## Supporting Information

# A Janus Protein-based Nanofabric for Trapping Polysulfides and Stabilizing Lithium Metal in Lithium-Sulfur Batteries 1. Experimental

**1.1 Sample preparation** 

# *Janus nanofabrics preparation.* Gelatin protein (from porcine skin, Type A, Mw = 50,000 ~ 100,000 g mol<sup>-1</sup>), pyrrole and Polyvinylpyrrolidone (PVP, Mw = 1,300,000 g mol<sup>-1</sup>) were all purchased from Sigma-Aldrich Co. Ltd. Gelatin protein was first dissolved and denatured in a solvent mixture of acetic acid (AA) and distilled water (DI) with a weight ratio of 8: 2. Then the viscous gelatin solution with a concentration 15 wt% was loaded into a plastic syringe and with a sctainless-steel nozzle connecting to high DC voltage and subjected to electrospinning. A high voltage of 17.5 kV, a constant flow rate of 1 ml h<sup>-1</sup> and a distance from nozzle to collector of 100 mm were used as the electrospinning parameters to obtain gelatin nanofabrics (G-nanofabric) with fine fibers. PVP was dissolved in Dimethylformamide (DMF) and then same amount of FeCl<sub>3</sub> was added and dissolved to prepare a PVP/FeCl<sub>3</sub> solution with a weight ratio of 1:1. The electrospinning parameters for the PVP/FeCl<sub>3</sub> solution was 20 kV, 0.5 ml h<sup>-1</sup> and 150 mm. Next, the PVP/FeCl<sub>3</sub> nanofibers and pyrrole monomer were placed in a sealed bottle and kept for 26 h after which we obtained polypyrrole (Ppy) coated PVP that is a conductive nanofabric (CNF). Then, 1 wt% gelatin solution was dropped on the CNF to get the gelatin-functionalized CNF (G@CNF). Finally, by combining the G@CNF and G-nanofabric, our Janus nanofabric was fabricated.

*Surfer cathode preparation*. The sulfur cathodes were prepared by mixing 63.8 wt% sulfur powders (Sigma-Aldrich), 24.1 wt% carbon black (CB) (Super C45, MTI Corp.), 2.1 wt% carbon nanofibers (Applied Science, Inc.) and 10 wt% PVDF in NMP using bowl-mill until homogeneity. The slurry was

then cast onto carbon-coated aluminum foil and dried at 50 °C overnight. The loading of sulfur active material was controlled about 1.5 mg cm<sup>-2</sup>. The liquid electrolyte solution was composed of 1 M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) dissolved in mixture solvents of 1,3-Dioxolane (DOL) and 1,2-Dimethoxyethane (DME) (1:1 by volume), and 2 wt% LiNO<sub>3</sub> was added as additive. The Li<sub>2</sub>S<sub>6</sub> solution was prepared by adding sulfur and lithium sulfide powders in a 5:1 molar ratio into a solvent mixture of DOL/DME (V/V=1:1) and stirred for 24 h under 60 °C.

## **1.2 Material characterizations**

*Morphology, porosity and electrolyte uptakes characterizations.* Scanning Electron Microscopy (SEM) (Quanta 200F) was used to characterize the morphology of separators and lithium metals as freshprepared or after disassembled from cycled batteries. For materials after cycling, they were first extracted from the coin cells in the glove box and followed by gentle rinse with DOL/DME (v/v: 1:1) to remove Li salt residue, and then dried in glove box for 24 h. The porosity of the nanofabric samples was measured via n-butanol uptake. Measurement of liquid uptake was carried out by dipping the gelatin and Celgard samples in the electrolyte solution. The excess electrolyte remaining on the surface of the samples was removed by wiping softly with a tissue paper. Sample was periodically weighed until no weight change was observed. The swelling ratio was defined by the weight ratio of the net liquid uptake to the dried polymer sample, can be written as

$$E = ((W_2 - W_1)/W_1) \times 100$$

where,  $W_1$  and  $W_2$  are the mass of the samples before and after dipping in the electrolyte solution, respectively. The measurement of each sample was repeated for at least five times for consistency. *Surface properties characterizations*. Fourier Transform Infrared Spectroscopy (FT-IR, Thermo ScientificTM NicoletTM iS50) was used to determine the functional groups present on materials. The wettability of Celgard, the G-nanofabric and G@CNF was analyzed by contact angle (OCA 25, Germany).

