

## Supporting Information

### **A robust network 1 binder with dual functions of Cu<sup>2+</sup> ions as ionic crosslinking and chemical binding agents for highly stable Li–S batteries**

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**Table S1** Comparisons of capacity retention rates of sulfur electrodes in binder-related studies.

Reference	Binder	Current density	Cycle number	Capacity (mAh g <sup>-1</sup> )	Retention rate <sup>1</sup>
<b>This work</b>	<b>SA-Cu6</b>	<b>0.2 C</b>	<b>100</b>	<b>925</b>	<b>83%</b>
<b>1</b>	Reduced graphene oxide-polyacrylic acid	0.5 C	100	635	77%
<b>2</b>	Poly(vinylidene difluoride-trifluoroethylene)	0.2 C	100	801	~67%
<b>3</b>	Sodium alginate	0.2 C	50	508	65%
<b>4</b>	LA132	0.2 C	50	885	76%
<b>5</b>	Gelatin	0.4 mA cm <sup>-2</sup>	50	408	36%
<b>6</b>	Mixture of SBR and CMC	100 mA g <sup>-1</sup>	60	580	67% <sup>2</sup>
<b>7</b>	Gum arabic	0.2 C	50	1090	79%
<b>8</b>	Perylene bisimide/PVDF composite	1.0 C	150	600	86%
<b>9</b>	Carbonyl- $\beta$ -cyclodextrin	0.2 C	50	1456	~77%
<b>10</b>	$\beta$ -cyclodextrin with a quaternary ammonium cation	100 mA g <sup>-1</sup>	100	928	67%

<sup>1</sup>Retention rate is computed for the initial discharge capacity. Some studies without the initial discharge capacity are not listed.

<sup>2</sup>Retention rate is computed for the capacity of the 5<sup>th</sup> cycle.

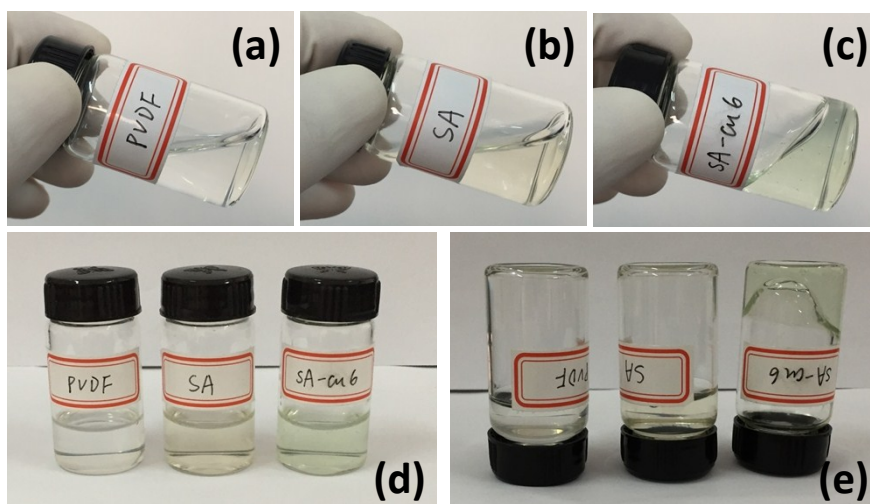
**Table S2** Comparisons of sulfur loadings of different sulfur electrodes in binder-related studies showing that using SA-Cu6 binder can achieve high-loading electrode.

Reference <sup>3</sup>	Binder	Sulfur loading (mg cm <sup>-2</sup> )
<b>This work</b>	<b>SA-Cu6</b>	<b>8.05</b>
20	Carbonyl- $\beta$ -cyclodextrin	3.0
43	Mixture of PAA and poly(3,4-ethylenedioxythiophene) : poly(styrenesulfonate)	0.8
44	Polyamidoamine dendrimers	4.4
45	LA132	1.0
46	Reduced graphene oxide-polyacrylic acid	0.8
47	Perylene bisimide/PVDF composite	1.0
48	Polypyrrole and polyurethane nanocomposite	4.6
49	Amino functional group binder	8.0
50	Cross-linked CMC-citric acid	14.9
51	Polymerization of hexamethylene diisocyanate with ethylenediamine	0.5
52	PEO <sub>10</sub> LiTFSI	4.0
53	PEO:PVP	5.26
54	Polyethylenimine	8.6
55	Polydiallyldimethylammonium	3.0

