Supporting Information
Multifunctional water-soluble binder for Li-S batteries

1. Experimental section

1.1 Preparation of binder PPB

First, 20 mg of polyvinyl alcohol (PVA, AR, General-reagent®) was added to 2 mL of deionized water and stirred at 90 °C until completely dissolved. A further 20 mg of polyacrylic acid (PAA, average Mv ~450,000, Aladdin) and 20/10 mg of boric acid (BA, AR, Sinopharm Chemical Reagent) were added with thorough mixing until complete dissolution. The mixture was put into an oven at 60 °C for 12 h. After removal, PPB-1 (20mg BA) and PPB-2 (10mg BA) were produced by freeze-drying, respectively.

1.2 Preparation of sulfur anode

Carbon nanotubes and sublimated sulfur were ground in a 3:7 mass ratio, and the mixture was heated at 155 °C for 12 h under an argon atmosphere to produce C-S composites. The C-S composite, conductive agent (Super P), and binder (PVDF/PPB-1/PPB-2) were ground at a ratio of 7:2:1. For comparison, the powder containing PVDF and the powder containing PPB were dissolved in NMP and thoroughly stirred to form a homogeneous slurry. The slurry was evenly coated on the carbon-coated aluminum foil with a scraper and dried in a vacuum oven at 60 °C for 12 h.

1.3 Battery assembly

Coin batteries were assembled in an argon-filled glove box using a CR2025 model battery case. A polypropylene film (Celgard 2500) was used as the separator between the sulfur cathode and the lithium anode. 1.0 M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) in DME and DOL (volume ratio of 1:1) containing 2 wt% $LiNO_3$ was applied as electrolyte.

1.4 Characterization and electrochemical testing

Field emission ambient scanning electron microscope (Thermo QuattroS Scanning

Electron Microscope) was used to characterize the sample morphology. Fourier transform infrared spectroscopy (FTIR) was obtained using a Thermo Scientific Nicolet iS5 spectrometer. Thermogravimetric analysis (TGA) was measured using a NETZSCH TG209F1-Thermo Nicolet iz10 from room temperature up to 600 °C at a rate of 10 °C min^{-1}. The contact angle test was measured using a LAUDA Scientific LSA100 contact angle meter, and the contact fluid was Li-S batteries electrolyte. The adsorption capacity of the binder on LiPSs was evaluated by analyzing the UV-Vis spectra (SHIMADZU 3600 Plus) performed on the solution after the binder adsorption. The effect of the binder on the cathode after charging and discharging was further analyzed by XPS (Thermo Fisher ESCALAB 250Xi). Charge-discharge curves between 1.6 V and 2.8 V were performed using LAND CT3002A. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were measured using AUTOLAB PGSTAT302N.

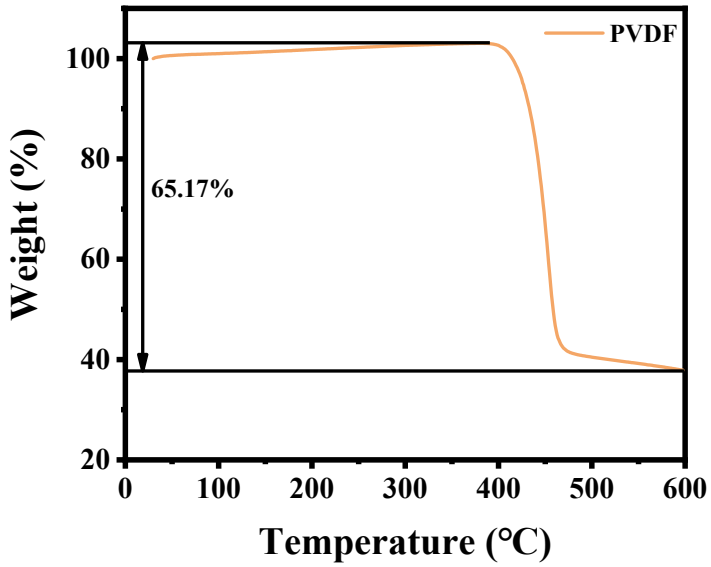

Fig. S1 Thermal gravimetric analysis curves of binder PVDF with different component ratios under Ar atmosphere.

