Supporting Information for

# Highly reversible Li/dissolved polysulfide batteries with binder free carbon nanofiber electrodes

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#### **Experiment Section**

**Brief estimation of the porosity of the CNF paper.** Given the density of graphite (~2.3 g cm<sup>-3</sup>) and weight of the circular electrode disk (~3.0 mg),<sup>1</sup> the volume of the circular disk made of solid graphite could be calculated to be 3.0 mg / 2.3 g cm<sup>-3</sup> =  $1.3 \times 10^{-3}$  cm<sup>3</sup>. Given the thickness of the CNF paper (0.4 mm) and the area of the circular electrode disk (1.13 cm<sup>2</sup>), the actual volume of the circular electrode disk could be calculated to be 0.4 mm  $\times 1.13$  cm<sup>2</sup> =  $4.5 \times 10^{-2}$  cm<sup>3</sup>. Therefore, the porosity of the CNF paper could be briefly estimated to be (1 -  $1.3 \times 10^{-3}/4.5 \times 10^{-2}$ )  $\times 100\% = 97\%$ .

### Estimation of the weight of the active materials conserved in the CNF paper

**electrode.** The CNF paper electrode after the  $1^{st}$  discharge was rinsed in a mixture of DOL/DME (vol, 1:1) for 30 min, followed by drying in the glove box for 6 h before weighing. The weight of the active material conserved in the electrode could be obtained by comparing the weights of the CNF paper electrode before and after the  $1^{st}$  discharge, assuming the majority of the polysulfides are converted to Li<sub>2</sub>S. Unreacted polysulfides at the end of the  $1^{st}$  discharge were lost during rinsing, rendering the weight obtained slightly less than the actual weight of the active material conserved in the CNF paper electrode.

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## **Supplemental Figures**



Figure S1. Schematic of the CNF paper/dissolved polysulfide cell.



**Figure S2.** 1<sup>st</sup> discharge voltage profiles of the Li/dissolved polysulfide cells with a CNF paper electrode and a MWCNT paper electrode, cycled at C/5 rate.



**Figure S3.** XRD patterns of the CNF paper electrode after 1<sup>st</sup>, 5<sup>th</sup>, and 10<sup>th</sup> discharge.



**Figure S4.** SEM images of the lithium metal anode (a) before cycling and after the (b)  $1^{st}$ , (c)  $5^{th}$ , and (d)  $10^{th}$  discharge.



**Figure S5.** (a) SEM image of the CNF electrode after the  $2^{nd}$  charge in the capacitycontrolled one-plateau cycling, (b) EDS sulfur and carbon mappings for the region shown in (a), with the sulfur signal in red and carbon signal in green, (c) magnified image of the region shown in (a), and (d) magnified image of the region shown in Fig. 3c.

### REFERENCES

1 J. A. Dean (ed), *Lange's Handbook of Chemistry*, McGraw-Hill, New York, 15th edn., 1999.