

Supplementary Information for

## **Atomic Layer Deposition Assisted Construction of Binder-Free Ni@N-Doped Carbon Nanospheres Films as Advanced Host for Sulfur Cathode**

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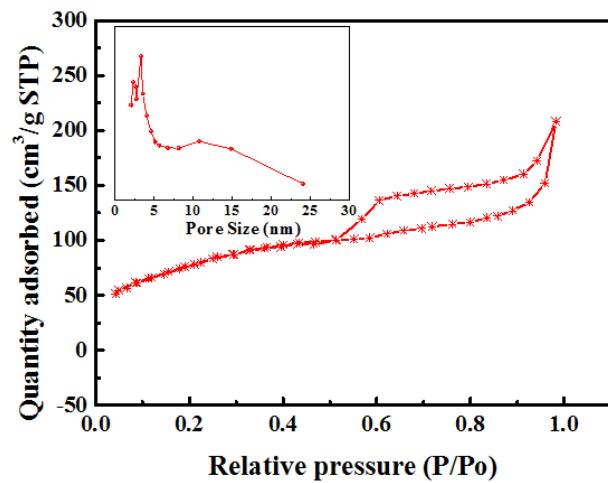
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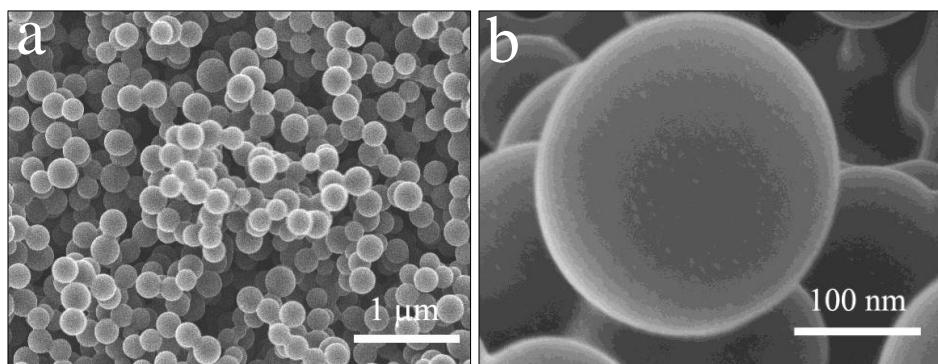
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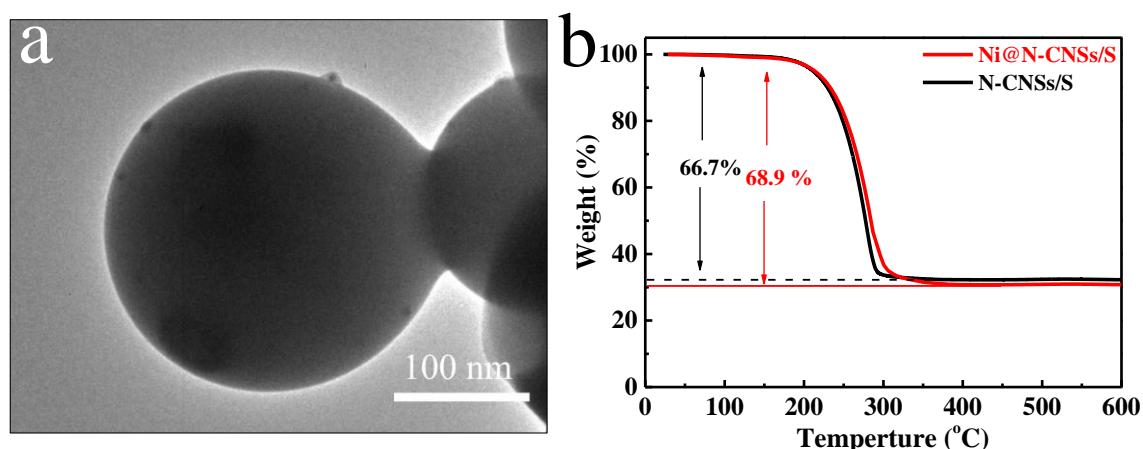
## Supplementary Figures and Table



