Electronic Supplementary Information

CNT threading N-doped porous carbon film as binder-free electrode for high-capacity supercapacitor and Li-S battery

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1. Equations for electrochemical calculation:

For supercapacitor, the specific capacitance, energy density and power density can be calculated according to the following equations:

$$Cg = It/(mV) \quad (1)$$

$$E = C_g V^2/(2*4*3.6) \quad (2)$$

$$P = E/t \quad (3)$$

where Cg (F g^{-1}) is the specific capacitance from galvanostatic charge-discharge (GCD) curves, I (A) is the current relating to the voltage V, V (V) is the voltage window, t (s) is the discharge time, m (g) is the mass of the active material, E (E, W h kg^{-1}) is the energy density and P (P, W kg^{-1}) is the power density.

For Li-S battery, the specific capacity is calculated from the charge-discharge curve, and the areal/volumetric capacity is calculated based on the active material (1C=1675 mA g^{-1}).

2. Additional Figures and tables.

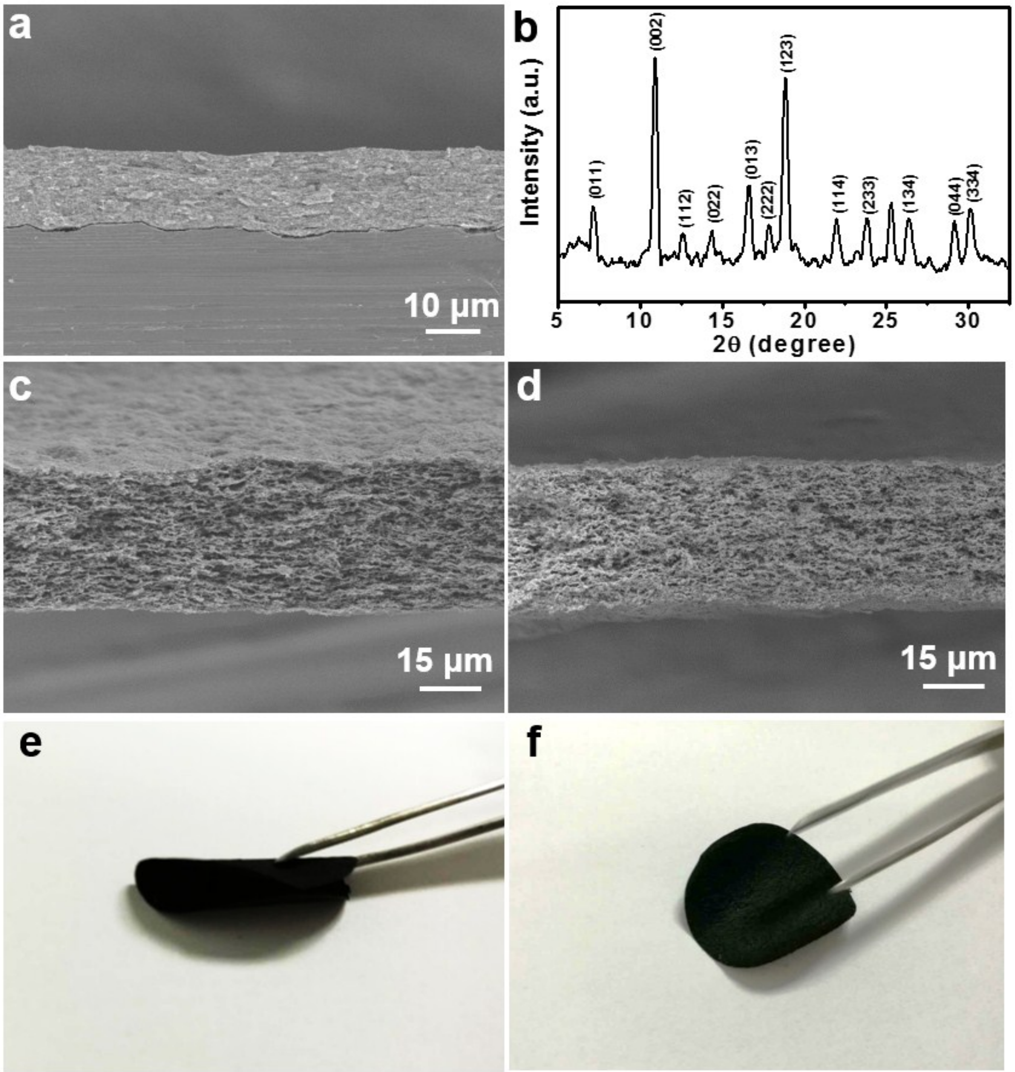

Figure S1. (a), (c) and (d) SEM of ZHN/CNT, CNCF-5/1 and CNCF-15/1, respectively; (b) XRD pattern of ZIF-8/CNT; (e) and (f) digital photograph of bended and released CNCF-10/1.

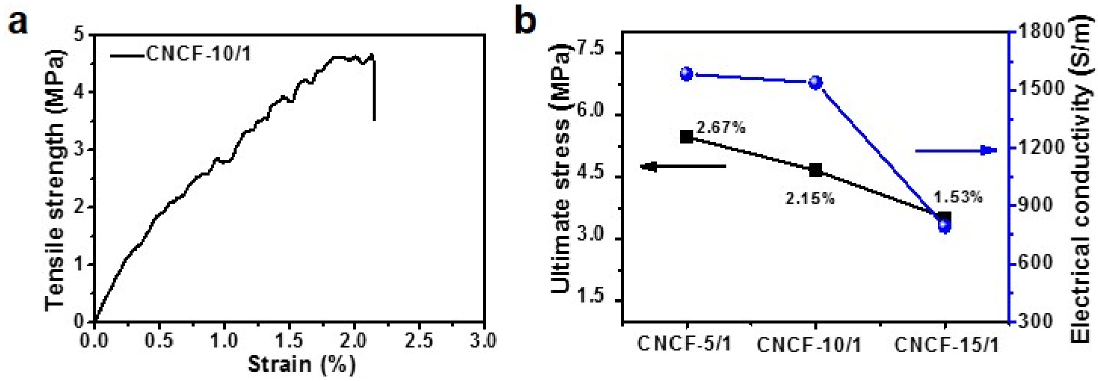

Fig S2. (a) Tensile strength–strain curves of HPCF4; (b) Ultimate stress, strain and electrical conductivity for HPCF1, 2, 3, 4 and 5.

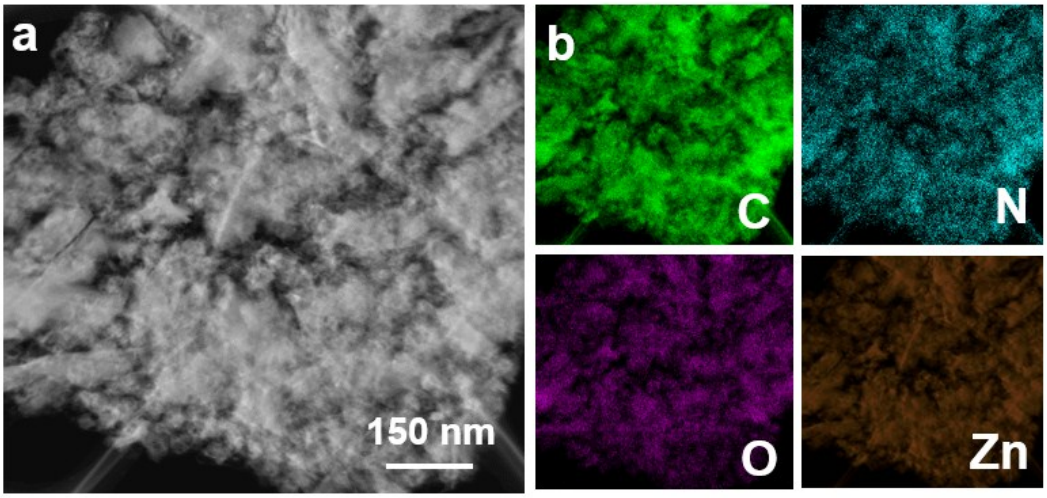

Figure S3. (a) TEM and (b) mapping results of CNCF-10/1@Zn.