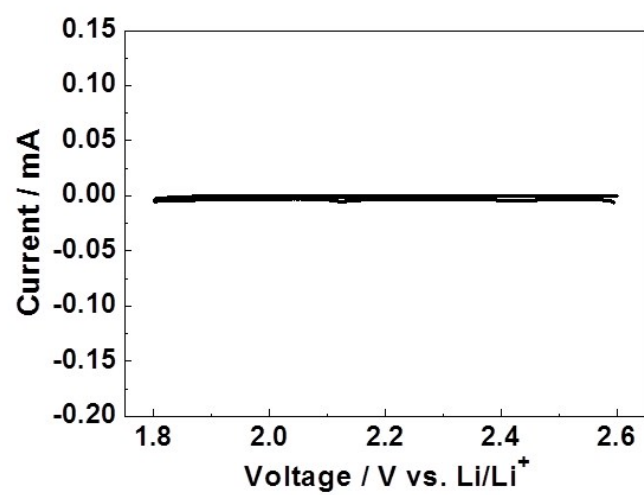
<sup>3</sup>References are listed in the maintext.

## References:

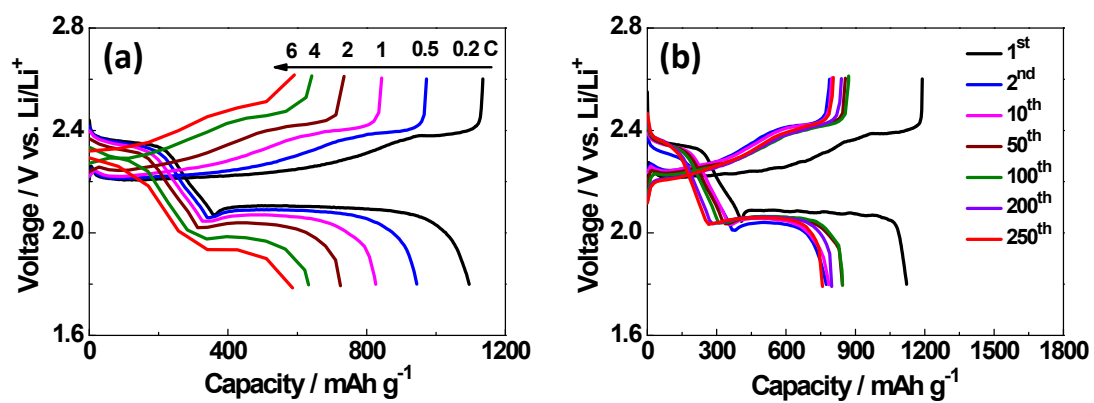
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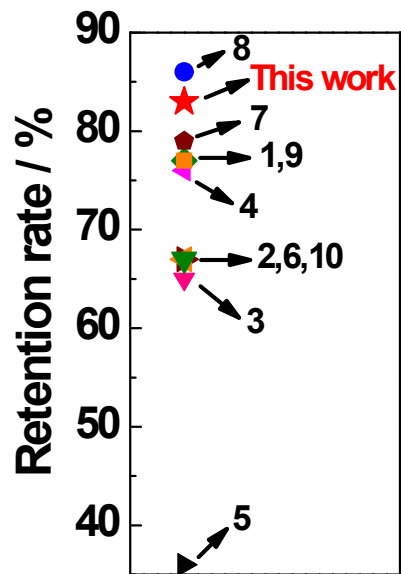
**Fig. S1** Photos of (a) PVDF binder, (b) SA binder, (c) SA-Cu6 binder, (d) erected binders, and (e) inverted binders, visually demonstrating the cross-linking effect of SA and  $\text{Cu}^{2+}$  ions.



**Fig. S2** CV curves of SA-Cu6 binder at  $0.1 \text{ mV s}^{-1}$ . The electrode consists of SA-Cu6 binder and super P conductive additive with a mass ratio of 1:1.

