3D interconnected porous carbon nanosheet/carbon nanotube as polysulfides reservoir for high performance lithium-sulfur batteries

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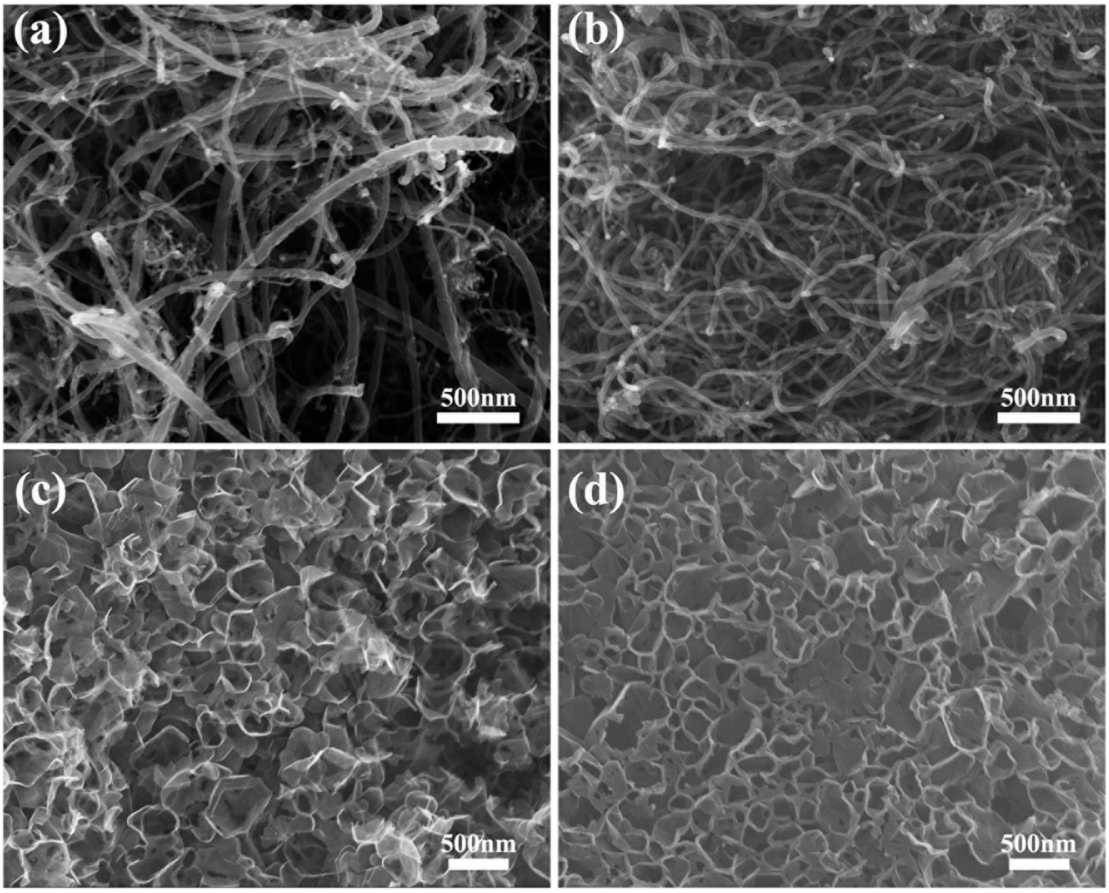

Fig. S1. SEM images of CNT (a) and S-CNT composite (b); SEM images of PC (c) and S-PC composite (d).

