

Supporting Information for

A Ternary Hybrid Material for High Performance Lithium-Sulfur Battery

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Preparation of mildly oxidized CNTs. CNTs were oxidized by a modified Hummers method. Multi-wall CNTs (CNano Tech. Ltd.) were purified by calcinations at 400 °C for 1 h and washed with 10 wt% HCl to remove metal residues. 1g of purified CNTs were dispersed into 23 ml of concentrated H₂SO₄ and the mixture was stirred at room temperature overnight. Next, the solution was heated to 40 °C in an oil bath. 350 mg of NaNO₃ was added, followed by the slow addition of 1 g of KMnO₄ while keeping the reaction temperature below 45°C. The solution was kept at 40°C under stirring for 30 min. 3 ml of water was added into the flask, followed by another 3 ml after 5 minutes. After another 5 minutes, 40 ml of water was added. 15 minutes later, the flask was removed from the oil bath and 140 ml of water and 10 ml of 30% H₂O₂ were added to end the reaction. Oxidized CNTs were collected, repetitively washed with 5 wt% HCl solution and then water, and finally lyophilized to acquire the mildly oxidized CNTs (Fig. S8).

Preparation of Li₂S₆ solution. Polysulfide solution was prepared by dissolving stoichiometric amounts of Li₂S and sulfur in DOL at 80 °C for 10 hours.

Polysulfide adsorption study. Test solutions were prepared by mixing 20 µl of 0.3 M Li₂S₆ in DOL, 1 ml of DOL and 1 ml of DME. 5 mg of CNT/NiFe₂O₄, CNT/NiFe₂O₄-2 or CNTs was added to each solution. The solutions were vigorously stirred for 20 min. All procedures were completed in an Ar-filled glove box. To further test whether the host materials can still effectively trap polysulfides after the long-term cycling tests, the cycled cathodes of CNT/NiFe₂O₄-S, CNT/NiFe₂O₄-S-2 and CNT-S with absorbed electrolyte were each directly soaked in 4 mL of DOL/DME (1:1, vol) mixed solvent for 24 hours. All procedures were completed in an Ar-filled glove box.

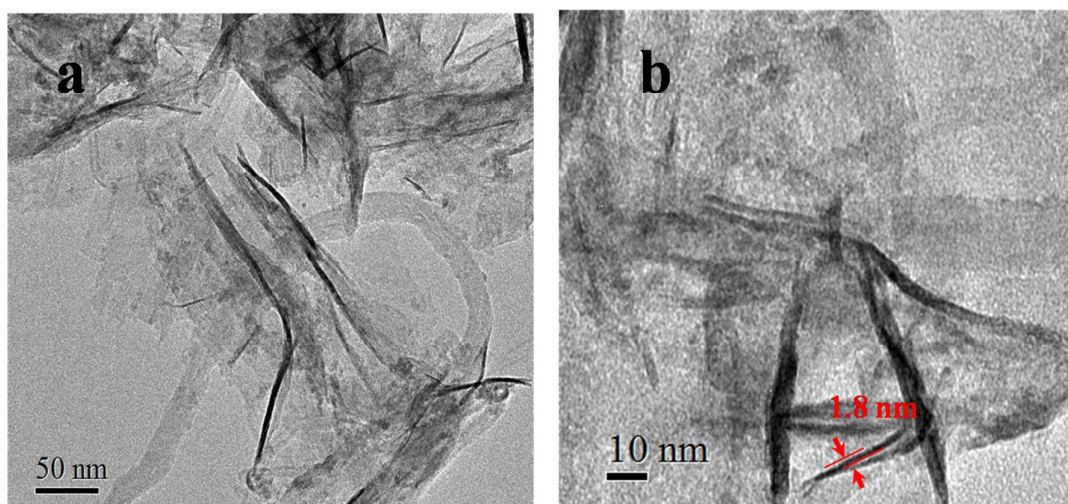


Fig. S1 TEM images of NiFe_2O_4 nanosheets grown on CNTs.

