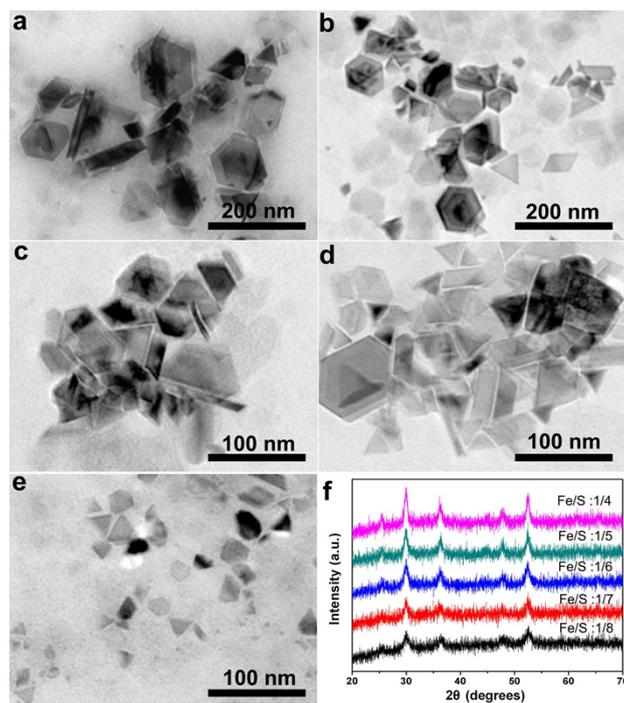
**Figure S6.** TEM images of the products captured at nucleation stage with the DE/ODA volume ratio of 0.7/1 (a), 1/1(b), 2/1 (c), 5/1 (d), and 11/1 (e), which reveal the increase of the number of FeS<sub>2</sub> nucleus as increasing DE.



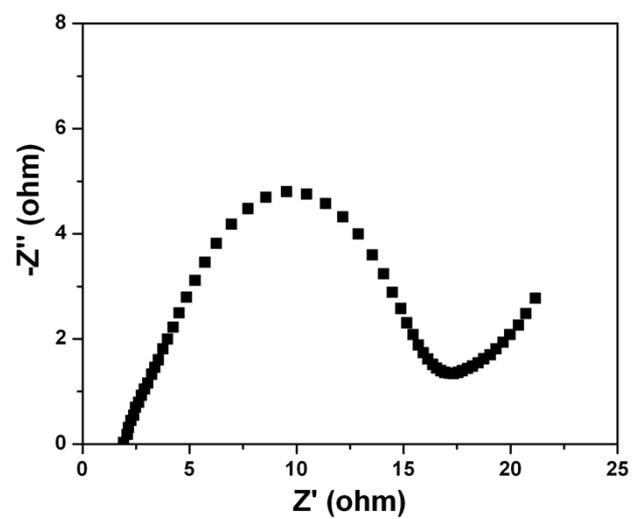
**Figure S7.** TEM images of the products captured at nucleation stage with the Fe/S feed ratio of 1/4 (a), 1/5 (b), 1/6 (c), 1/7 (d), and 1/8 (e), which reveal the decrease of the number of FeS<sub>2</sub> nucleus as decreasing Fe source.



**Figure S8.** The temporal evolution of Fe<sub>3</sub>S<sub>4</sub> NPs which is revealed by TEM observation. (a) 0 min, (b) 5 min, (c) 30 min, (d) 60 min, and (e) 180 min. (f) The corresponding absorption spectra. The DE/ODA volume ratio is 1/1.



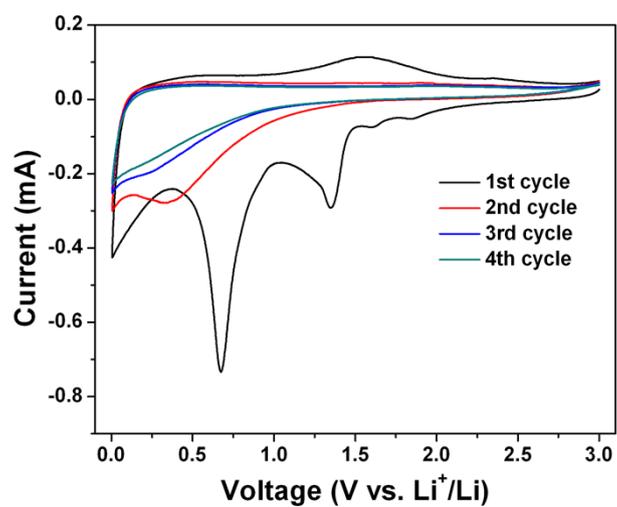
**Figure S9.** TEM images of the  $\text{Fe}_3\text{S}_4$  NPs synthesized with the Fe/S molar feed ratio of 1/4 (a), 1/5 (b), 1/6 (c), 1/7 (d), and 1/8 (e), which reveal the decrease of lateral size as increasing S. (f) The corresponding XRD patterns.



**Figure S10.** EIS profile of a fresh Fe<sub>3</sub>S<sub>4</sub>-based coin cell.

<b>Samples</b>	<b><i>Re</i></b>	<b><i>Rf</i></b>	<b><i>Rct</i></b>
3rd	2.4	25.9	146.9
5th	2.8	28.6	128.5
7th	5.3	15.1	72.5
15th	3.6	17.2	38.3
20th	3.8	16.8	36.7

**Table S2.** Kinetic parameters of the Fe<sub>3</sub>S<sub>4</sub>-based electrode at different cycles.



**Figure S11.** Cycle voltammetry profiles of the first four cycles for the Fe<sub>3</sub>S<sub>4</sub>-based electrode at a scanning rate of 0.1 mV/s in the voltage window of 0.001-3.0 V (Li<sup>+</sup>/Li).