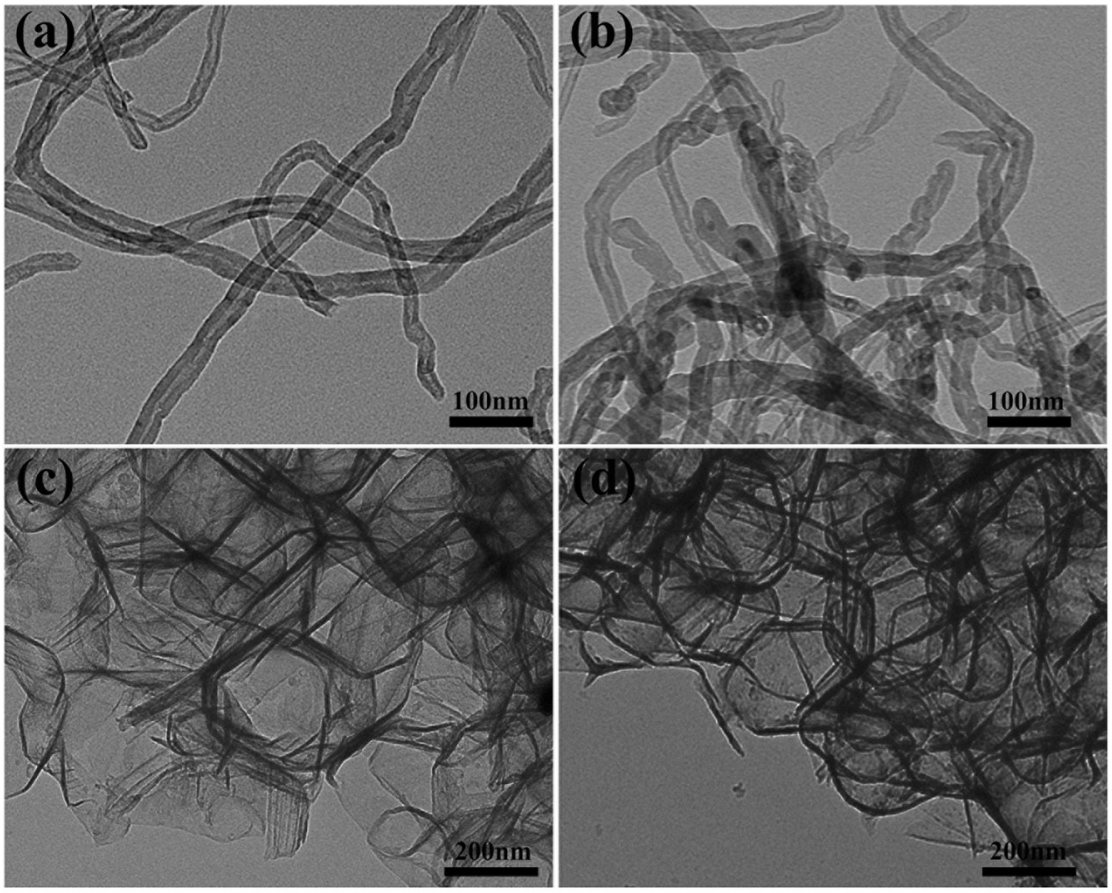

Fig. S2. TEM images of CNT (a) and S-CNT composite (b); TEM images of PC (c) and S-PC composite (d).