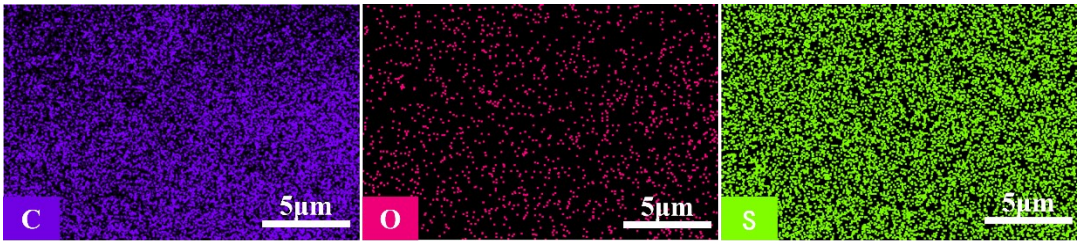

Fig. S2 EDS mapping of C, O and S elements in the cathodes of PPB-based Li-S batteries.

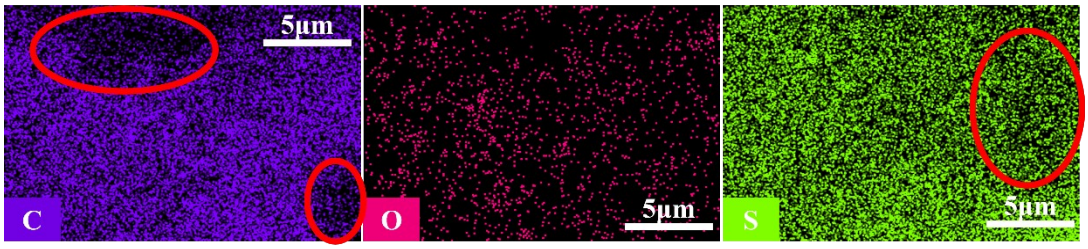

Fig. S3 EDS mapping of C, O and S elements in the cathodes of PVDF-based Li-S batteries.

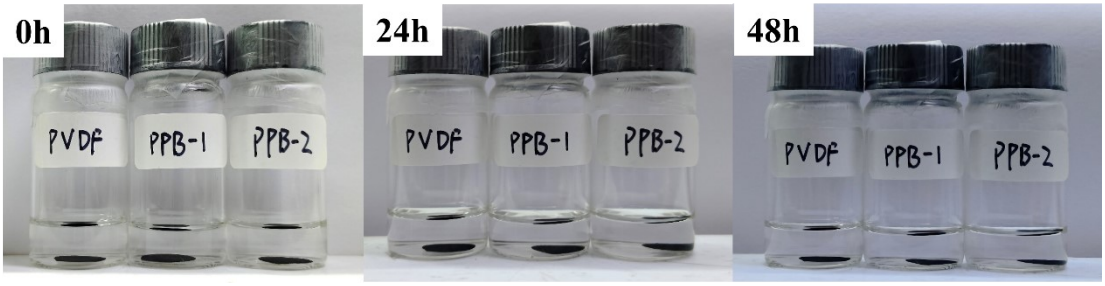

Fig. S4 Digital images of sulfur cathodes using PVDF, PPB-1 and PPB-2 binders after immersion in Li-S batteries electrolyte for 0h, 24h and 48h, respectively.

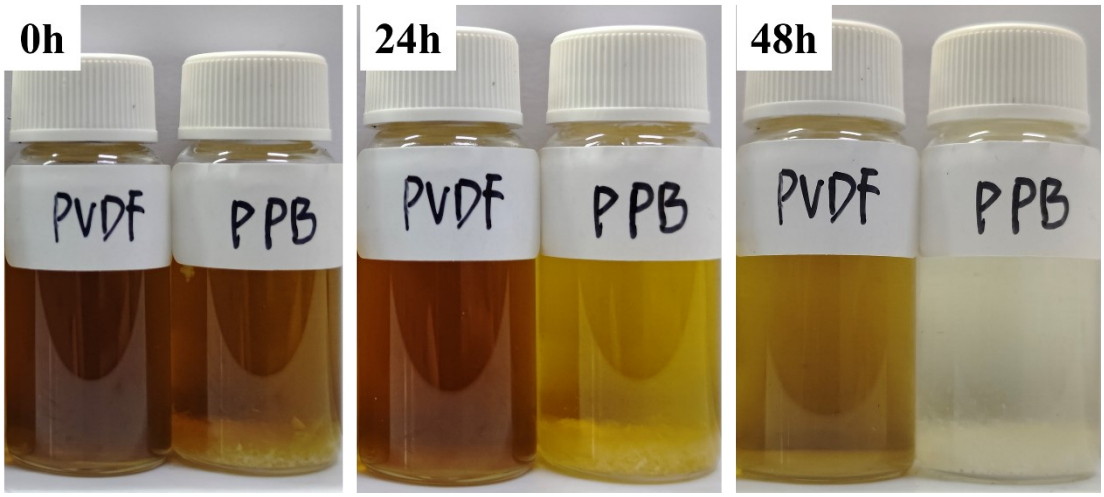

Fig. S5 Digital photographs of binder PVDF and PPB before and after Li_2S_6 absorption.

