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Supporting Information

A Core-shell Cathode Substrate for Developing High-loading, High-performance Lithium-sulfur Batteries

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Supporting Figures

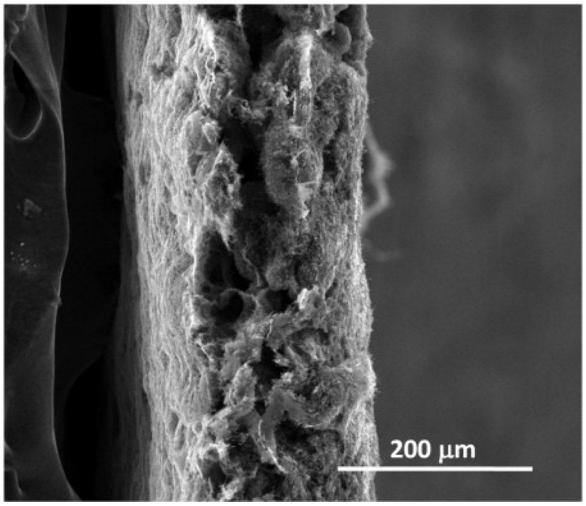


Fig. S1. Cross-sectional view of the core-shell CNF substrate.

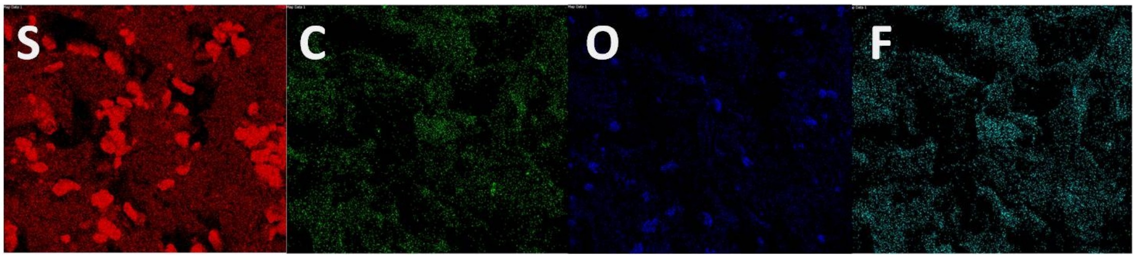


Fig. S2. EDX mapping results (elemental S, C, O, and F signals) of Figure 1e.

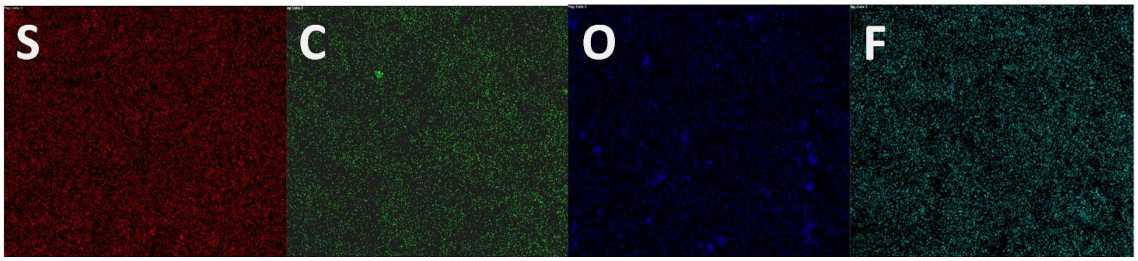


Fig. S3. EDX mapping results (elemental S, C, O, and F signals) of Figure 1f.

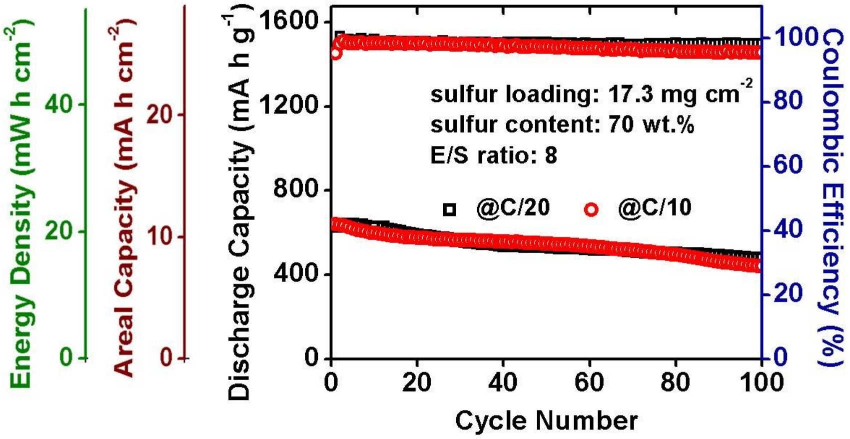


Fig. S4. Cycling performance of the core-shell cathodes with a sulfur loading of 17.3 mg cm^{-2}.