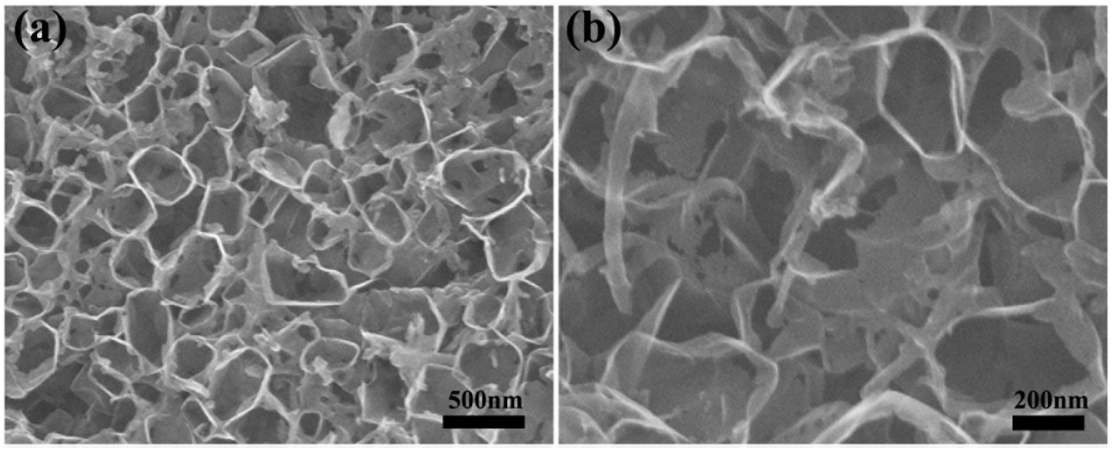

Fig. S3. SEM images of PC/CNT composite at different magnifications: (a) at a low magnification; (b) at a high magnification

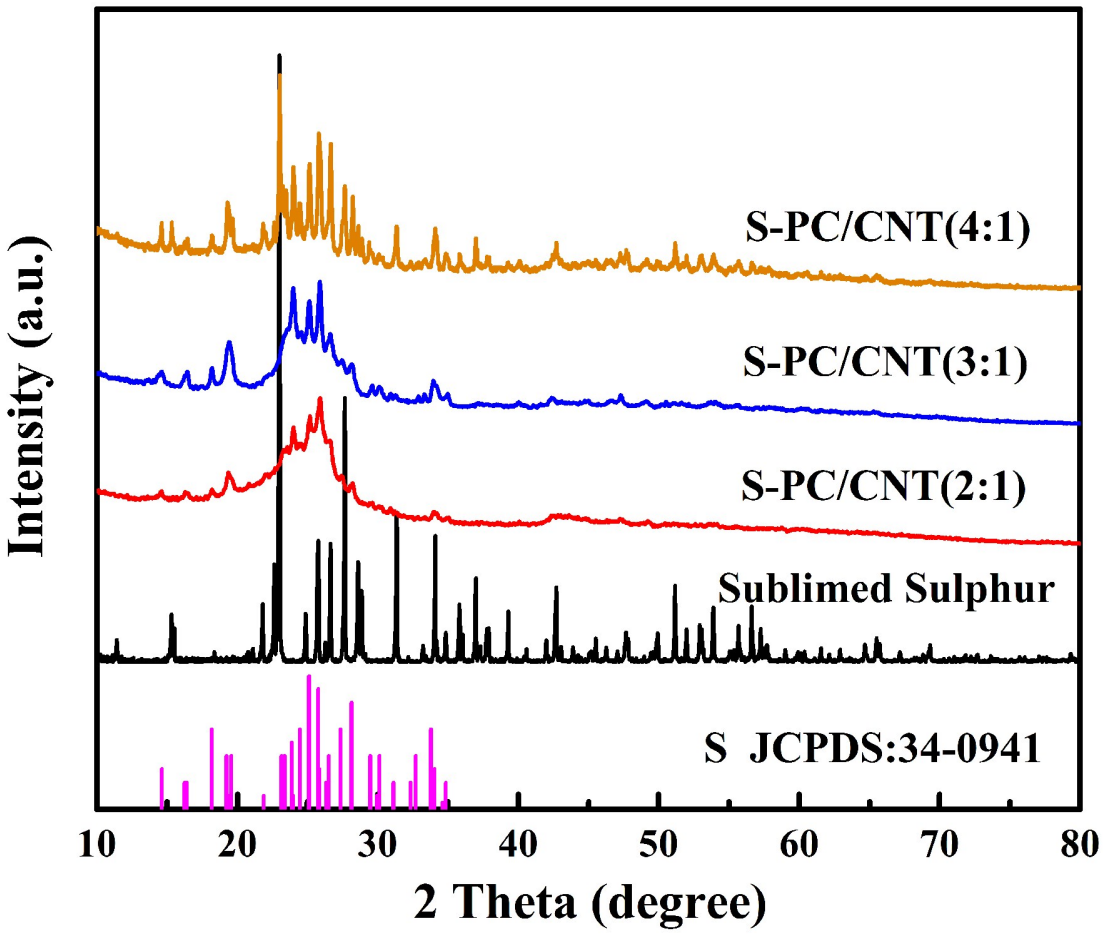

Fig. S4. XRD patterns of S-PC/CNT with different sulfur loading.

