Electronic Supplementary Material

$(FeO)_2FeBO_3$ nanoparticles attached on interconnected nitrogen-doped carbon nanosheets serving as sulfur hosts for lithium–sulfur batteries

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1 Materials characterization

Crystal structures of samples were examined by X-ray diffraction (XRD; Shimadzu XRD-6000) using the high-intensity Cu Kα radiation with a wavelength of 1.54178 Å, and the morphologies and element distributions were investigated through scanning electron microscopy (SEM; Hitachi S8100) accompanied with energy-dispersive X-ray spectroscopy (EDS). Transmission electron microscopy (TEM; Hitachi HT-7700) and high-resolution transmission electron microscopy (HRTEM; JEOL-2010 TEM) were performed to determine the microstructures of samples. Raman spectra (Renishaw inVia) were obtained using a 532 nm laser source. Brunauer–Emmett–Teller (BET) specific surfaces areas (SSAs) and pore volumes of samples were measured using a Micromeritics ASAP 2460 unit. Thermogravimetric analysis (TGA; Setaram Labsys Evo SDT Q600) was carried out at a heating rate of 5 °C·min^{-1} from room temperature to 700 °C in air. X-ray photoelectron spectroscopy (XPS; ESCALAB 250) was used to analyze the surface elemental compositions and chemical bondings.

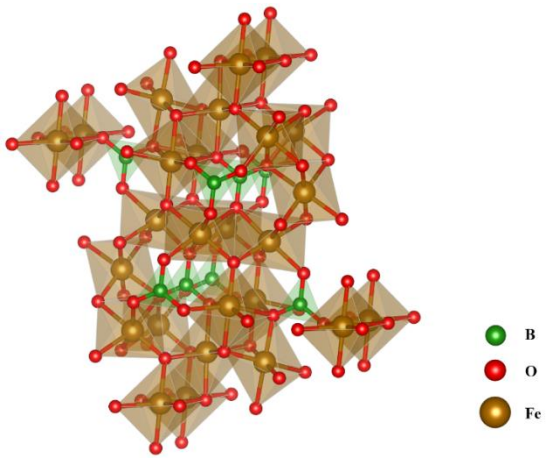

Fig. S1 Diagram of the atomic structure of orthorhombic $(FeO)_2FeBO_3$.

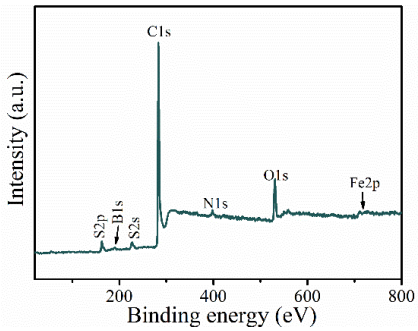

Fig. S2 XPS survey spectrum of the $(FeO)_2FeBO_3$@NCNs/S composite.