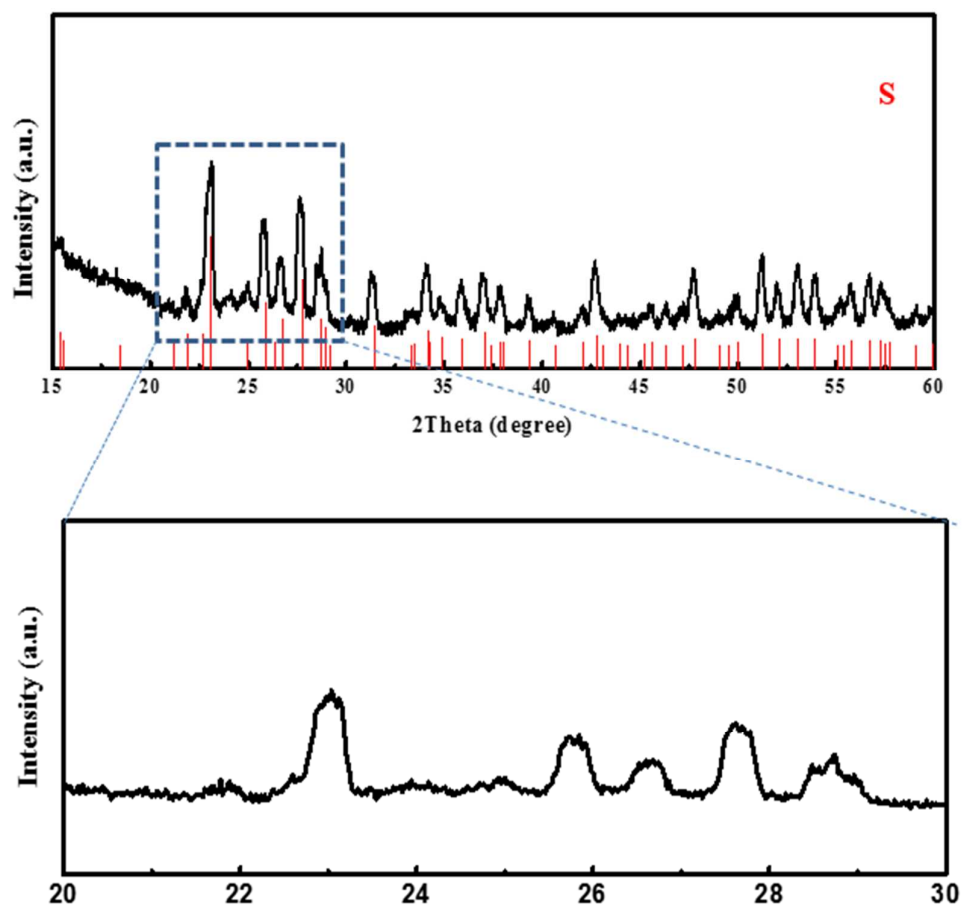


Fig. S2 XRD pattern of the CNT/ NiFe_2O_4 -S ternary hybrid material.

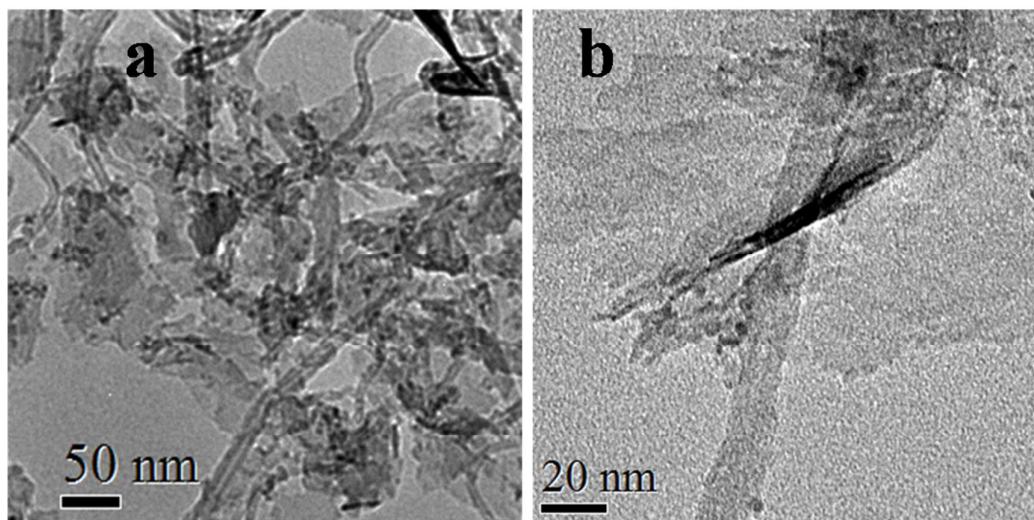


Fig. S3 TEM images of CNT/NiFe₂O₄-S after long-term cycling.

