Supporting Information

Natural hematite-derived NiFe$_2$O$_4$ as a separator modification material for improved Li-S battery performance

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Experimental Section

Chemicals and Instrumentations

The Natural purified hematite is purchased from Shanlin Shiyu Mineral Co., Ltd. Commercial sulfur powder, N-methyl-2-pyrrolidone (NMP), and polyvinylidene difluoride (PVDF) were acquired from Shanghai Aladdin Bio-Chem Technology Co., Ltd, located in China. Acetylene black, Ferric nitrate nine-hydrate, Ferric chloride hexahydrate，Sodium acetate, 1,2-Diaminopropane，Diethylenetriamine and Ethanol were obtained from Sinopharm Chemical Reagent Co., Ltd, China. All reagents were utilized without undergoing additional purification. For all the experiments, ultrapure water (Millipore, 18.25 MΩ cm-1) was utilized.

Instrumentations

Wide-angle X-ray diffraction (WAXRD, Bruker D8 advance) with Cu Kα radiation characterized the crystal structures of the samples in the 2θ range of 5° to 80°. Scanning electron microscopy (SEM, FEI nanoSEM450) and transmission electron microscopy (TEM, Jem-2100F) were used to test the morphologies of the samples. The sample's elemental chemical environment was characterized using X-ray photoelectron spectroscopy (XPS, Thermofisher escalab 250xi).

Synthesis of NFO

Natural hematite powder and nickel hydroxide were mixed in a molar ratio of 1:1, and dry-ground in a mortar for 30 minutes. The ground sample was placed in a tube furnace, nitrogen was passed through,

and heated at 950 °C for 4 hours at a heating rate of 5 °C/min.

Preparation of NFO@PP

A slurry was prepared by mixing NFO, polyvinylidene fluoride (PVDF), and acetylene black in a ratio of 8:1:1, using N-methyl-2-pyrrolidone (NMP) as the dispersant. The resulting slurry was coated onto a Celgard 2400 separator, followed by vacuum drying at 60°C for 24 hours to obtain a modified separator. The modified separator was then punched into discs with a diameter of 19 mm for use in 2016 coin cells.

Preparation of the sulfur cathode

A sulfur slurry was obtained by dispersing sulfur powder, conductive carbon black and PVDF at a weight ratio of 8:1:1 in NMP. The slurry was then coated on Al foil, which was completely dried in a vacuum oven to evaporate the NMP solvent and then cut into small circular working electrodes with a diameter of 10 mm. For each electrode disk, the loading amount of active material was about 1.5 mg.