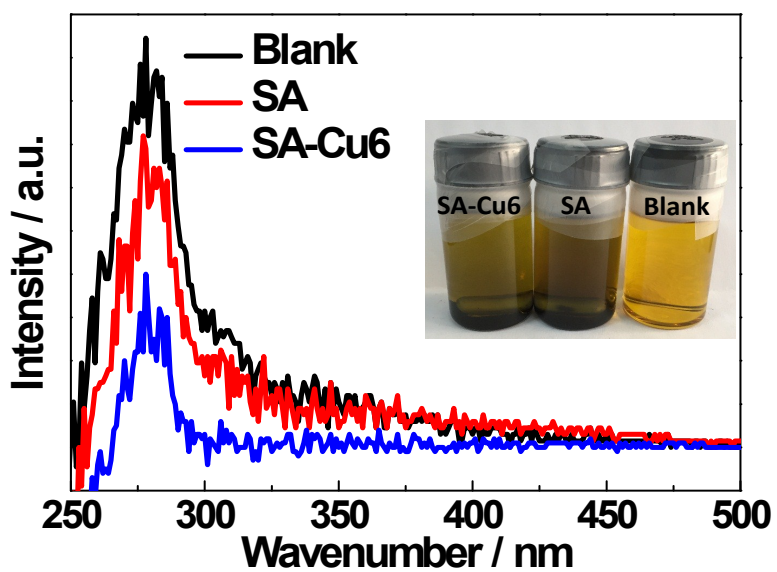


**Fig. S3** Charge-discharge curves of S@SA-Cu6 electrodes (a) at different rates and (b) at 1 C for different cycles (the first cycle is at 0.2 C).

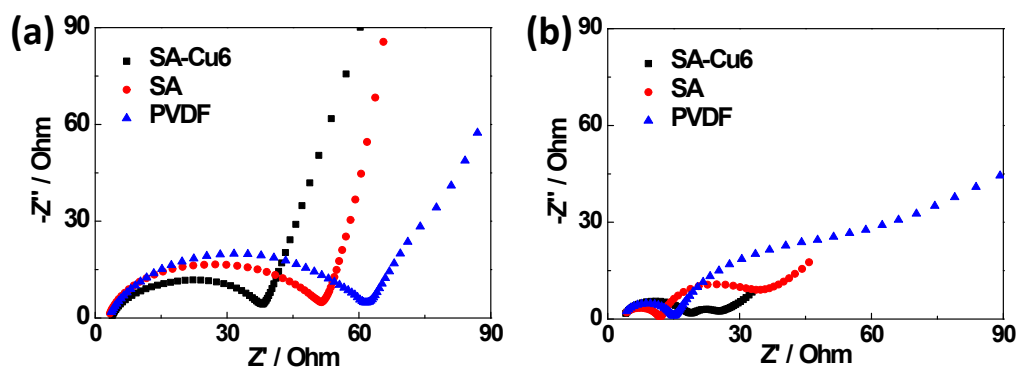


**Fig. S4** Comparisons of capacity retention rates of sulfur electrodes in binder-related studies, according to Table S1.