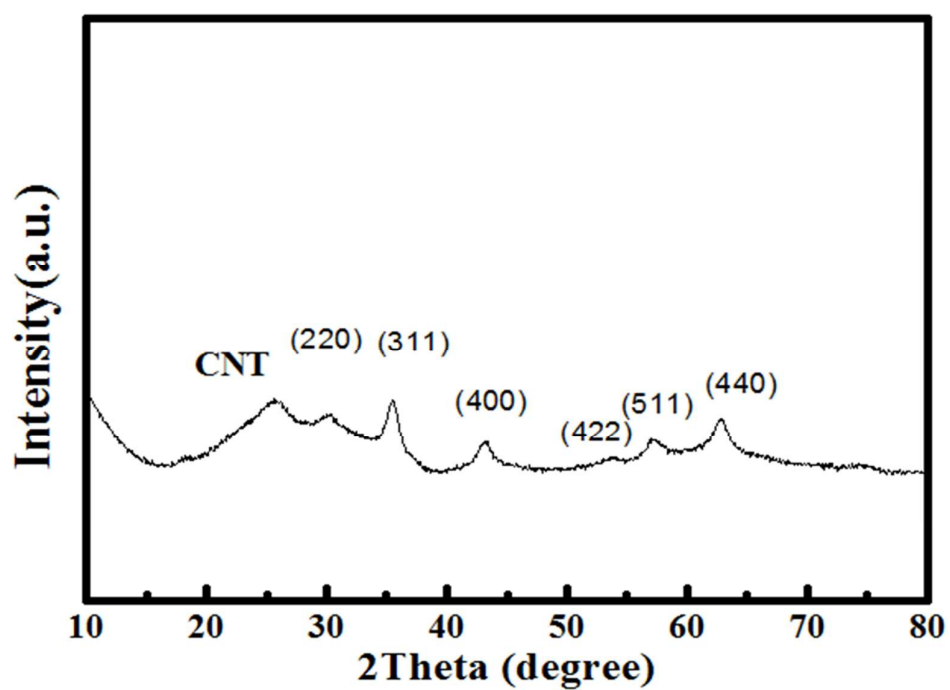


Fig. S4 XRD pattern of the discharged electrode material after hundreds of cycles.

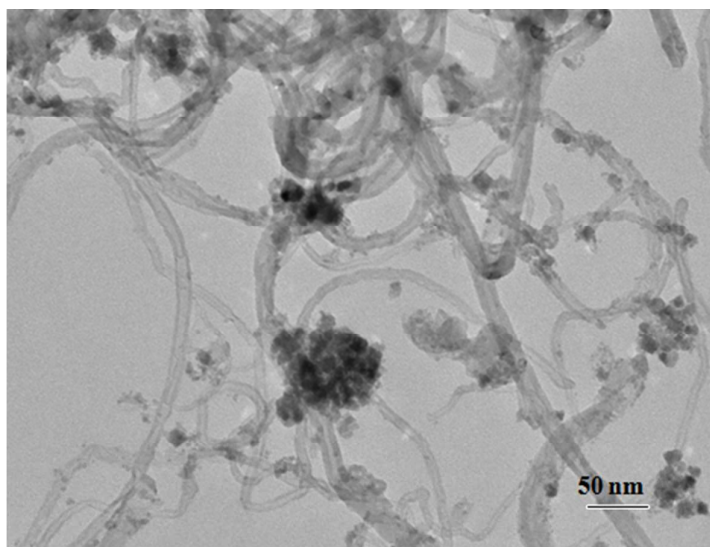


Fig. S5 TEM image of NiFe₂O₄ nanoparticles grown on CNTs.

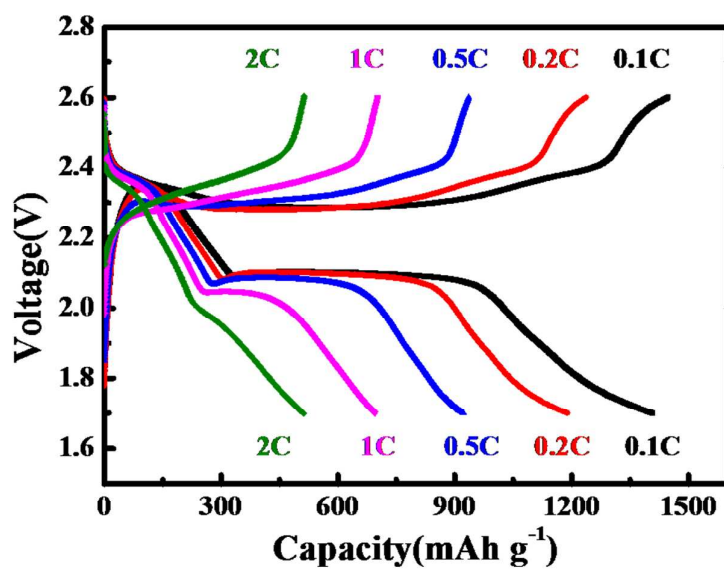


Fig. S6 Charging/discharging voltage profiles of the CNT-S at various C rates from 0.1 to 2 C.