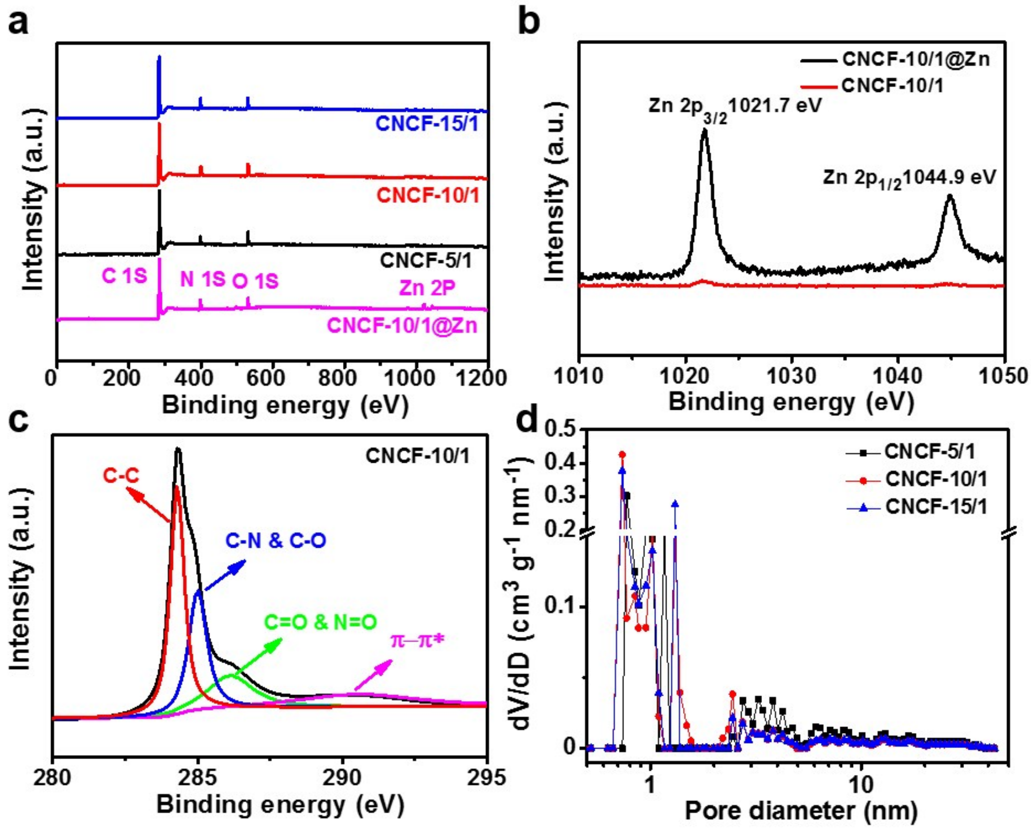

Figure S4. (a) Overview XPS spectra of CNCF-5/1, 10/1, 15/1 and CNCF-10/1@Zn; (b) comparison of Zn 2p spectra between CNCF-10/1@Zn and CNCF-10/1; (c) C 1s spectra of CNCF-10/1; (d) DFT pore-size distribution of CNCF-5/1, 10/1 and 15/1.

