Supporting Information

High Energy Density Li_2S@C Nanocomposite Cathode with a Nitrogen-doped Carbon Nanotube Top Current Collector

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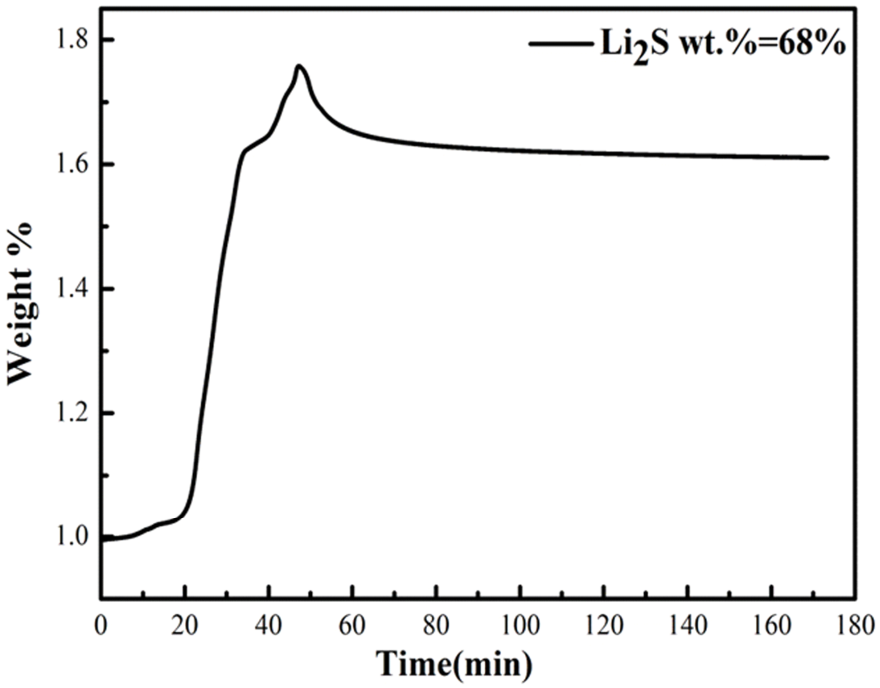

Figure S1. TGA curve of Li_2S@C composites heating in air.

wt % (Li_2S) = wt % (Li_2SO_4) × M (Li_2S) / M (Li_2SO_4)

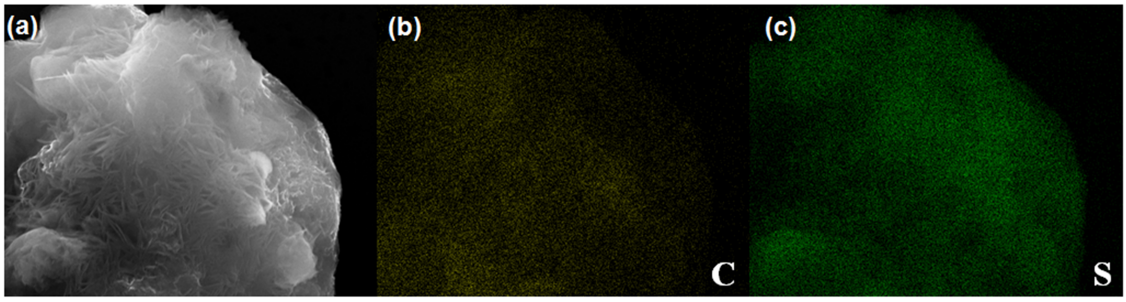

Figure S2. (a) SEM image of Li_2S@C composites; (b) elemental mapping results of C; (c) elemental mapping results of S.

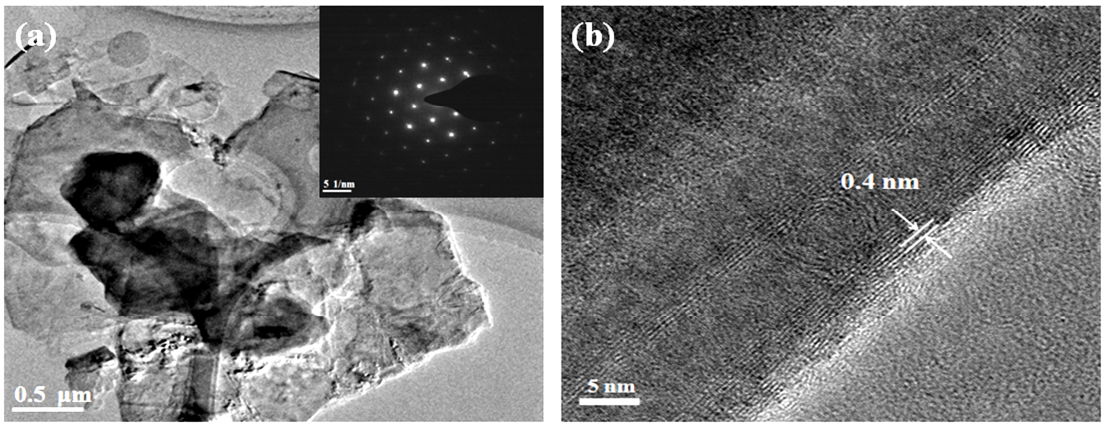

Figure S3. (a) A representative FETEM image of C_{AG} and SAED pattern of C_{AG}; (b) HRTEM image of C_{AG}.