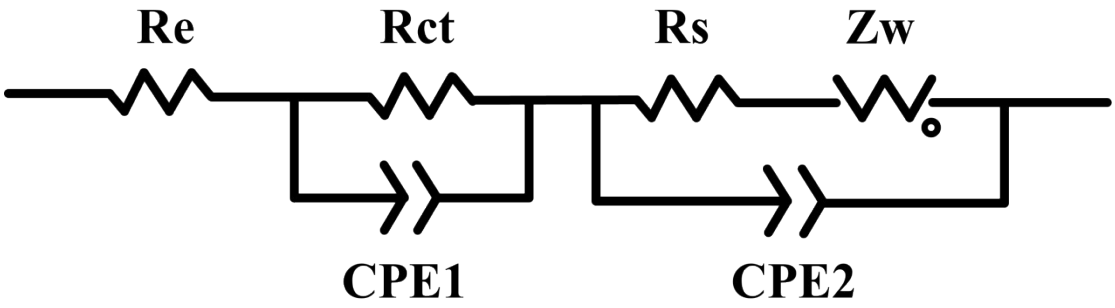

Fig. S6 The equivalent circuits used to fit the impedance spectrum.

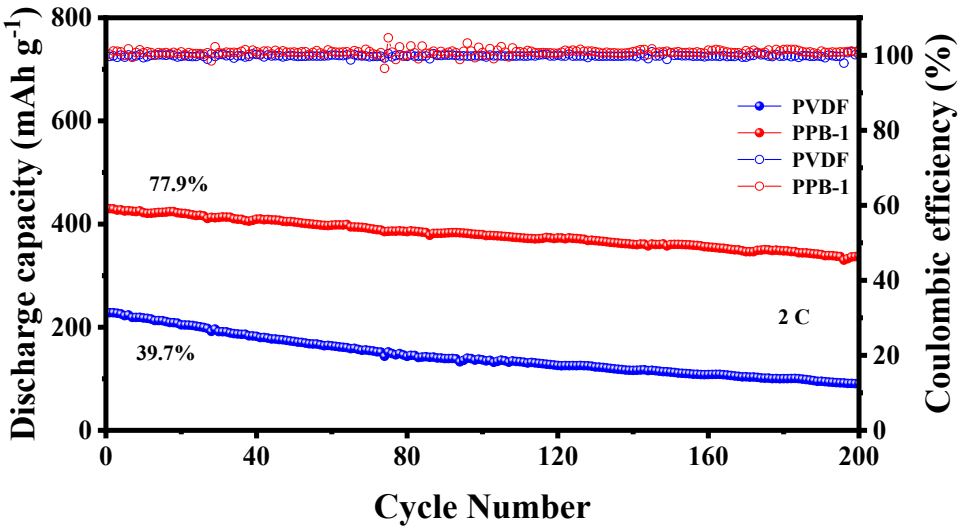

Fig. S7 Long cycling performance and Coulombic efficiencies at 2C of Li-S battery based on PVDF and PPB-1.