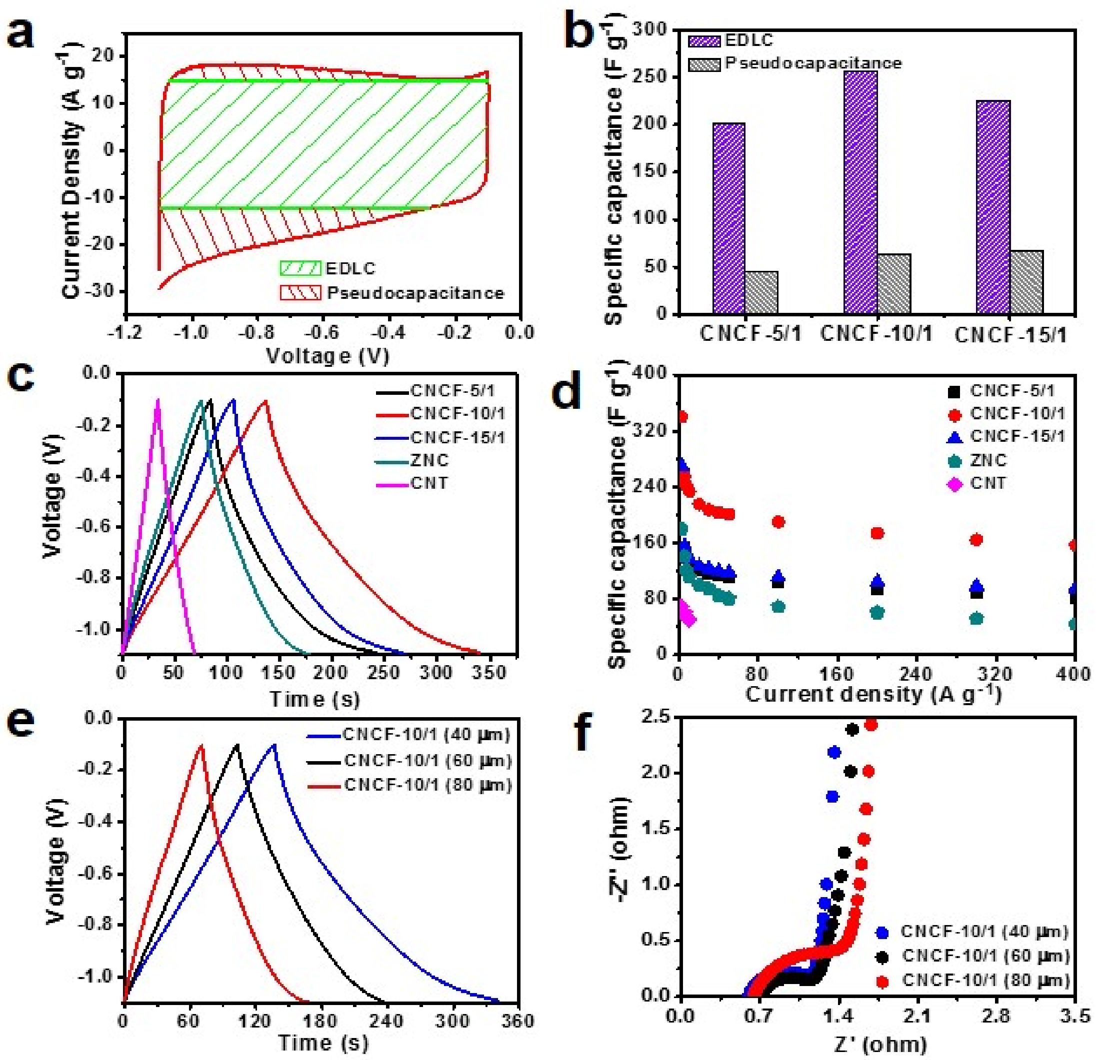

Figure S5. (a) The CV curve of CNCF-10/1 sample divided into EDLC and pseudocapacitance area at 100 mV s^{-1}; (b) the EDLCs and pseudocapacitances of all samples obtained from the CV curves at 100 mV s^{-1}; (c) the GCD curves of the CNCFs, CNT and ZNC at 2 A g^{-1}; (d) specific capacitances at different current densities; (e) the GCD curves of the CNCF-10/1 electrode with different thickness at 2 A g^{-1}; (f) EIS results of the CNCF-10/1 electrode with different thickness.