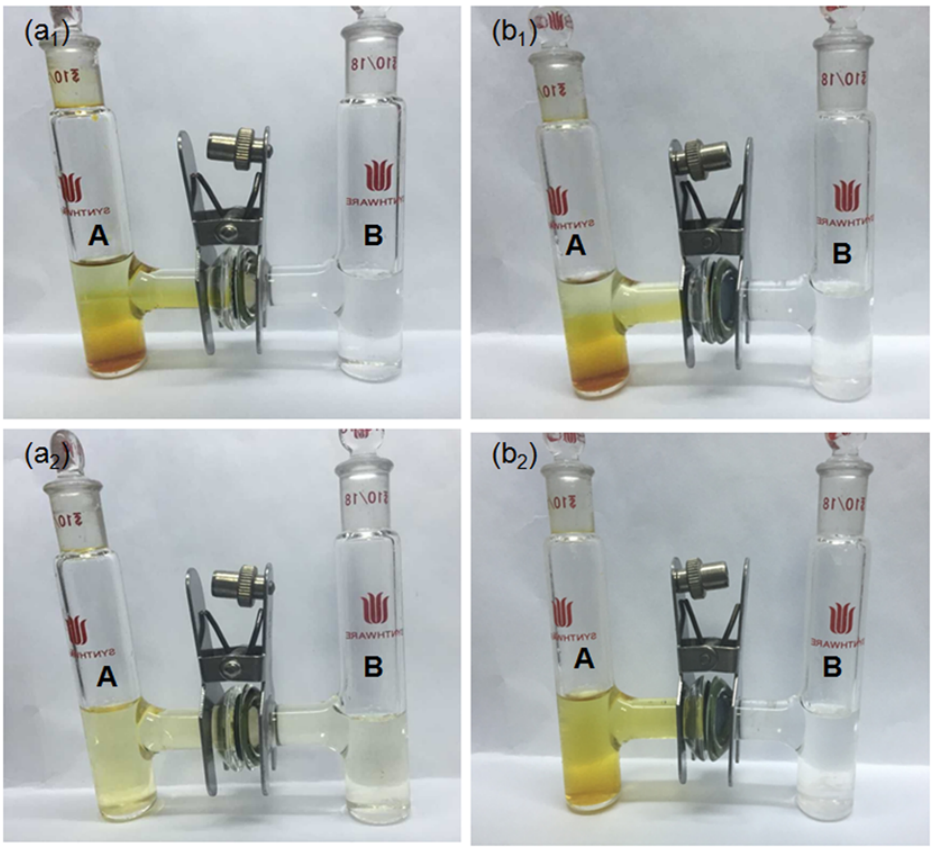

Figure S4. Photos of the H-type glass containers with "side A" containing electrolyte with Li_2S_8 and "side B" containing pure electrolyte: (a_1) pure separator at 0 min; (a_2) pure separator after 20h; (b_1) pure separator with N-CNT film at 0 min; (b_2) pure separator with N-CNT film after 20 h.