Generally, sublimed sulphur corresponds to the orthorhombic phase sulfur (JCPDS No. 08-0247). After sulfur loading, some peaks of S-PC/CNT (2:1 or 3:1) corresponds to the monoclinic phase sulfur (JCPDS No. 34-0941). It is observed that most of the sulfur exist in amorphous state in the hierarchical pores of PC/CNT and a small amount of orthorhombic phase sulfur are converted into monoclinic phase sulfur.

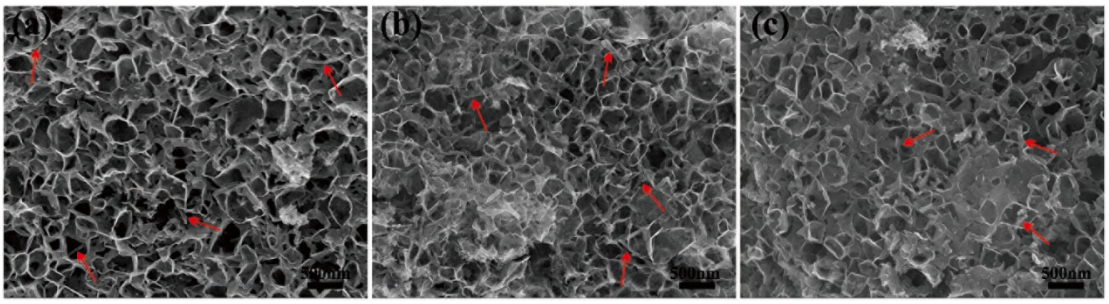

Fig. S5. SEM images of S-PC/CNT composite with different sulfur loading: (a) S-PC/CNT (2:1); (b) S-PC/CNT (3:1); (c) S-PC/CNT (4:1). The red arrows point to CNT.