Table S1. Comparisons of the NPCF-10/1 electrode with MOF derived porous carbon materials in aqueous electrolyte using three-electrode configuration.

Material	Electrolyte	Current density	Cg/ F g^{-1}	Ref
CNT/NPC	0.5 M H_2SO_4	2.5 A g^{-1}	286	1
N-doped carbon/CNTs	1 M Na_2SO_4	1 A g^{-1}	250	2
rGO/CMOF-5	6 M KOH	0.5 A g^{-1}	312	3
3D hybrid porous carbon	6 M KOH	0.5 A g^{-1}	332	4
N-Doped Porous Carbon	6 M KOH	0.5 A g^{-1}	149	5
Hollow carbon nanospheres	1 M H_2SO_4	20 mV s^{-1}	91	6
Hierarchically porous carbons	6 M KOH	1 mV s^{-1}	170	7
PC1000/C	6 M KOH	0.5 A g^{-1}	225	8
N-doped porous carbon	6 M KOH	0.1 A g^{-1}	213.8	9
N-doped porous carbon/CNT	1 M H_2SO_4	5 mV s^{-1}	308	10
CNCF-10/1	**6 M KOH**	**2 A g^{-1}**	**340**	**This work**

Table S2. Comparison of electrochemical performance of flexible and/or self-standing carbon film.

S_{BET}/ m^2 g^{-1}	Electrolyte	Current density	Cg/ F g^{-1}	Ref
656	6 M KOH	0.2 A g^{-1}	105	11
541	1 M H_2SO_4	0.5 A g^{-1}	165	12
--	1 M H_2SO_4	0.2 A g^{-1}	170	13
299.4	6 M KOH	0.5 A g^{-1}	196	14
590	6 M KOH	0.5 A g^{-1}	207	15
--	1 M H_2SO_4	0.4 A g^{-1}	209	16
12.7	1 M H_2SO_4	0.4 A g^{-1}	221.7	17
--	0.5M H_2SO_4	50 mV s^{-1}	223.8	18
113.5	1 M HCl	1 A g^{-1}	251	19
584.5	3M KOH	0.3 A g^{-1}	259	20
814.5	6 M KOH	1 A g^{-1}	267.6	21
550	6 M KOH	10 A g^{-1}	289	22
1223	6 M KOH	200 mV s^{-1}	289	23
119	6 M KOH	5 mV s^{-1}	80	24
691	6 M KOH	1 A g^{-1}	152	25
812.5	6 M KOH	2 A g^{-1}	175	26
1317	6 M KOH	20 A g^{-1}	210	27
679	6 M KOH	0.5 A g^{-1}	255	28
763	6 M KOH	1 A g^{-1}	251	29
803	6 M KOH	1 mV s^{-1}	223.8	30
645.2	**6 M KOH**	**2A g^{-1}**	**340**	**This work**

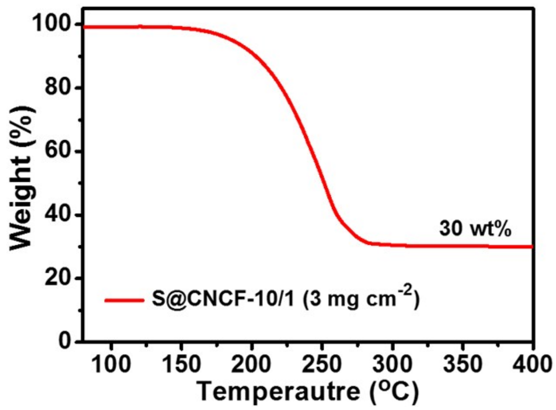

Figure S6. (a) The TGA curve of S@CNCF-10/1 electrode with a sulfur loading of 3 mg cm^{-2} in N_2 with a heating rate of 5 ºC min^{-1}, indicating sulfur content of ~70 wt%.