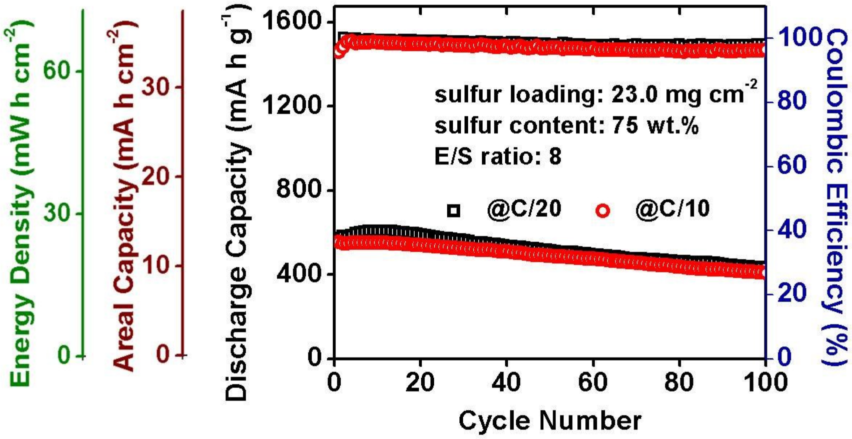


Fig. S5. Cycling performance of the core-shell cathodes with a sulfur loading of 23.0 mg cm^{-2}.

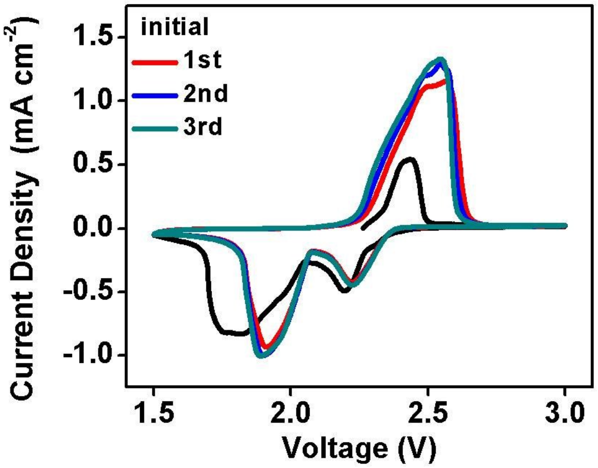
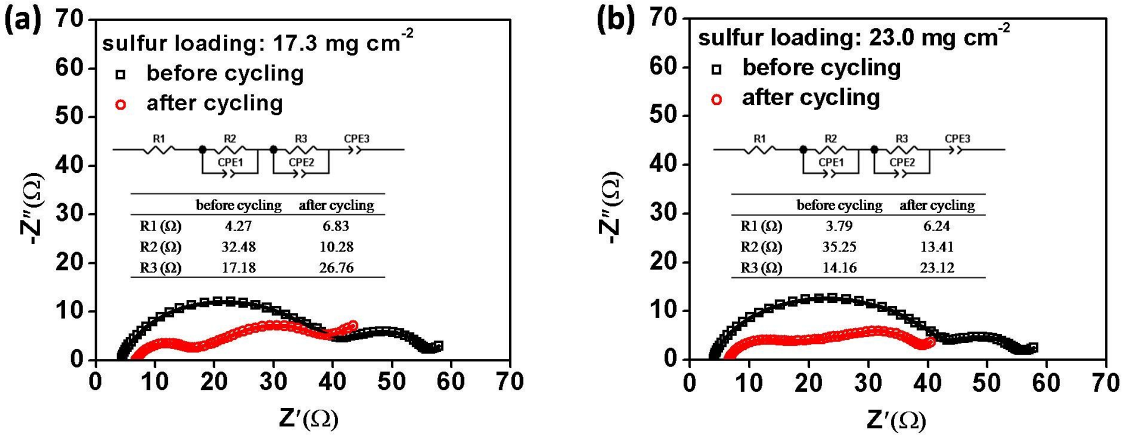


Fig. S6. CV curves of the core-shell cathode with a sulfur loading of 23.0 mg cm^{-2}.



(a)

	before cycling	after cycling
R1 (Ω)	4.27	6.83
R2 (Ω)	32.48	10.28
R3 (Ω)	17.18	26.76

(b)

	before cycling	after cycling
R1 (Ω)	3.79	6.24
R2 (Ω)	35.25	13.41
R3 (Ω)	14.16	23.12

Fig. S7. Electrochemical impedance spectra and the fitting results of the core-shell cathodes with sulfur loadings of (a) 17.3 mg cm^{-2} and (b) 23.0 mg cm^{-2} before and after cycling.

