

Supporting Information



Fig. S1: (a) SEM of PUF. (b) the quality of Al current collector and (c) NCF current collector. (d) TEM of NCF.

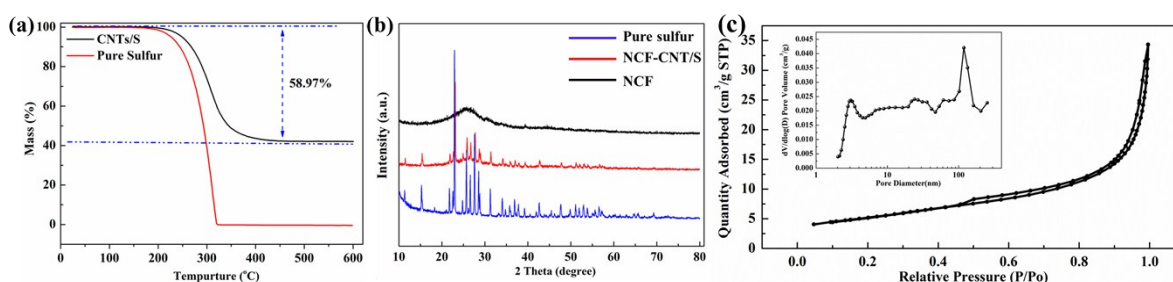


Fig. S2: (a) TG of CNT/S composites and pure sulfur. (b) XRD of pure sulfur, NCF and NCF-CNT/S cathode. (c) N₂ adsorption-desorption isotherms of NCF-CNT/S (the inset of pore diameter distribution curves).

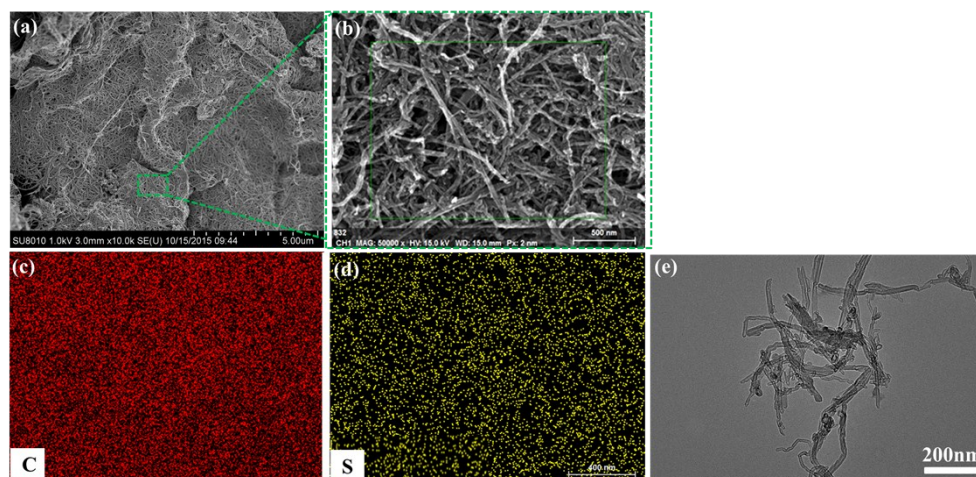


Fig. S3 (a) FESEM of CNT/S composites. (b) FESEM of the selection area. (c)-(d) EDS of CNT/S composites. (e) TEM images of CNT/S composites.

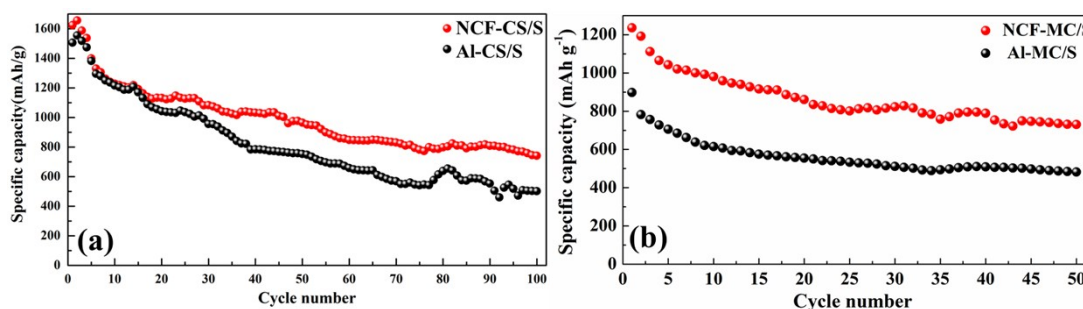


Fig. S4: Cycle performance of (a) the NCF-CS/S cathodes and Al-CS/S. (b) NCF-MC/S cathode and Al-MC/S

The carbon hollow spheres (CS) were prepared as previously reported.¹ Briefly, first, 0.08 g sodium dodecyl sulfate (SDS) and 3.5 g glucose were dissolved in 35 ml water and keep stirred for 10min to form a homogeneous solution. Then, transferred the solution into a 60 ml Teflon-lined stainless steel autoclave and hydrothermally treated at 160 °C for 16 h. Second, the puce products were centrifuged and washed with distilled water and absolute ethanol several times. The carbon precursor was obtained after oven-drying at 60 °C for 12 h. Finally, the final CS (Fig. S 5a†) was get after carbonized carbon precursor by heating at 800 °C for 2 h under N₂ flow with a heating rate of 5 °C min⁻¹ in a tube furnace.