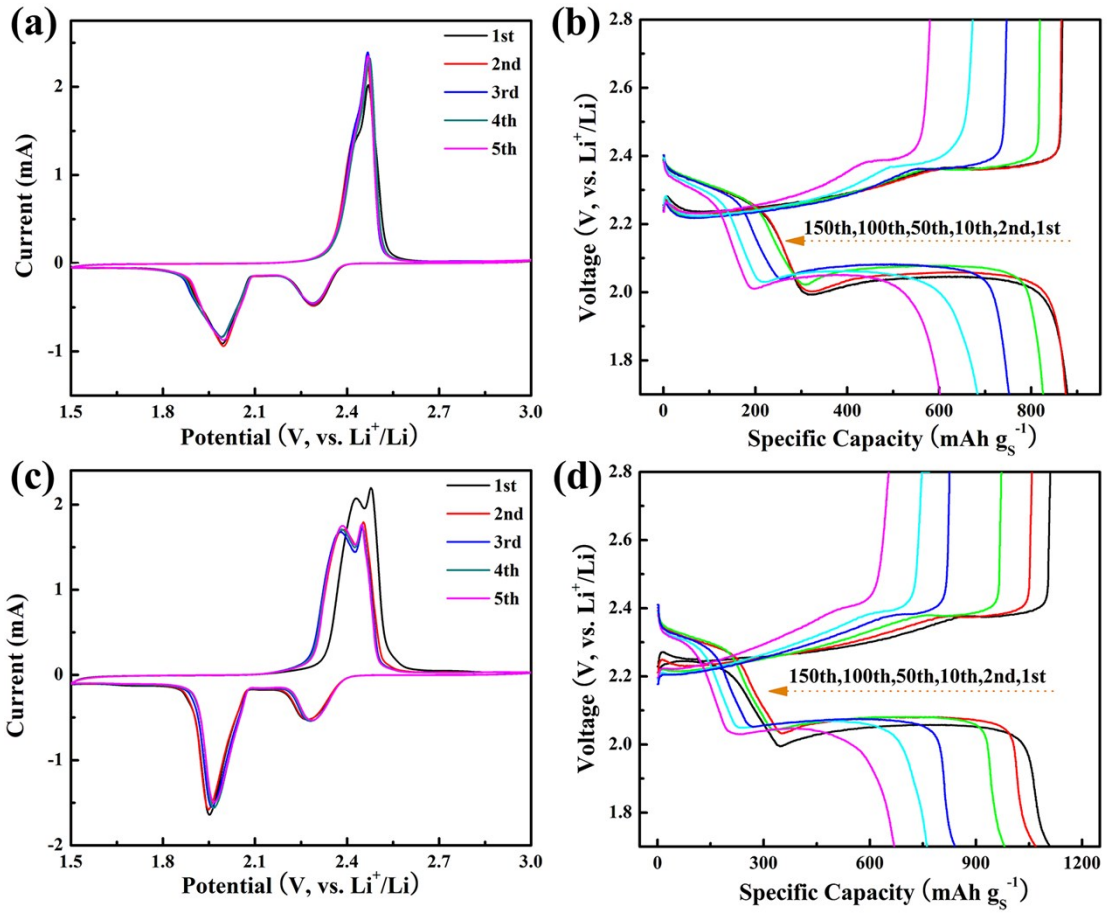

Fig. S6. CV curves and charge/discharge profiles at 0.5 C of S-CNT cathode (a, b); CV curves and charge/discharge profiles at 0.5 C of S-PC cathode (c, d).