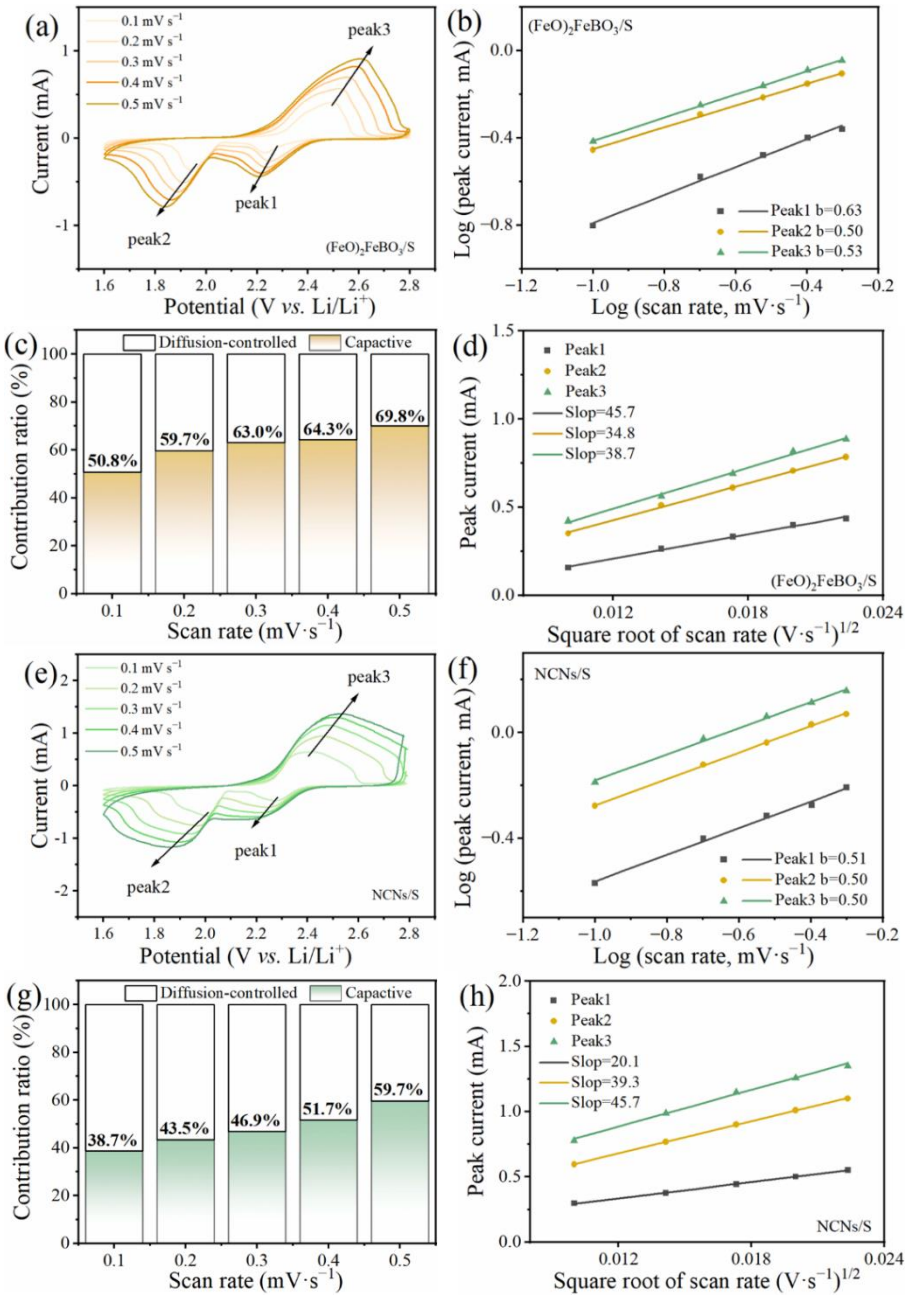

Fig. S3 (a) CV curves for the $(FeO)_2FeBO_3$/S cathode at a series of scan rates. **(b)** Plots of lgI_p *vs*. lg$v$ for the $(FeO)_2FeBO_3$/S cathode. **(c)** Contribution ratios of capacitance- and diffusion-controlled processes for the $(FeO)_2FeBO_3$/S cathode. **(d)** Plots of I_p *vs*. $v^{1/2}$ for the $(FeO)_2FeBO_3$/S cathode. **(e)** CV curves for the NCNs/S cathode at a series of scan rates. **(f)** Plots of lgI_p *vs*. lg$v$ for the NCNs/S cathode. **(g)** Contribution ratios of capacitance- and diffusion-controlled processes for the NCNs/S cathode. **(h)** Plots of I_p *vs*. $v^{1/2}$ for the NCNs/S cathode.

2 Electrochemistry measurements

Electrochemical performances of $(FeO)_2FeBO_3$@NCNs-1/S, $(FeO)_2FeBO_3$@NCNs-2/S, $(FeO)_2FeBO_3$@NCNs-3/S, NCNs/S, and $(FeO)_2FeBO_3$/S were tested with a button cell (CR2032). The composite (75 wt.%), carbon black (CB; 15 wt.%), and polyvinylidene fluoride (PVDF; 10 wt.%) were mixed with a certain amount of N-methylpyrrolidone (NMP), and the resulted mixture was stirred into a uniform slurry. Al foil (Shanghai Aladdin Reagent Co., Ltd.) was coated evenly 14 mm in diameter and 0.015 mm in thickness, followed by vacuum-drying at 60 °C for 12 h. The cells were subsequently assembled in an Ar-filled glove box ($c(O_2) < 0.01$ ppm, $c(H_2O) < 0.01$ ppm; Mikrouna, Super 1220/750/900). The electrolyte was 1 mol·L^{-1} bis(trifluoromethane)sulfonamide lithium salt (LiTFSI; Sigma Aldrich) at a 1:1 volume ratio of 1,3-dioxolane (DOL) and 1,2-dimethoxyethane (DME) with 1 wt.% $LiNO_3$ additive. Galvanostatic discharging–charging curves as well as cycle and rate performances from the Neware battery test system (Neware CT-3008) were obtained in the potential range of 1.6–2.8 V. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were performed on an electrochemical workstation (Chenhua CHI-660E) from 0.01 Hz to 100 kHz.