Equally, the mesoporous carbon (MC) were prepared as previously reported.² 0.1 mol ferric nitrate (Fe(NO₃)₃) and 0.2 mol citric acid were added to 100mL deionized water and stired strongly for 30 min to make sure to form a homogeneous solution. The sol heated at 120 °C in the oven for 24 h, a gel with hierarchical structure was shaped and then calcined at 800 °C at a heating rate of 5 °C min⁻¹ under N₂ atmosphere for 2 h. The grey powder was then neutralized in 1 M H₂SO₄ to thoroughly etch away the decomposition products decomposed from Fe(NO₃)₂. The final MC (Fig. S 5b†) product was collected after filtration, repeated washing with deionized water and vacuum drying.

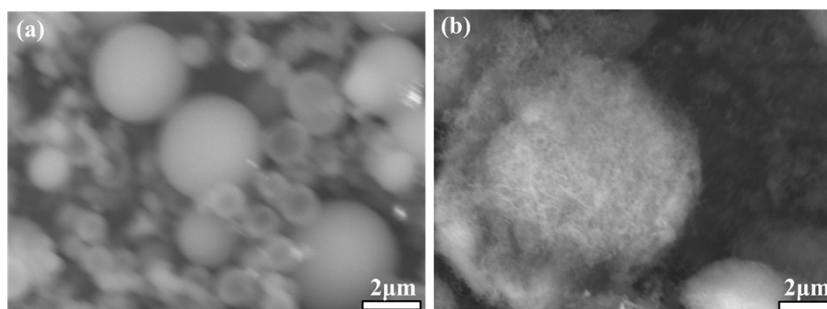


Fig. S5: SEM of (a) CS and (b) MF.

The CS/S composite, MC/S composite and the corrsponding NCF-C/S cathodes were the same to the CNTs/S composite and NCF-CNTs/S cathode, respectively.

Samples	Density (kg/m ³)	Fiber diameter	Mesh diameter	Main application
Nano-foam	6.5-8.5	Dozens of nm	Dozens of nm	Cleaning supplies
High density nano-foam	15-17	Dozens of nm	Dozens of nm	Cleaning supplies
Common Foam	4-12	~5μm	10-30μm	Automobile or cosmetics
This work	~8	~5μm	10-30μm	Construction industry

Table S1. Physical properties and mainly applications of some others PU foam precursor

Samples	Sulfur loading (mg/cm ²)	Rate (C)	Specific capacity (mAh/g)	EV (kWh/L)	reference
3D CNT/graphene/sulfur hybrid sponge	1.0	0.5	1150	2.93	3
3D few-layer graphene foam	2.0	0.5	800	4.08	4
Nano-cellular carbon	2.2	0.5	1200	5.25	5
Pie-like electrode	3.6	0.5	800	5.7	6
Flexible all-carbon electrode	2.3	0.5	1000	5.85	7
Hollow carbon nanofibers	3.5	0.5	900	6.27	8
3D biomass derived active carbon	5.1	0.5	950	9.64	9
Carbon paper	6.7	0.5	800	10.66	10
This work	1.2	0.5	1124	5.37	
	1.8	0.5	1012	7.52	

Table S2. The specific volumetric energy density of 3D electrodes

Cycles	R1	CPE1-T	CPE1-P	R2	CPE2-T	CPE2-P	R3	W1-R	W1-T	W1-P
1	6.04	9.84e-6	0.84	31.56	0.11	0.18	35.54	8.95	3.26	0.31
2	5.73	1.22e-5	0.81	25.22	0.06	0.15	30.11	10.03	3.10	0.37
10	5.27	1.21e-5	0.85	17.45	0.05	0.14	29.36	12.38	2.91	0.31
20	5.24	1.32e-5	0.80	55.70	0.03	0.52	27.67	14.14	4.6	0.44
50	6.29	1.05e-5	0.83	58.98	0.06	0.21	37.80	12.64	0.95	0.35
100	4.32	1.28e-5	0.79	67.23	0.02	0.69	56.50	23.49	0.33	0.44

Table S3. Fitted values of the impedance spectra in Fig. 6c.

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