Electrochemical measurements

All Li-S battery tests were carried out by standard 2016-type coin cells. Typical, the slurry was made via uniform mixing S/CNT, acetylene black and PVDF (8:1:1). Subsequently, the slurry was applied onto an aluminum foil. The coin cells were assembled with lithium metal as a counter electrode and polypropylene membrane as the separator, and the electrolyte was 1.0 M Li bis (tri-fluoromethanesulphonyl) imide (LiTFSI) dissolved in 1, 3 dioxolane (DOL) and 1, 2-dimethoxyethane (DME) (1:1 by volume) with 1.0 wt.% $LiNO_3$ as additive. The batteries were built within a glovebox that contained ultra-pure Ar gas, ensuring that the levels of water and oxygen remained below 0.01 p.p.m. The galvanostatic charge-discharge experiments were performed at a temperature of 25 °C using a Land battery tester (Land CT3001A, China) at various rates within the voltage range of 1.7-2.8 V. Cyclic voltammetry (CV) was conducted utilizing an electrochemical workstation (CHI760E Shanghai, China). The specific capacity is determined by the mass of Sulphur (S) present in the active material, while the areal sulfur loading is approximately 1 mg cm^{-2}.
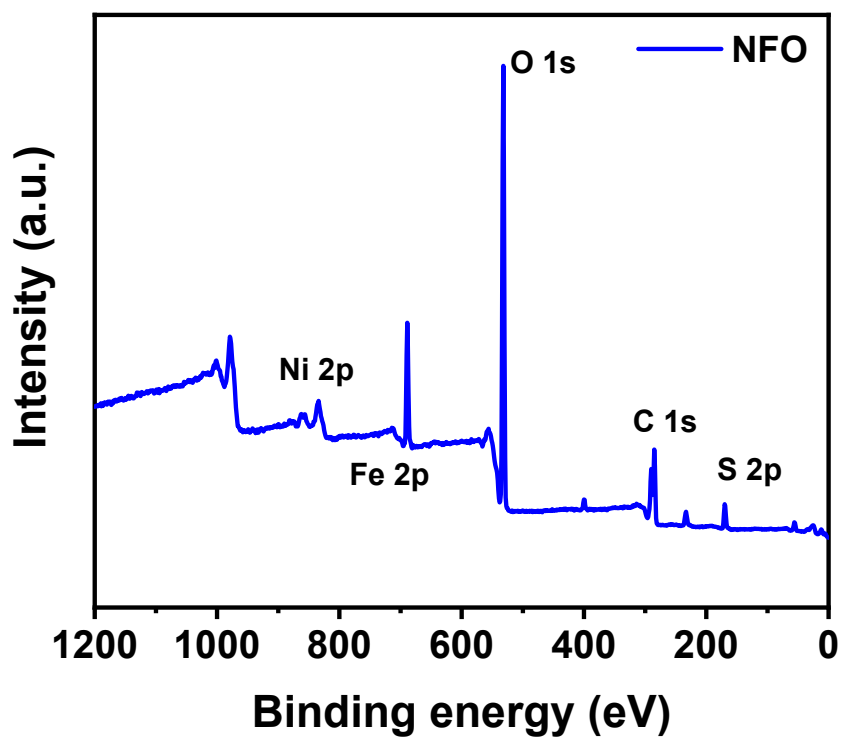
Supplementary figures
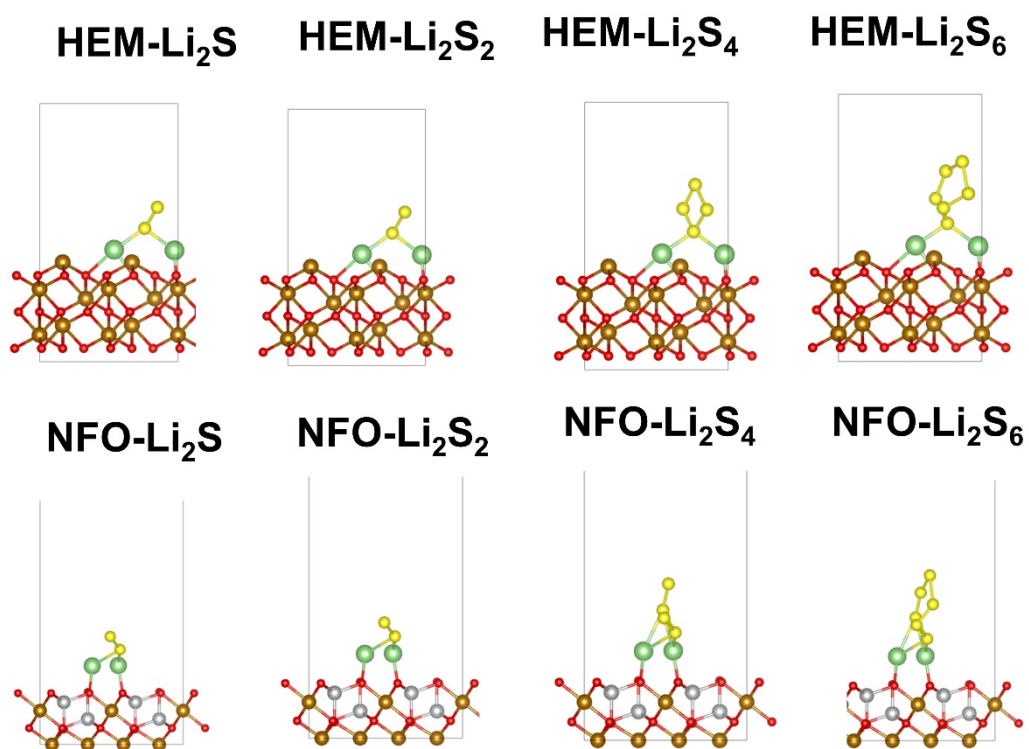

Fig. S1. XPS survey spectrum of NFO.