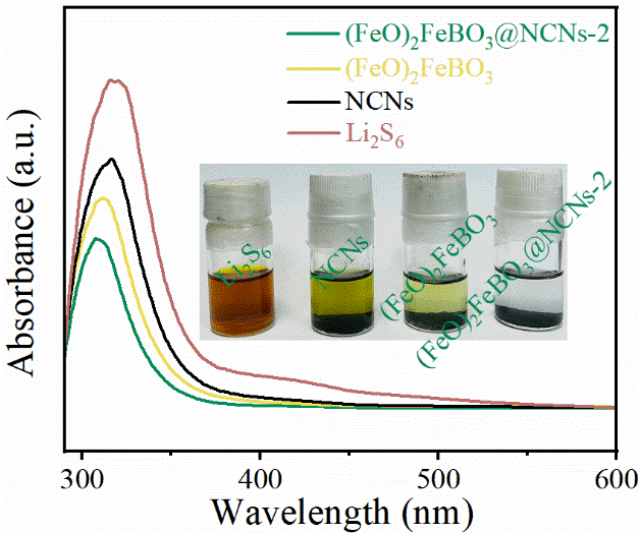

Fig. S4 UV–vis spectra of residual solutions after the adsorption for 24 h. Inset shows the optical image of a sealed vial containing the Li_2S_6/DOL/DME solution and those in contact with NCNs, $(FeO)_2FeBO_3$, and $(FeO)_2FeBO_3$@NCNs-2 after 24 h.

Table S1 Cycling performances of the $(FeO)_2FeBO_3$@NCNs/S electrode and other reported cathodes [44,50–54]

Material	Preparation method	i/(A·g^{-1})	Cycle number	C/(mA·h·g^{-1})	Ref.
$Fe(OH)_3$@GO/S	Hydrothermal method	0.835	500	520.4	[44]
Fe, N co-doped mesoporous carbon spheres	Calcination method	0.334	200	547	[50]
(GQD)/Fe_2O_3@S@SnO_2	Calcination method	0.668	350	450	[51]
Co_3O_4/CoO/GNS/h-BN	Hydrothermal method	1.67	250	356.2	[52]
N-Co_3O_4@N-C/rGO	Calcination method	0.334	500	568	[53]
Fe_3O_4-NC/S	Calcination method	1	500	532	[54]
$(FeO)_2FeBO_3$@NCNs/S	Soaking–drying–calcination	0.2	200	605.9	This work
		0.5	1000	458.3	
		1	1000	436.8	

Notes: i, current density; C, capacity.

3 Adsorption experiment

0.01 mol·L^{-1} Li_2S_6 solution was prepared by mixing sulfur powder and Li_2S at a mole ratio of 5:1 with the DME/DOL solution at a volume ratio of 1:1 in an Ar-filled glove box. The solution was then stirred at 80 °C for 15 h. Finally, 10 mg of NCNs, $(FeO)_2FeBO_3$, and $(FeO)_2FeBO_3$@NCNs-2 were separately added into 3 mL of the Li_2S_6 solution. Adsorption properties of the LiPS solution were analyzed after culturing for 24 h.

Table S2 EIS fitting parameters of NCNs/S, $(FeO)_2FeBO_3$/S, and $(FeO)_2FeBO_3$@NCNs/S cathodes

Electrode material	R_1/Ω	R_{ct}/Ω	R_2/Ω
NCNs/S (before cycling test)	2.3	145.0	–
NCNs/S (after 500 cycles)	1.6	474.6	20.34
$(FeO)_2FeBO_3$/S (before cycling test)	1.5	137.5	–
$(FeO)_2FeBO_3$/S (after 500 cycles)	2.0	135.6	51.9
$(FeO)_2FeBO_3$@NCNs-2/S (before cycling test)	1.4	92.7	–
$(FeO)_2FeBO_3$@NCNs-2/S (after 500 cycles)	1.6	70.1	16.8