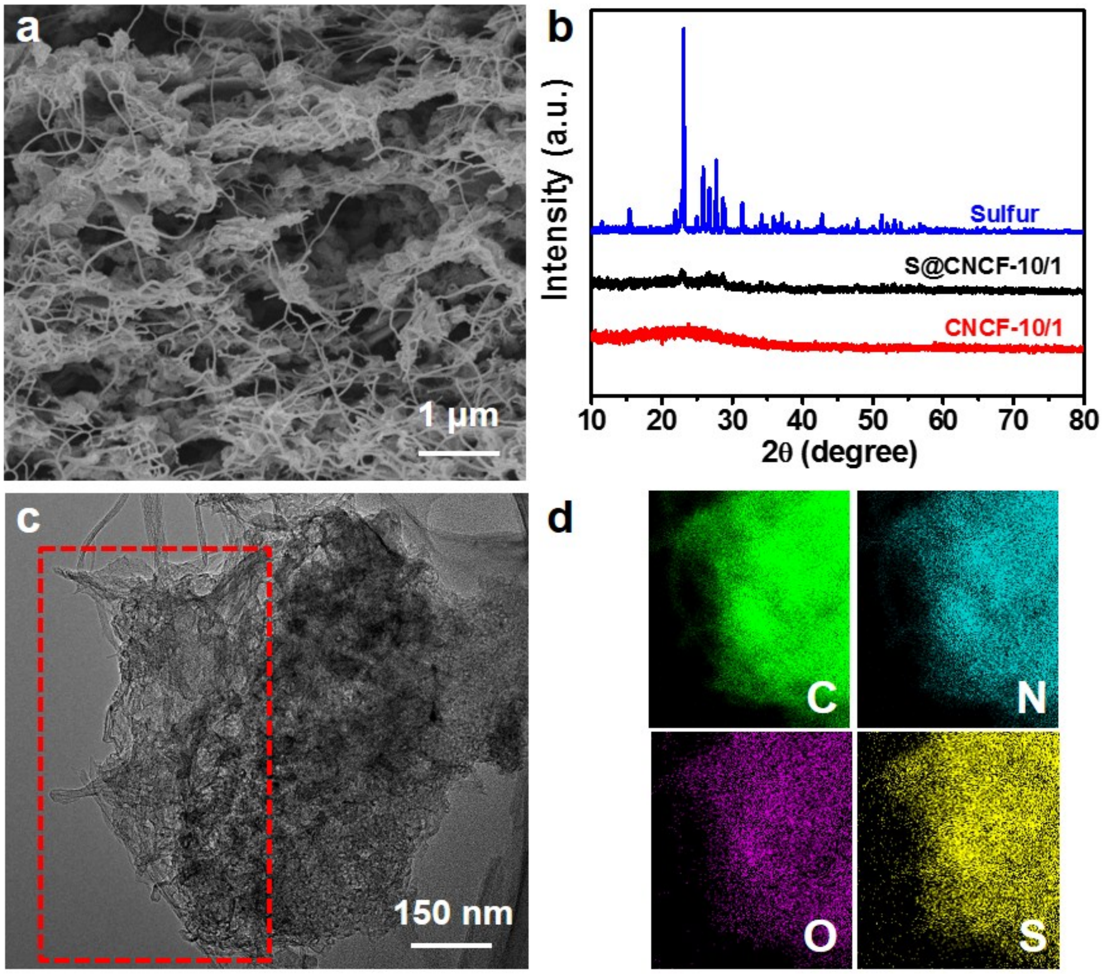

Figure S7. (a) SEM image of S@CNCF-10/1 electrode; (b) XRD pattern of CNCF-10/1 and S@ CNCF-10/1; (c) and (d) TEM image and mapping results of S@CNCF-10/1.