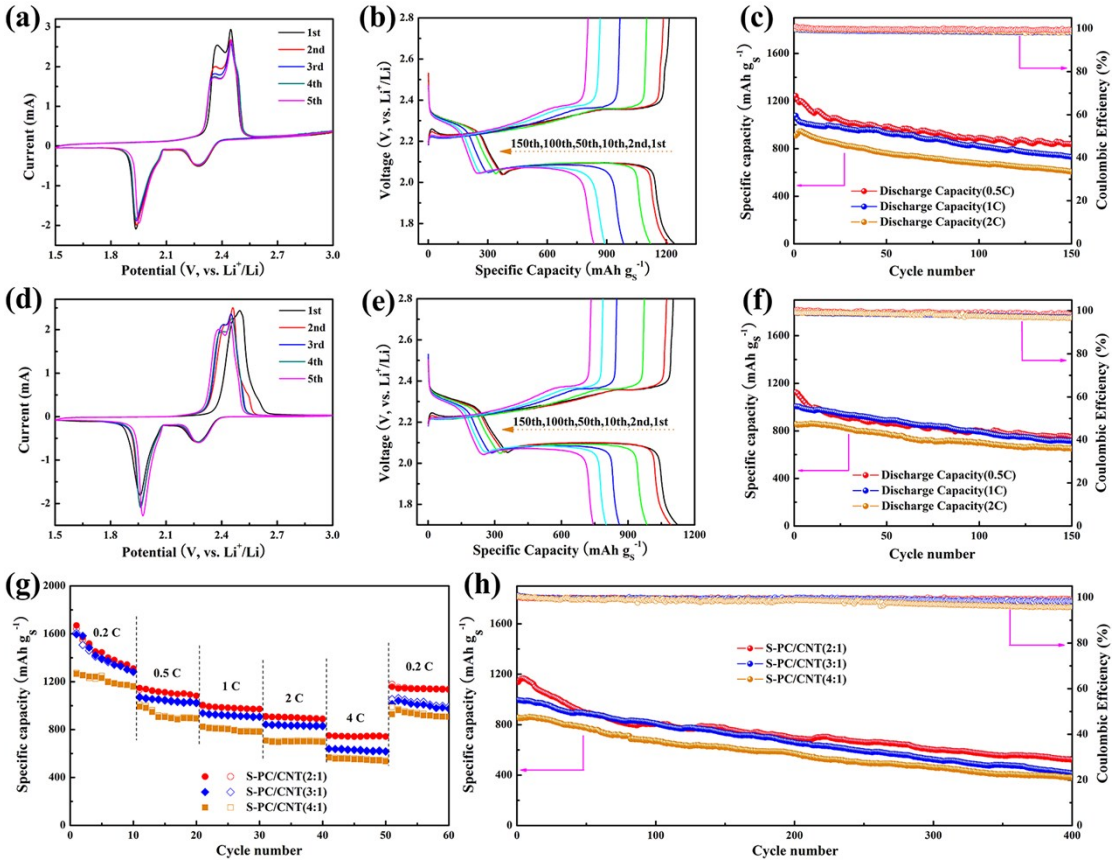

Fig. S7. CV curves and charge/discharge profiles at 0.5 C of S-PC/CNT (3:1) cathode (a, b); Cycling performance of S-PC/CNT (3:1) cathode at charge/discharge rates of 0.5, 1, 2 C (c); CV curves and charge/discharge profiles at 0.5 C of S-PC/CNT (4:1) cathode (d, e); Cycling performance of S-PC/CNT (4:1) cathode at charge/discharge rates of 0.5, 1, 2 C (f); Rate performance of S-PC/CNT cathodes with different sulfur loading (g); Long-term cycling stability of S-PC/CNT cathodes with different sulfur loading at 2 C (h).

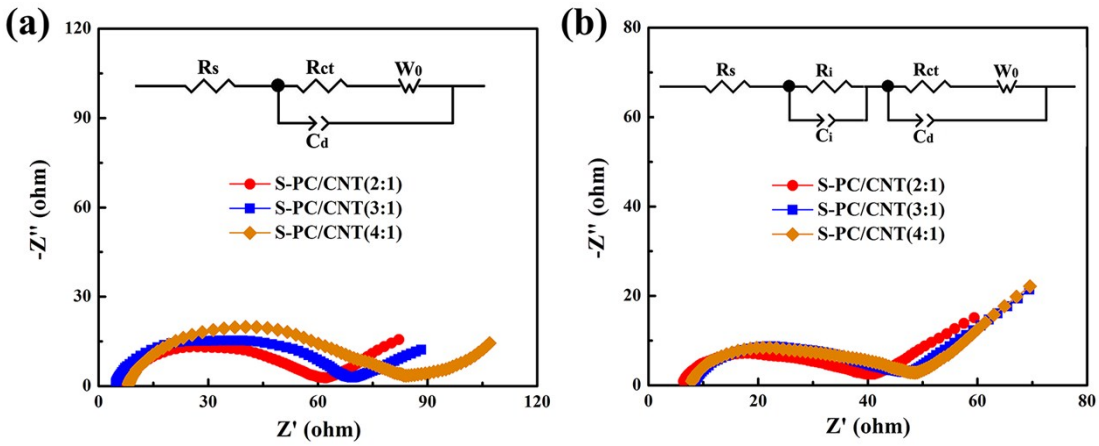

Fig. S8. Nyquist plots of S-PC/CNT cathodes with different sulfur loading (a) before cycling (b) after 200 cycles.