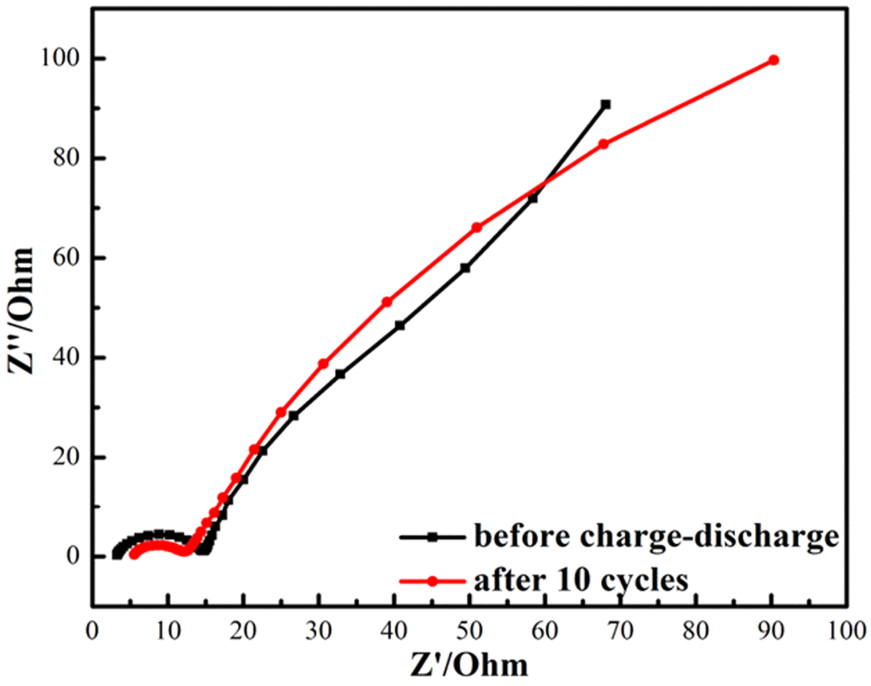

Figure S5. Electrochemical impedance spectra of Li_2S@C composites with N-CNT top current collector.

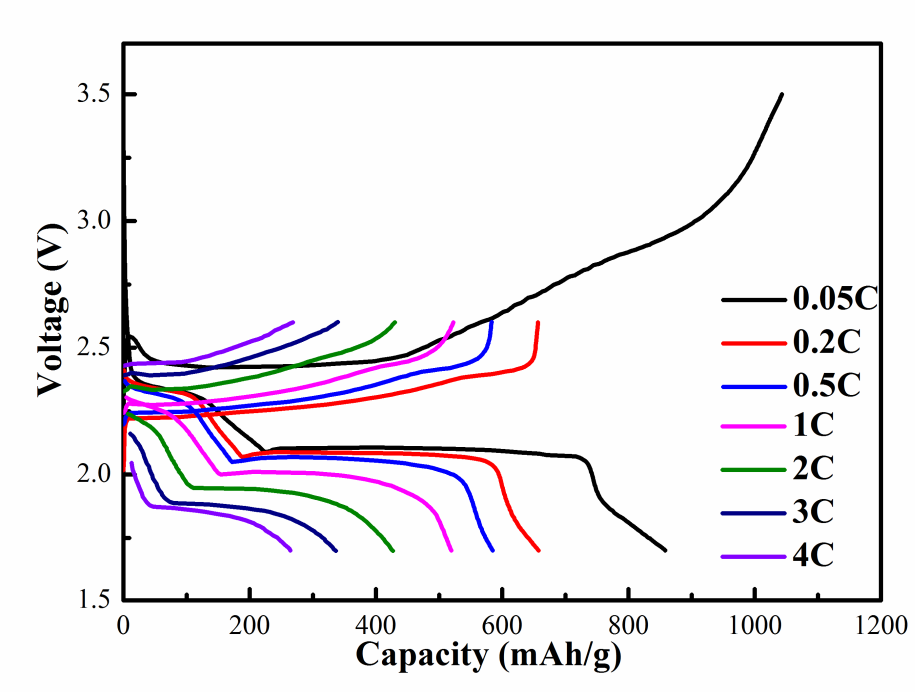

Figure S6. Voltage-capacity profiles of the top current collector cell with Li_2S@C cathode at various discharge/charge current rates.

The calculation method of Li_2S wt % and S wt % in electrode:

$$Li_2S\ \text{wt\% in electrode} = \frac{\text{areal weight of } Li_2S}{\frac{\text{areal weight of } Li_2S}{Li_2S\ \text{wt\% in electrode materials}} + \text{areal weight of Al}}$$

$$\text{areal weight of S} = \text{areal weight of } Li_2S \times 32 / 46$$

$$\text{areal weight of (C+B)} = \frac{\text{areal weight of } Li_2S}{Li_2S\ \text{wt\% in electrode materials}} - \text{areal weight of } Li_2S$$

$$\text{S wt\% in electrode} = \frac{\text{areal weight of S}}{\text{areal weight of (C+B)} + \text{areal weight of S} + \text{areal weight of Al}}$$

In equations, C means conductive carbon and B means binders.

The specific energy of electrode was calculated by:

$$\text{Specific energy of electrode} = \text{Specific capacity of electrode} \times 2.1$$

Table S1 The specific capacity and energy density calculated based on the total electrode for Li/Li_2S cells.