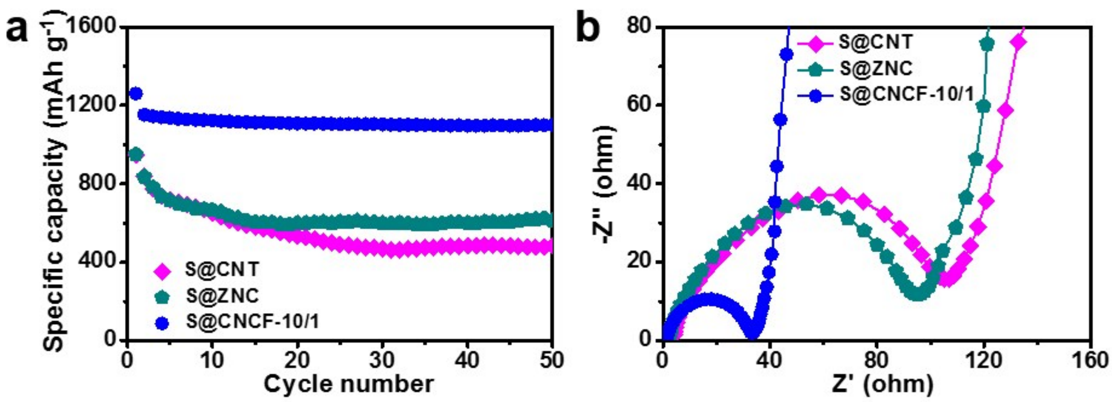

Figure S8. (a) cycling performance of S@CNT, S@ZNC and S@CNCF-10/1; (b) EIS spectra of the samples.

Table S3. Comparison of the S@ CNCF-10/1 electrode with representative high-performance nitrogen-doped carbon material.

Sulfur loading/mg cm^{-2}	Sulfur content/wt%	Rate/C	Cycle number	Capacity retention/mAh g^{-1}	Ref.
3	**70**	**1**	**100**	**959**	**This work**
			200	**926**	
			600	**807**	
			1800	**614**	
--	53	0.2	100	800	31
--	85	0.2	100	980	32
2	--	0.2	300	670	33
3.4	75	0.2	350	519	34
1.2-1.4	--	0.3	200	1002	35
--	63	0.3	300	450	36
1.2	64.5	0.3	300	629	37
1.5	73	0.5	200	896	38
2	67	0.5	300	700	39
	65	0.5	300	740	40
1.5	72	0.5	300	833	41
2	--	0.5	300	844	42
1.1	--	0.5	500	650	43
1.6	--	0.5	1200	566	44
--	74	0.5	1800	230	45
--	76	1	100	800	46
1	53.3	1	200	600	47
--	62	1	200	682	48
0.68	68.1	1	200	786	49
--	70	1	400	706	50
3.3	--	1	500	600	51
--	60	1	500	652	52
1.5	64	1	600	438	53
0.7	60	1	600	500	54
--	63	1.2	120	962	55
1.2	--	2	200	461	56
--	54	2	250	621	57
0.54	--	2	300	450	58

The active material loading, current rate and specific capacities are calculated based on elemental sulfur (1C=1675 mA g^{-1})

Table S4. Comparison of volumetric capacity and sulfur weight content of the S@ CNCF-10/1 with previously reported self-standing sulfur electrode with high sulfur-loading.

Rate/C	Thickness/ μm	Areal sulfur loading/mg cm^{-2}	Sulfur content/ wt%	Volumetric sulfur loading/g cm^{-3}	Volumetric capacity/Ah cm^{-3}	Ref.
0.5	80	6.8	52	0.85	0.66	59
0.2	65	4.5	68	0.69	0.58	60
0.9	145	6.1	45	0.42	0.42	61
0.2	100	5.1	44	0.51	0.36	62
0.5	160	4.7	70	0.29	0.29	63
0.1	150	3.3	44	0.22	0.20	51
0.5	150	3	40	0.20	0.17	64
0.2	140	2.5	77	0.18	0.16	65
0.2	100	2.1	57	0.21	0.23	66
0.2	**78**	**6.9**	**71**	**0.88**	**0.84**	**This work**

The volumetric capacity is valued by the 50^{th} cycle. The active material loading, current rate and specific capacities are calculated based on elemental sulfur (1C=1675 mA g^{-1})

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