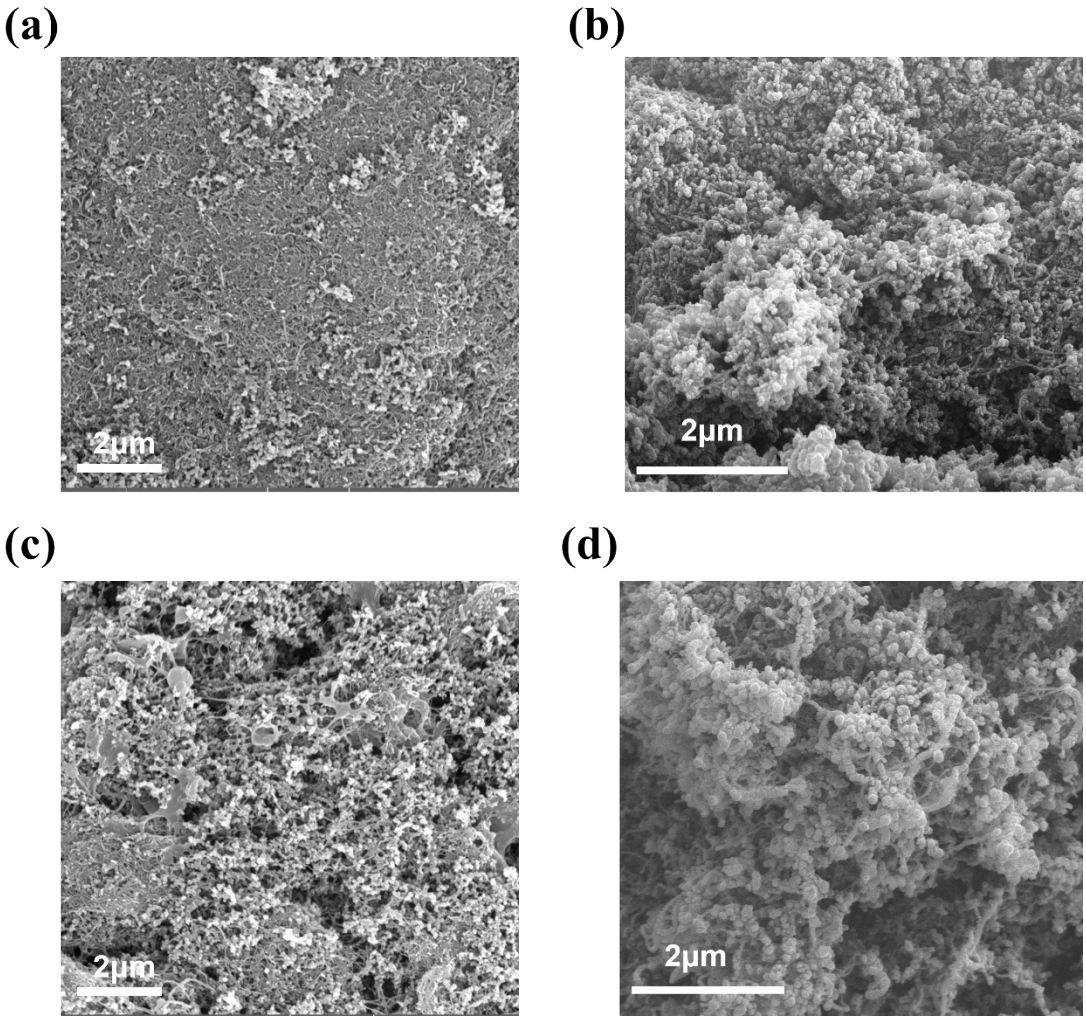

Fig. S8 SEM image (a) and cross-sectional SEM image (b) before cathode reaction for S@PPB. SEM image (c) and cross-sectional SEM image (d) before cathode reaction for S@PVDF.

Table S1 Mass percentage of C,O,S elements in cathode sheet at different magnifications

Material	Carbon	Oxygen	Sulfur
S@PPB (10KX)	89.04	1.93	9.03
S@PVDF (10KX)	85.87	6.81	7.32
S@PPB (20KX)	89.95	1.63	8.42
S@PVDF (20KX)	87.15	5.92	6.93

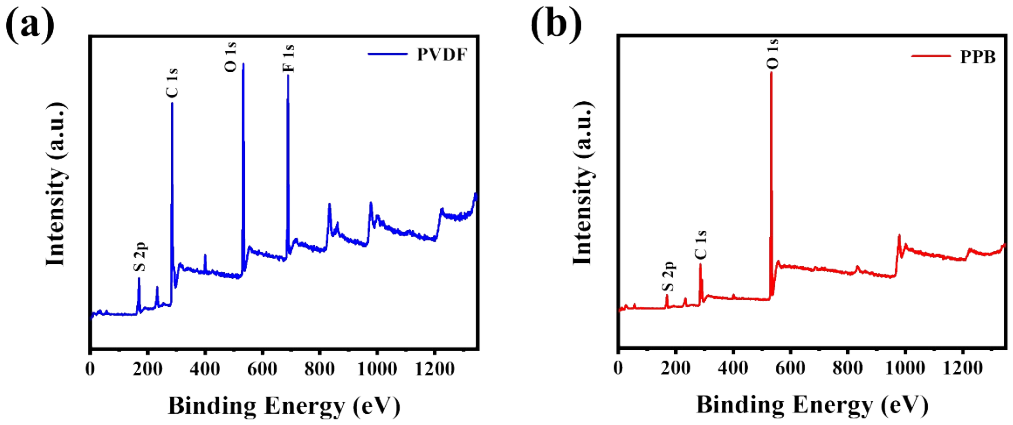

Fig. S9 (a) XPS full spectrum of S@PPB cathode. (b) XPS full spectrum of S@PVDF cathode.

Table S2 Comparison of the performance of Li-S batteries assembled with PPB and recent Li-S batteries binders reported in the literature

Capacity Retention	C Rating	Cycle Number	Binder
84.4%	0.5 C	200	PPB-1 (This work)
49.18%	0.5 C	500	CAB[1]
62.5%	0.2 C	100	γ-PGA[2]
82%	0.5 C	500	N2200-OE[3]
48.9%	0.5 C	500	PAA-B-HPRN^{+4}
61.17%	0.1 C	300	NH_4PA[5]

References

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