As shown in Fig. S7(a), it is found that the impedance spectra before cycling composed of semicircle in the high frequency region and inclined line in the low frequency region. The semicircle in the high frequency region is associated with charge-transfer processes while the inclined line is related to a combined effect of the diffusion of lithium ions. After cycling, the Nyquist plots of samples display two depressed semicircle in the high frequency region (corresponding to the interphase contact resistance on the surfaces of two electrodes, which is caused by the formation of Li_2S/Li_2S_2 on the carbon matrix at cathode) and middle frequency region (corresponding to the charge transfer resistance). The equivalent circuit models for analysing the impedance spectra are shown in the insets of Fig. S7. The R_s represents the bulk resistance including resistance of the electrolyte, separator and electrodes, *Rct* is the charge transfer resistance, CPE is a constant phase element which is used instead of capacitance, R_i is the interfacial resistance of SEI, and W_0 is the Warburg impedance .

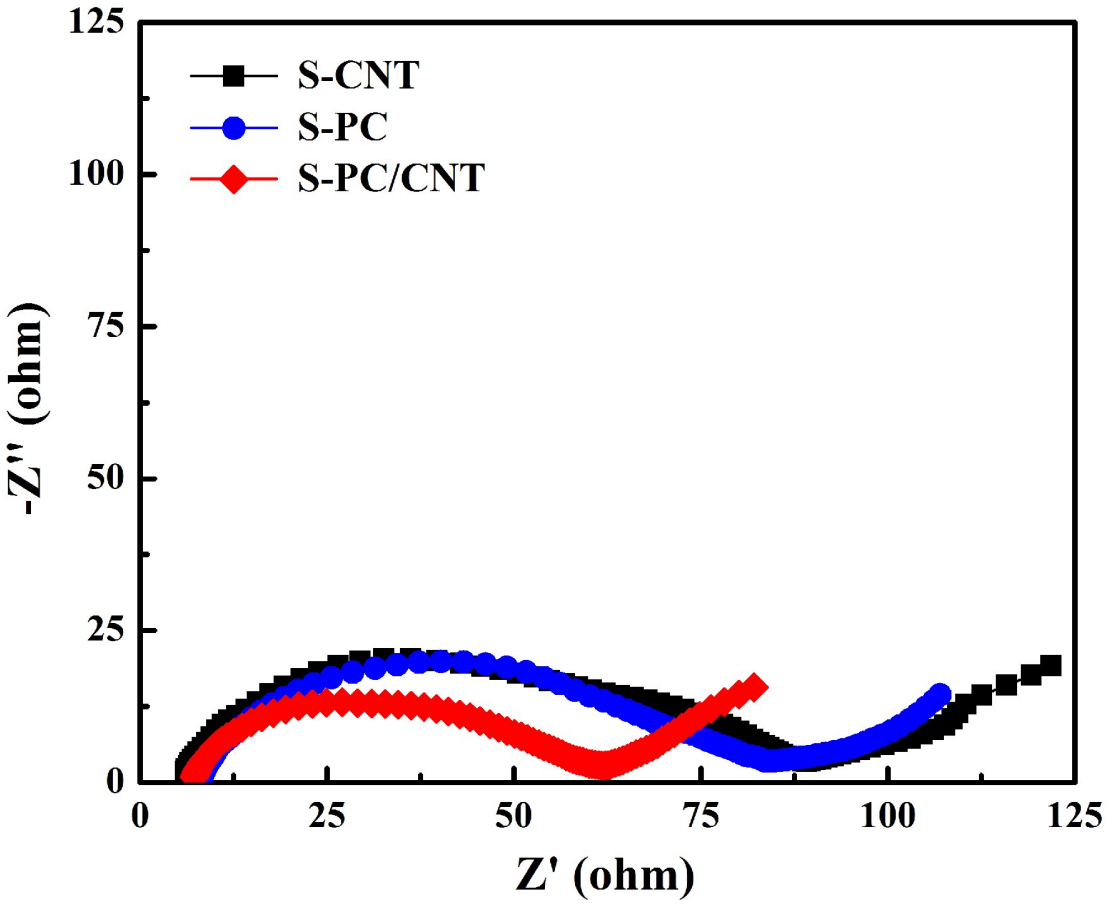

Fig. S9. Nyquist plots of different composite cathodes before cycling.