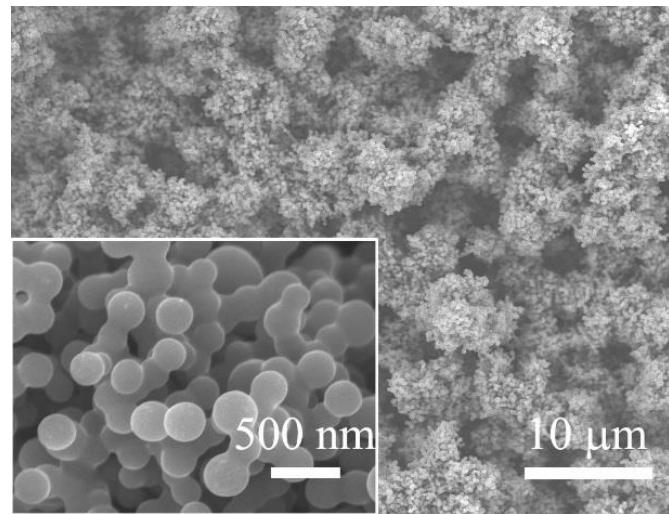
**Fig. S1** Adsorption-desorption isothermal curves of N-CNSs films (pore size distribution in inset)



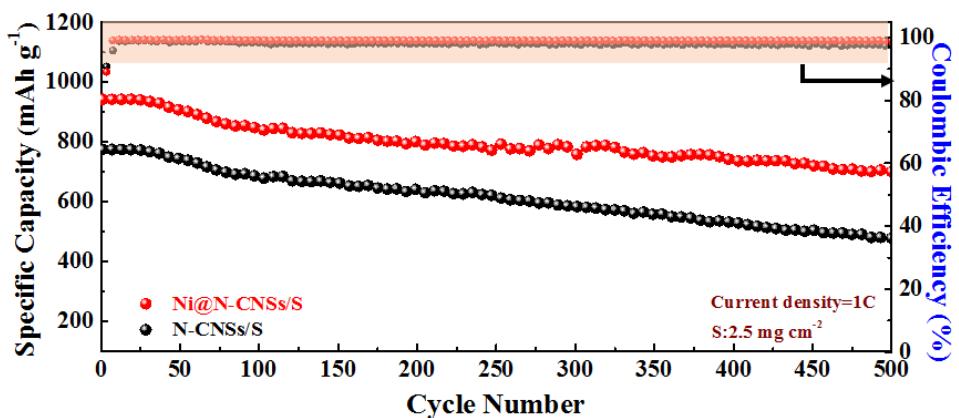
**Fig. S2 a, b** SEM images N-CNSs/S films



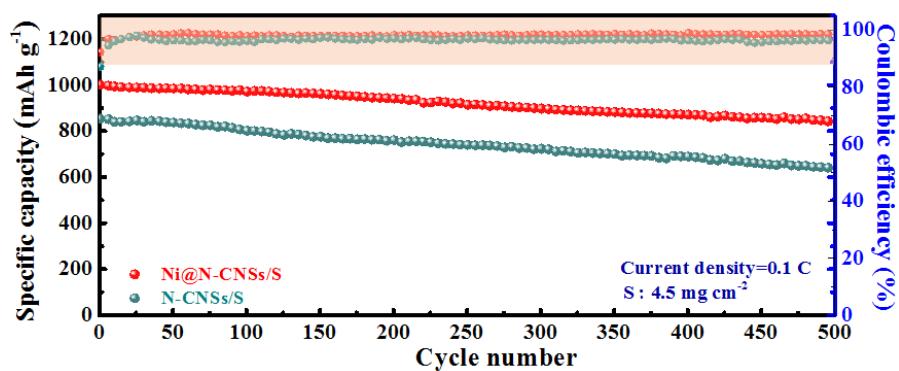
**Fig. S3 a** TEM images N-CNSs/S. **b** TGA curves of Ni@N-CNSs/S and N-CNSs/S electrodes