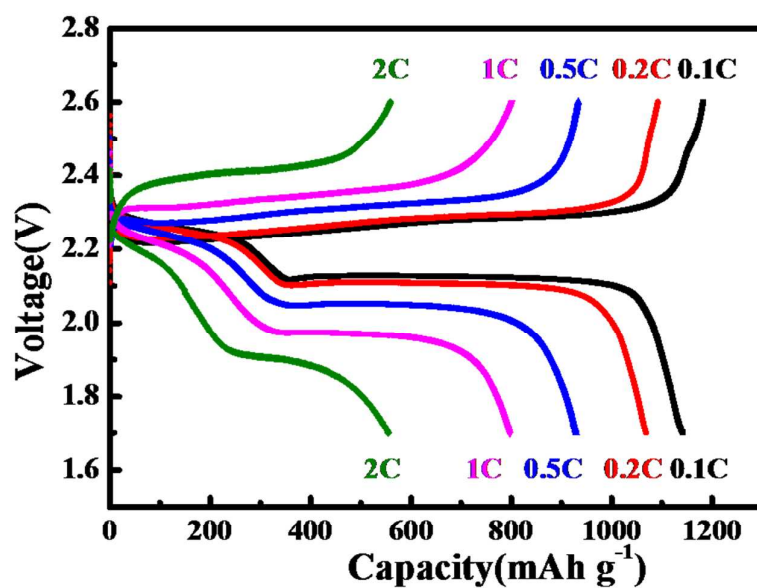


Fig. S7 Charging/discharging voltage profiles of the CNT/NiFe₂O₄-S-2 at various C rates from 0.1 to 2 C.

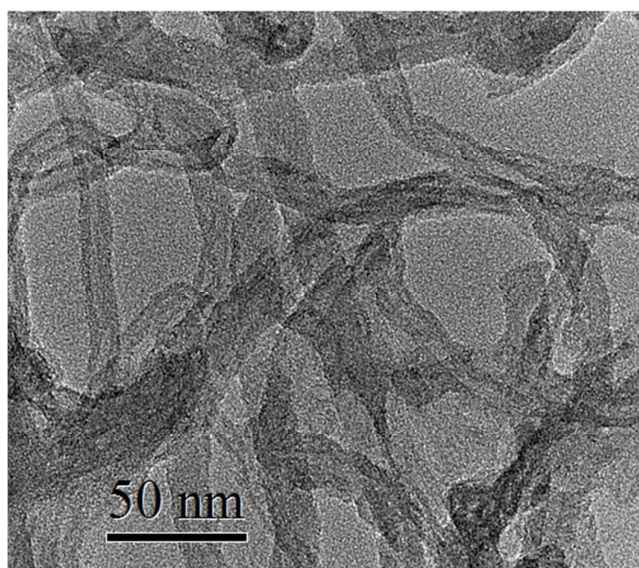


Fig. S8 TEM image of the mildly oxidized CNTs.

| Host Material | Sulfur Loading | Capacity loss per cycle | |
|---|----------------|-------------------------|-----------------|
| | | Cycles | Loss (%) |
| CNT/NiFe ₂ O ₄ nanosheet <i>This work</i> | 76 wt% | >500 | 0.009 (at 1C) |
| ITO-Carbon Fiber ¹ <i>Ref.1</i> | 57 wt% | 500 | 0.036(at 0.2C) |
| MnO ₂ nanosheet ² <i>Ref. 2</i> | 75 wt% | 2000 | 0.036 (at 2C) |
| TiO ₂ hollow sphere ³ <i>Ref.3</i> | 71 wt% | 1000 | 0.033 (at 0.5C) |
| Ti ₄ O ₇ ⁴ <i>Ref.4</i> | 70 wt% | 500 | 0.06 (at 2C) |
| Amino-functionalized reduced graphene oxide ⁵ <i>Ref.5</i> | 60 wt% | 350 | 0.057 (at 0.5C) |
| Covalently bonded CNT ⁶ <i>Ref.6</i> | 83 wt% | 500 | 0.021 (at 0.5C) |
| Graphene ⁷ <i>Ref.7</i> | 70 wt% | 300 | 0.1 (at 1C) |
| N-Doped Graphene ⁸ <i>Ref.8</i> | 60 wt% | 700 | 0.068 (at 1C) |
| CNT-interpenetrated mesoporous N-doped carbon sphere ⁹ <i>Ref.9</i> | 70 wt% | 200 | 0.05 (at 0.2C) |
| N-Doped Double-Shelled Hollow Carbon Sphere ¹⁰ <i>Ref.10</i> | 78 wt% | 200 | 0.19 (at 0.5C) |
| Ultra-high-surface-area hollow carbon nanosphere ¹¹ <i>Ref.11</i> | 67 wt% | 500 | 0.053 (at 1C) |
| Porous trithiocyanuric acid ¹² <i>Ref.12</i> | 63 wt% | 450 | 0.037 (at 0.5C) |

Table S1 Comparison of cycling stability of representative S cathode material structures in the literature.

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