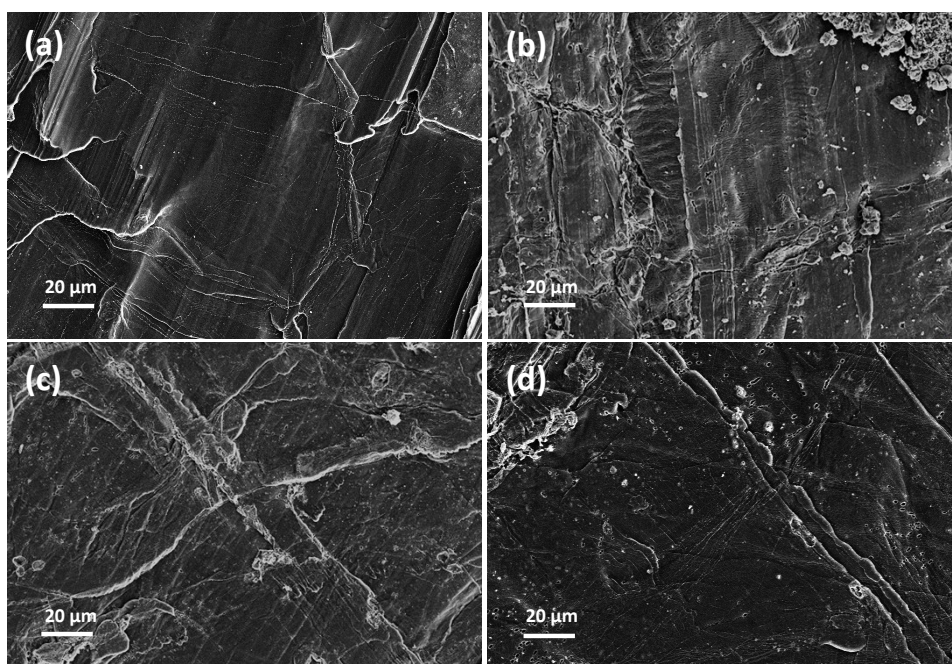




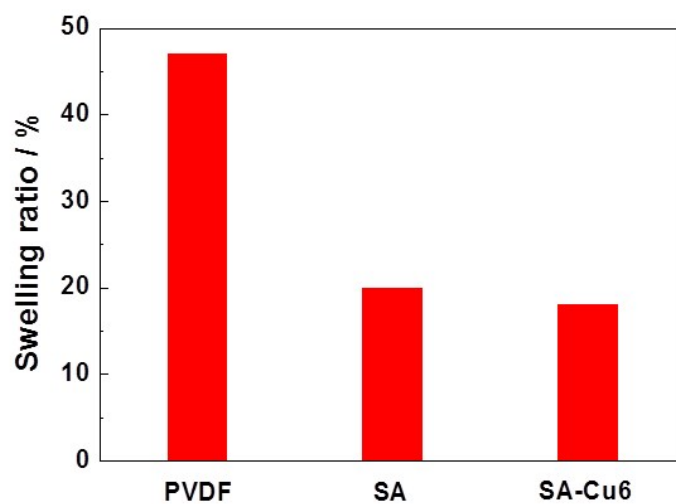
**Fig. S5** UV-vis spectra of SA-Cu6/super P and SA/super P soaking in 2 mM  $\text{Li}_2\text{S}_6$  solutions and pristine 2 mM  $\text{Li}_2\text{S}_6$  solution, the inset is digital image of these solutions.



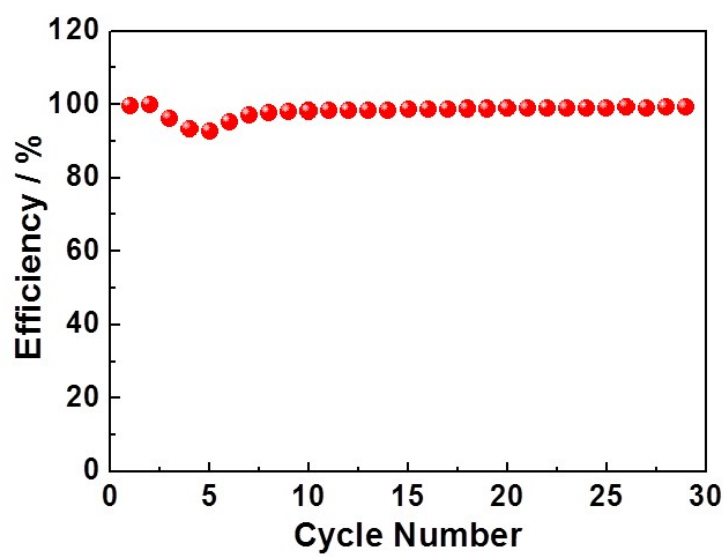
**Fig. S6** (a) EIS of fresh cells and (b) cells after 20 cycles at 0.2 C fabricated with different binders.



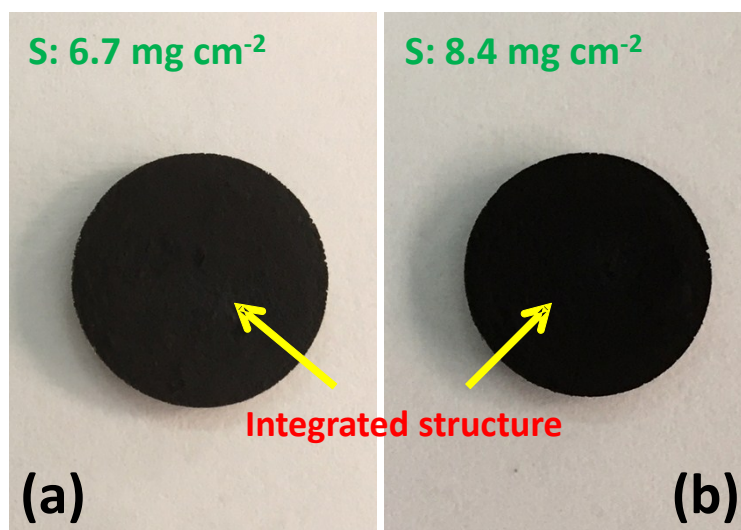
**Fig. S7** SEM images of (a) fresh lithium foil anode and lithium foil anodes detached from cells fabricated with (b) S@PVDF electrode, (c) S@SA electrode, and (d) S@SA-Cu6 electrode after 20 cycles at 0.2 C.



**Fig. S8** Swelling ratios of different binders. The swelling ratio is defined as the percent of the increased weight of binder after soaking in the electrolyte for 24 h and the initial weight of binder.



**Fig. S9** Coulombic efficiency of high-loading S@SA-Cu6 electrode with a sulfur loading of 8.05 mg cm<sup>-2</sup>.



**Fig. S10** Photos of high-loading S@SA-Cu<sub>6</sub> electrodes with sulfur loadings of (a) 6.7 and (b) 8.4 mg cm<sup>-2</sup> showing the crack-free electrode structure. The electrodes consist of S/super P composite and SA-Cu<sub>6</sub> binder without 1D and 2D carbon materials.