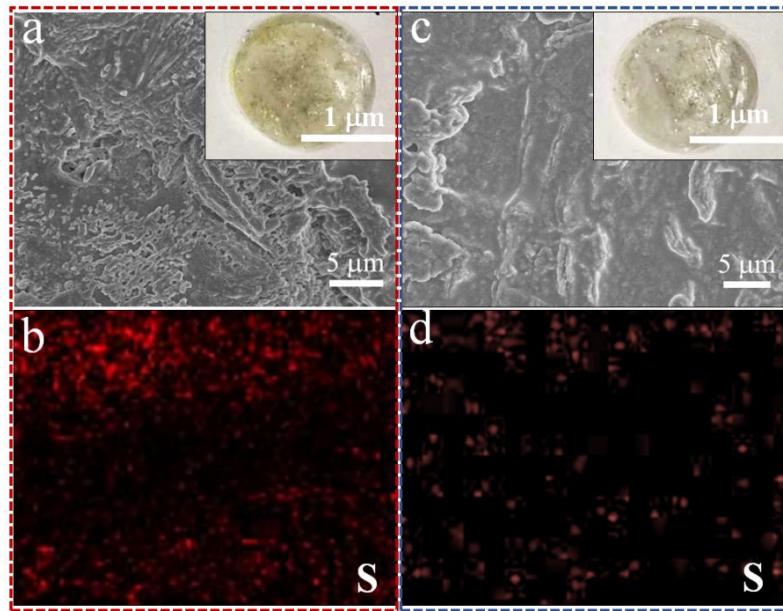
**Fig. S4** SEM image of Ni@N-CNSs/S electrode after 200 cycles at 0.1 C



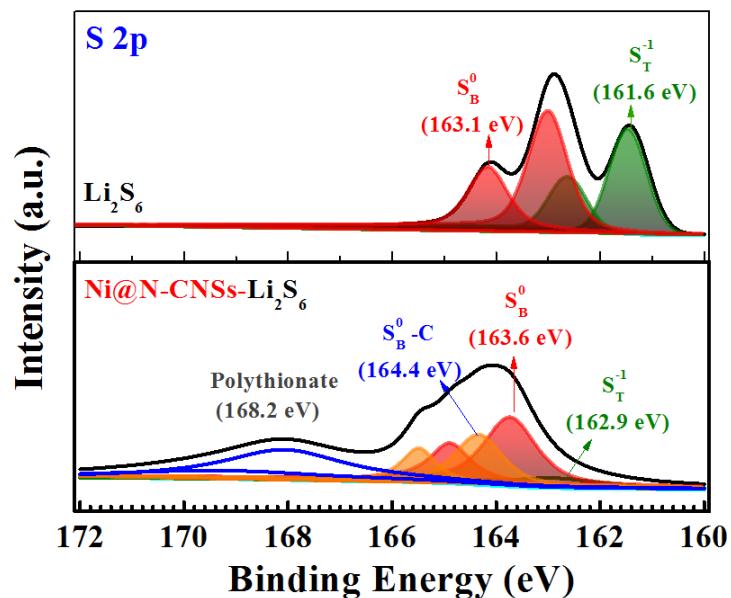
**Fig. S5** Cycling performance and coulombic efficiency of N-CNSs/S and Ni@N-CNSs/S electrodes with S mass of  $2.5 \text{ mg cm}^{-2}$  at 1 C