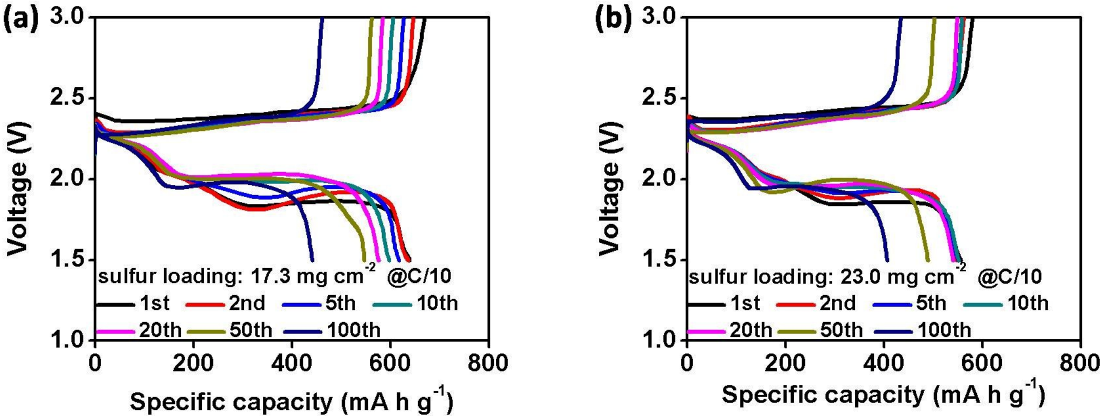


Fig. S8. Voltage profiles of the core-shell cathodes with sulfur loadings of (a) 17.3 mg cm^{-2} and (b) 23.0 mg cm^{-2} at C/10 rate.

Supporting Table

Table S1 Electrochemical performances of state-of-the-art lithium-sulfur battery cathodes

	sulfur loading (mg cm^{-2})	sulfur content (wt.%)	E/S ratio (μL mg^{-1}s)	area capacity (mA h cm^{-2})	cycle number	capacity retention	C-rate / current density
S1	11.6	50*	6	11.2	20	62%	0.2 mA cm^{-2}
S2	12	59	/	13.6	40	67%	40 mA g^{-1}
S3	12.5	42.5*	/	3.1	70	81%	C/10
S4	13.9	55*	15	14.6	100	70%	C/20-C/3
S5	10.8	72.3*	27.8	10.7	50	76%	1.2 mA cm^{-2}
S6	11.9	80	/	7.4	300	61%	C/10
S7	10	60	15	7.5	200	62%	C/10
S8	11.33	70	4.4	7.5	50	86%	C/5
S9	10.2	63*	4	10.3	1	/	C/10
S10	16	70	12	13.3	100	57%	C/10
	32	70	12	16.5	100	58%	C/10
S11	19.3	75	4	3.9	50	58%	C/10
S12	10.9	54	8	9.4	30	85%	C/20-C/10
	17.3	54	7	15.1	17	79%	C/20-C/10
S13	11.1	75*	4	11.2	12	75%	50 mA g^{-1}
S14	10	60*	13	13.3	50	91%	C/3
S15	13	41	4	8.4	30	54%	0.5 mA cm^{-2}
	13	48	4	10.1	20	74%	0.5 mA cm^{-2}
this work	**23**	**75**	**8**	**14.0**	**100**	**71%**	**C/20-C/10**

* does not include the mass of current collector (Al foil or Al foam).

Comparative Assessment of the Core-shell Carbon Substrate

Compared with the exiting work in the literature, the novelty and advancement of this core-shell design lies in three aspects: First, the three-dimensional, core-shell substrate has quite different internal and external structures. As shown in Figures 1b and 1, the CNF substrate contains simultaneously a porous core and a dense shell. However, in most of the existing work in the literature, two carbon-based layers with the same morphology are used to encapsulate the active material.[S16,S17] More importantly, the unique structure in this work is the self-assembly through the phase inversion method. Second, the CNF substrate has integrated internal and external regions, which distinguishes this work from other core-shell design. For example, in our previous work, we fabricated the carbon shell with carbon papers and carbon paper O-ring[S18]. Compared to the divided carbon papers and carbon paper O-ring, the CNF substrate in this work attains better structural integrity. The continuous electron/ion pathways through the whole structure facilitate the redox reaction. Third, the active material achieves intimate contact with the conductive agent. Herein, the "core" is not only the pure sulfur core but also the porous CNF network core. The woven CNFs fully wrap the active material, which improves the reaction kinetics. The CNF network core also increases the tortuosity of the internal region and hinders the migrating of polysulfides. Additionally, the porous space helps to buffer the volume changes during the lithiation/delithiation reaction.

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