Fig. S2. Optimized geometric configurations for the binding of Li_2S_n (n = 2, 4, 6, 8) to HEM and NFO
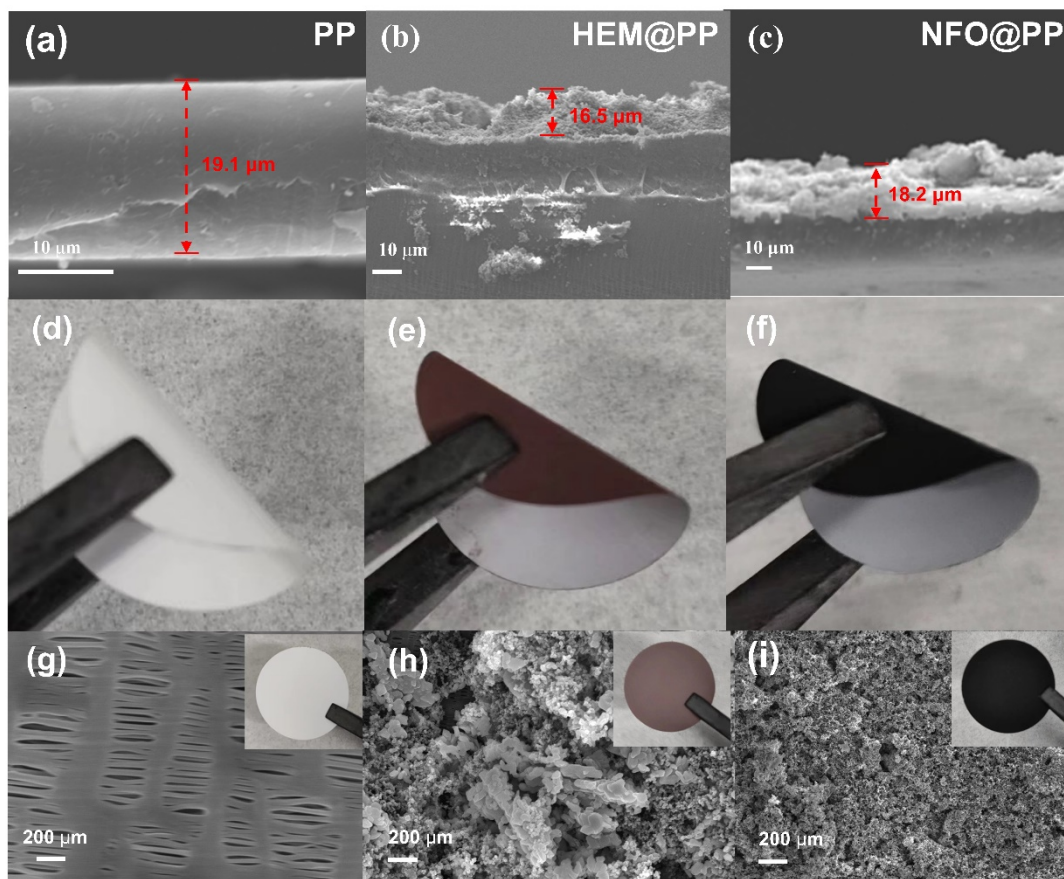
Fig. S3. SEM image of the cross section of (a) PP, (b) HEM@PP and (c)NFO@PP. Flexibility of (d) PP, (e) HEM@PP and (f) NFO@PP. SEM images and digital photographs of (g) PP, (h) HEM@PP and (i) NFO@PP.
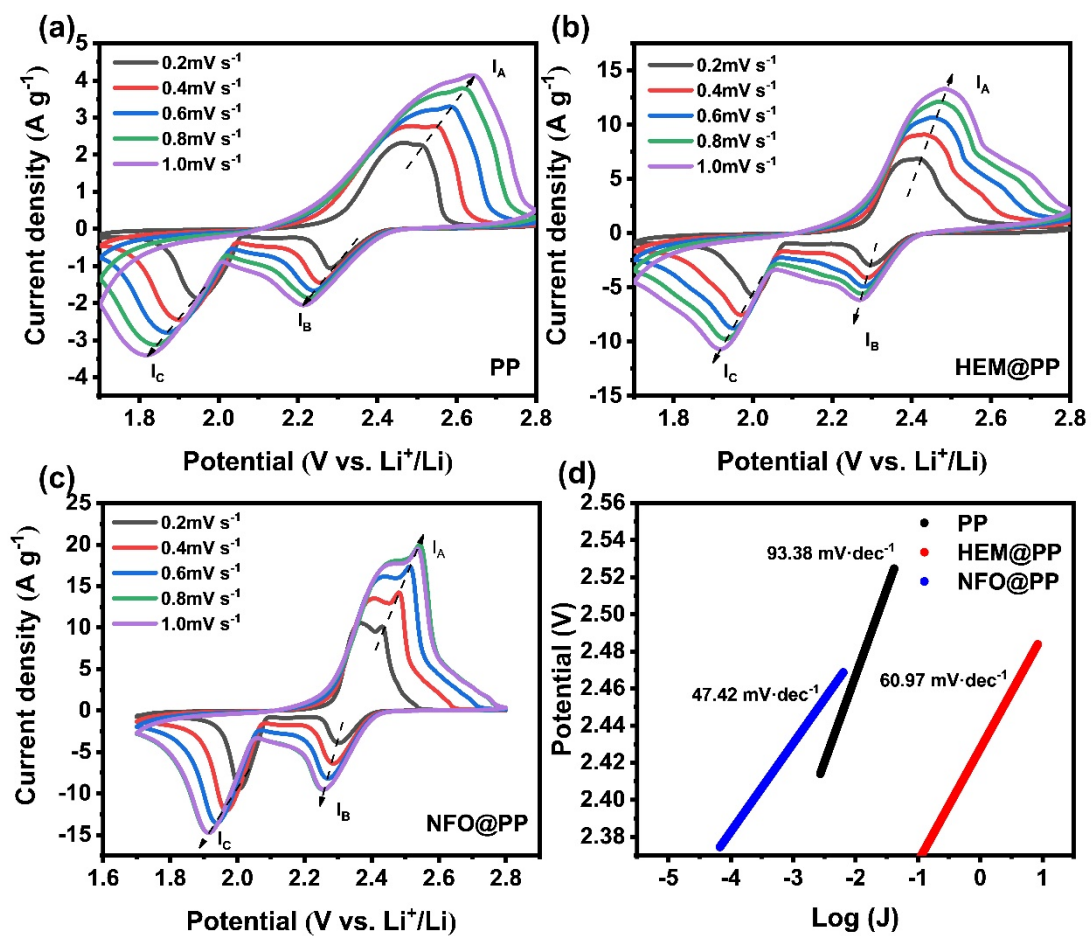
Fig. S4. CV curves (0.2 to 1.0 mV s^{-1}) of (a) Pure PP, (b) HEM@PP and (c) NFO@PP. (d) Tafel curves of PP, HEM@PP, and NFO@PP
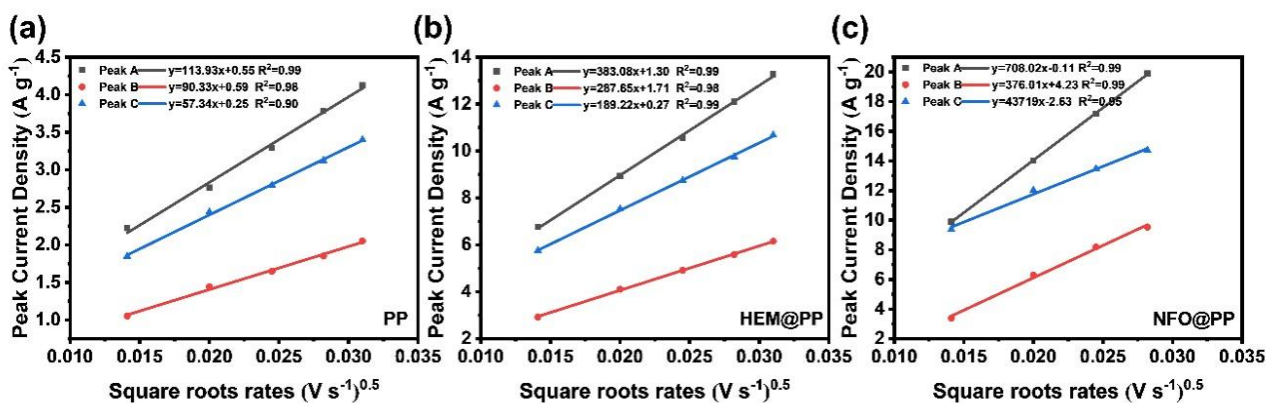
Fig. S5. Slope of (a) PP, (b) HEM@PP and (c) NFO@PP.

Table S1. Electrochemical performance comparison for Li-S coin batteries.

Materials Type	Rate	Specific capacity (mAh·g^{-1})	Ref.(SI)
Co-Mo$_2$C@NCNTs//PP	0.5C	648.15 (500th)	1
Co-NC-X	2C	470.5 (500th)	2
Fe-NG/PP	1C	678 (500th)	3
Sn-MOF@ZIF-8	0.5C	515 (500th)	4
SnO$_2$-CNT-PANI‖PP=|SnO$_2$	1C	433.6 (800th)	5
Y$_2$O$_3$/YS@C	0.5C	530.5 (400th)	6
NFO@PP	**0.5C**	**612.1 (1000th)**	**This work**

References

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