	Li_2S areal Weight (mg/cm^2)	*Li_2S wt % in composite (wt %)*	*Li_2S wt % in cathode materials [a] (wt %)*	*Li_2S wt % in total electrode [b] (wt %)*	*Initial specific capacity of Li_2S (mAh/g Li_2S) at specific current for cycling*	*Initial specific capacity of electrode (mAh/g electrode)*	*initial specific energy of electrode (Wh/kg electrode)*
$Li_2S@C_{(BM)}$ [9]	**5.78**	**67.5**	**50.6**	**43.47**	**342 at 0.5C**	**148.6**	**312**
$Li_2S@C_{(NF)}$ [37]	**1.5**	**72.2**	**60**	**20**	**620 at 0.5C**	**124**	**260**
$Li_2S@C_{(polypyrole)}$ [41]	**1**	**86**	**51.6**	**16.4**	**670.5 at 0.5C**	**110**	**230.7**
$Li_2S@C_{(RF\ gel)}$ [38]	**0.54**	**62**	**49**	**11.6**	**330 at 0.5C**	**38.3**	**80.4**
In-stiu TG-Li_2S [47]	**1.3**	**67**	**53**	**18.9**	**750 at 0.5C**	**142**	**298**
$Li_2S@C_{(CB)}$ [48]	**4**	**81**	**78**	**41.7**	**700 at 0.1C**	**292**	**613**
Li_2S@C(our work)	**3~4**	**68**	**68**	**49**	**667 at 0.5C**	**327**	**686**

a. Cathode materials include active materials, binders and conductive additives;

b. Total electrode includes current collector and cathode materials; and the area weight with 4.2 mg/cm^2 of Al current collector was used here to calculate Li_2S wt% in total cathode in these compared works.

Table S2 Comparison of the sulfur weight percentage, the capacity of Li_2S composite cathodes was converted to sulfur. The specific capacity and specific energy calculated based on the total electrode for our cells, several representative solid sulfur cathodes, solid Li_2S cathodes and liquid polysulfide cathodes.

	S areal Weight (mg/cm^2)	*S wt % in composite (wt %)*	*S wt % in cathode materials [a](wt %)*	*S wt % in total electrode [b](wt %)*	*Initial specific capacity of S (mAh/g S) at specific current for cycling*	*Initial specific capacity of electrode (mAh/g electrode)*	*initial specific energy of electrode (Wh/kg electrode)*
S/CMK3[42]	**1.13**	**70**	**58.8**	**19.1**	**1005 at 0.1C**	**191.9**	**403.1**
S/CNT@MPC[43]	**1**	**40**	**32**	**14.0**	**1120 at 0.5C**	**156.8**	**329.3**
GO/S/CTAB[7]	**0.8**	**80**	**56**	**14.7**	**860 at 1C**	**126.4**	**265.5**
S@PVP[44]	**1**	**70**	**49**	**16.5**	**990 at 0.5C**	**163.4**	**343.0**
Liquid Li_2S_6 [45]	**~1.5**	**40.6 [c]**	**40.6 [c]**	**30.8**	**1224 at 0.5C**	**377.4**	**793**
$Li_2S@C_{(BM)}$[9]	**4.05**	**47**	**35.2**	**36.9**	**488.6 at 0.5C**	**180.2**	**378.6**
$Li_2S@C_{(NF)}$[37]	**1.04**	**50**	**41.7**	**18**	**886 at 0.5C**	**159.5**	**335**
$Li_2S@C_{(polypyrole)}$[41]	**0.7**	**60**	**36**	**12**	**957.9 at 0.5C**	**115**	**241**

$Li_2S@C_{(RF\ gel)}$[38]	**0.38**	**43**	**34**	**9**	**471 at 0.5C**	**42.4**	**89**
S/PNCF[46]	**1.9**	**42.8**	**42.8**	**42.8**	**988 at 0.5C**	**422.8**	**888**
In-stiu TG-Li_2S[47]	**0.91**	**47**	**37**	**14**	**1071 at 0.5C**	**150**	**315**
$Li_2S@C_{(CB)}$[48]	**2.8**	**56.7**	**54.6**	**31.7**	**1000 at 0.1C**	**317**	**666**
Li_2S@C(our work)	**2.1~2.8**	**47**	**47**	**40.2**	**953 at 0.5C**	**383**	**804**

a. Cathode materials include active materials, binders and conductive additives;

b. Total electrode includes current collector and cathode materials; and the area weight with 4.2 mg/cm^2 of Al current collector was used here to calculate S wt% in total electrode in these compared works.

c. S wt% in liquide electrode= $\frac{\text{areal weight of } Li_2S_6}{\text{areal weight of } Li_2S_6 + \text{areal weight of electrolyte } (\text{in liquide } Li_2S_6)} \times \frac{6M_S}{M_{Li_2S_6}}$

The electrolyte density of 1.805 g/mL was used here to calculate the weight of electrolyte in liquid cathode and the “real” electrolyte part in cells was not counted.