*Conductivity testing.* The ambient ionic conductivity was measured via AC impedance spectroscopy with an electrochemistry workstation (Biologic VSP EC-Lab). The bulk resistance and the ionic conductivity can be calculated based on the following equation:

$$\sigma = L/(AR_b)$$

where *L* is the thickness of the testing samples, cm; *A* is the contact area for the sample and electrodes, cm<sup>2</sup>;  $R_b$  is the bulk resistance that can be read at the intersection of *x* axis,  $\Omega$ . Electrical resistance of interlayers was measured by four-probe method using Keithley 2400. The top cover of the sample holder was attached with four needles parallelly aligned with 2 mm spacing. The inner and outer two needles connected with one resistivity tester to test the voltage and current, respectively. The samples were placed in the sample holder with intimate contact with the needles. Five measurements were conducted to ensure consistency.

*Electrochemical performance testing.* The lithium-ion transference number ( $t_{Li+}$ ) was evaluated by the method of chronoamperometry in the cell Li/separator/Li with a polarization voltage of 0.01 V. The  $t_{Li+}$  was obtained according to the equation (1):

$$t_{\text{Li+}} = \frac{I_t \times R_{bt} \times (\Delta V - I_0 \times R_{i0})}{I_0 \times R_{b0} \times (\Delta V - I_t \times R_{it})}$$
(1)

Where  $I_0$  and  $I_t$  were the initial and steady-state current that were recorded by chronoamperometry for 1000 s.  $R_{b0}$  and  $R_{bt}$  were the bulk resistance at initial and steady state, respectively.  $R_{i0}$  and  $R_{it}$  were the interfacial resistance between the electrolyte and separator before and after the test.

Electrochemical impedance spectroscopy (EIS) was used to characterize the impedance of the cells via an electrochemical workstation (Biologic VSP EC-Lab) over a frequency range of 0.01-10<sup>6</sup> Hz.

The electrochemical stability window was obtained from linear sweep voltammetry using the cell of Li/separator/Stainless steel. Cyclic voltammetry (CV) measurements of Li-S cells performed between 1.7 and 2.8 V (Li/Li<sup>+</sup>) at a scan rate of 0.1 mV s<sup>-1</sup> and the lithium ion diffusion property was characterized via CV measurements at scan rates of 0.2, 0.3, 0.4, 0.5 mV s<sup>-1</sup>, respectively. The galvanostatic charge/discharge measurements were monitored using the LANHE battery testing system (CT2001A, Wuhan LAND electronics Co., China) within a voltage range of 1.8 - 2.8 V.

## 2. Supporting tables and figures



**Figure S1.** Diameter distributions of PVP/FeCl<sub>3</sub> fibers obtained by electrospinning solutions with different concentrations.



**Figure S2.** Pore size distributions of PVP/Ppy fibers (polymerization time of 26 h) obtained by electrospinning solutions with different concentrations.



Figure S3. Photographs of the flexible (a) PVP/FeCl<sub>3</sub> nanofabric and (b) PVP/Ppy nanofabric (CNF).



**Figure S4**. (a) SEM images of G-nanofabric, (b) the fiber diameter distribution and (c) pore size distribution.



Figure S5. Contact angle between electrolyte and G@CNF.



Figure S6. FTIR spectrum of G-nanofabric.



**Figure S7.** Interface impedance spectrum of Li-Li cells with (a) Celgard and (b) G-nanofabric separators before and after polarization



**Figure S8.** Voltage profiles of Li/Cu cells during lithium stripping/plating cycling at a current density of 0.25 mA cm<sup>-2</sup>.



**Figure S9.** Voltage profile of Li/Cu cell with G-nanofabric separator during lithium stripping/plating cycling at a current density of 0.5 mA cm<sup>-2</sup>.



Figure S10. The morphology of fresh lithium metal.

	R <sub>b</sub>	R <sub>CT</sub>	R <sub>SEI</sub>
Celgard	4.6	67.8	194.9
G-nanofabric	1.4	36.8	83.1
G@CNF/Celgard	4.2	66.6	-
Janus nanofabric	5.1	60.85	-

Table S1. Fitting results of EIS testing on fresh cells at discharged status



Figure S11. C-ratel performance of Li-S batteries with PVP/Ppy (CNF) interlayers electrospun form 7 wt% and 9 wt% solutions.



Figure S12. Cycle performance of Li-S batteries with CNF and G@CNF interlayers.



Figure S13. Voltage profiles at a current density of 0.3 A g<sup>-1</sup>.



Figure S14. Digital photos showing the diffusion process of polysulfides in a system separated by Celgard separator (left) and Janus separator (right).



Figure S15. G-nanofabric separator from a cycled Li-S battery