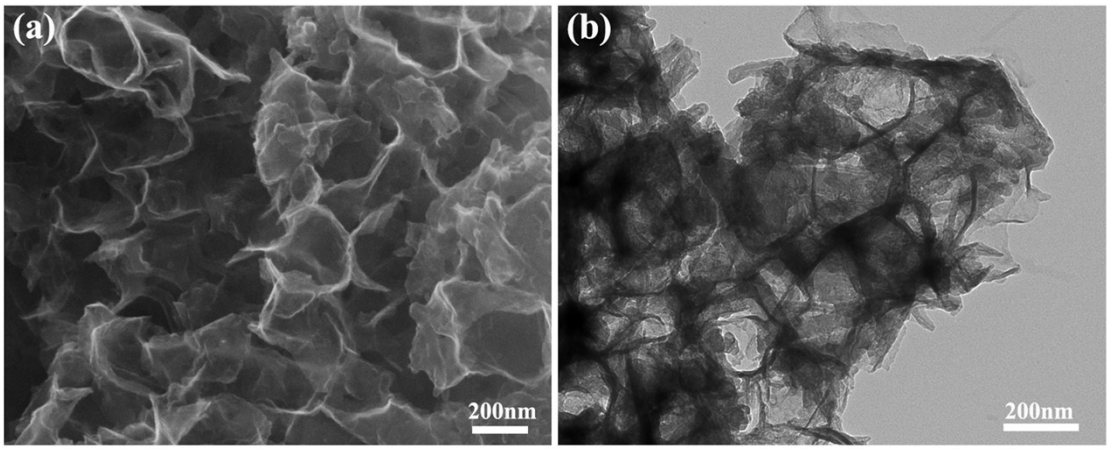

Fig. S10. SEM (a) and TEM (b) images of S-PC/CNT cathode after 200 charge/discharge cycles at 2 C.

Table S1. Electric conductivity of CNT, PC, PC/CNT and other carbon samples measured at 7 MPa.

Samples	CNT	PC	PC/CNT	Commercial acetylene black	Graphene
Electric conductivity (S cm^{-1})	27.6	15.5	19.8	20.2	36.1

Table S2. Summarization of the electrochemical performances of S-PC/CNT composite cathode with different sulfur loading for Li–S batteries.

Rate	S-PC/CNT (2:1)			S-PC/CNT (3:1)			S-PC/CNT (4:1)		
	Capacity ($mAh\ g^{-1}$)		Capacity retention	Capacity ($mAh\ g^{-1}$)		Capacity retention	Capacity ($mAh\ g^{-1}$)		Capacity retention
	1st cycle	200th cycle		1st cycle	200th cycle		1st cycle	200th cycle	
0.5 C	1485.4	825.1	55.5%	1243.0	777.7	62.6%	1123.9	689.7	61.4%
1 C	1300.3	821.0	63.1%	1076.8	734.1	68.2%	1007.0	659.8	65.9%
2 C	1138.0	693.2	60.9%	995.7	665.8	66.9%	857.2	596.5	69.6%

Table S3. Impedence parameters of S-PC/CNT composite cathode with different sulfur loading calculated from equivalent circuit model.

Samples	Cycle number	Resistance		
		$R_s(\Omega)$	$R_i(\Omega)$	$R_{ct}(\Omega)_t$
S-PC/CNT(2:1)	Before cycling	5.63	—	53.62
	After 200 cycles	6.02	18.95	14.83
S-PC/CNT(3:1)	Before cycling	5.47	—	63.96
	After 200 cycles	7.68	18.64	20.81
S-PC/CNT(4:1)	Before cycling	5.90	—	78.28
	After 200 cycles	7.31	18.56	21.59