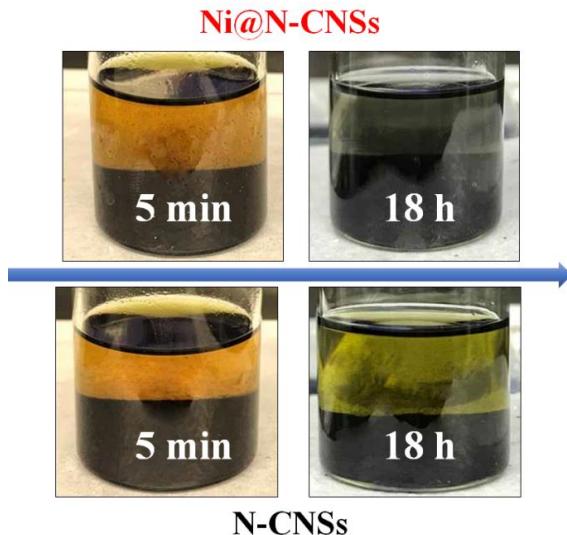
**Fig. S6** Cycling performance and coulombic efficiency of N-CNSs/S and Ni@N-CNSs/S electrodes with S mass of  $4.5 \text{ mg cm}^{-2}$  at 0.1 C



**Fig. S7** SEM image and mapping image of S for cycled lithium anodes after 100 cycles at 0.1 C: **a, b** Cycled lithium anode with N-CNSs/S; **c, d** Cycled lithium anode with Ni@N-CNSs/S cathode (digital photos of cycled separators in inset)



**Fig. S8** XPS analysis of pristine  $\text{Li}_2\text{S}_6$  and  $\text{Ni@N-CNSs-Li}_2\text{S}_6$  samples: S 2p spectra

**Fig. S9** Adsorption test for  $\text{Li}_2\text{S}_6$  solution with Ni@N-CNSs and N-CNSs**Table S1** Electrochemical performance of other carbon/S composite cathodes

Samples	Initial capacity	Cyclability ( $\text{mAh g}^{-1}$ )
CNSs/S powder [S1]	1031 $\text{mAh g}^{-1}$ at 0.5 C	477 $\text{mAh g}^{-1}$ after 100 cycles at 0.5 C
Porous CNSs/S powder [S2]	1150 $\text{mAh g}^{-1}$ at 1 C	680 $\text{mAh g}^{-1}$ after 100 cycles at 1 C
CNTs/graphene/S powder [S3]	829 $\text{mAh g}^{-1}$ at 0.5 C	89% retention after 200 cycles at 0.5 C
S/C nanosphere (PSCs) powder [S4]	809 $\text{mAh g}^{-1}$ at 0.5 C	775 $\text{mAh g}^{-1}$ after 200 cycles at 0.5 C
Hollow CNSs/S powder [S5]	$\sim$ 1318 $\text{mA h g}^{-1}$ at 0.5 C	$\sim$ 700 $\text{mAh g}^{-1}$ after 200 cycles at 0.5 C
TiO@CNSs/S powder [S6]	1285 $\text{mAh g}^{-1}$ at 0.1 C	76% retention after 200 cycles at 0.1 C
Yolk–shell CNSs/S powder [S7]	1106 $\text{mAh g}^{-1}$ at 0.05 C	832 $\text{mAh g}^{-1}$ after 200 cycles at 0.05 C
CNSs-trithiocyanuric acid /S powder [S8]	1227 $\text{mAh g}^{-1}$ at 0.2 C	443 $\text{mAh g}^{-1}$ after 200 cycles at 0.2 C
S@C@MnO <sub>2</sub> /S powder [S9]	1345 $\text{mAh g}^{-1}$ at 0.1 C	1043 $\text{mAh g}^{-1}$ after 200 cycles at 0.1 C
N-CNSs@MnO <sub>2</sub> /S powder [S10]	1249 $\text{mAh g}^{-1}$ at 0.5 C	1110 $\text{mAh g}^{-1}$ after 200 cycles at 0.1 C
<b>This work</b>	<b>1350 <math>\text{mAh g}^{-1}</math> at 0.1C</b>	<b>1175 <math>\text{mAh g}^{-1}</math> after 200 cycles at 0.1 C</b>

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