Supplementary Information

In-situ monitoring viscosity change of electrolyte in a Li-S battery

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Electrode preparation

Sulfur powder (0.2 g, Alfa Aesar, precipitated, 99.5%) and Denka black (0.2 g) were first thoroughly ground in an agate mortar, then dispersed in polyvinylidene difluoride (PVDF, Solef®) solution (4 g, 2.5 wt% in anhydrous N-methyl-2-pyrrolidone, Sigma-Aldrich, 99.5%) with a planetary centrifugal mixer (ARE-310, Thinky). The obtained slurry was evenly cast on an aluminum foil and dried at 60 °C. The areal sulfur loading is around 4 mg cm^{-2}. The electrode was cut into a strip with a dimension of 0.8 × 7.0 cm (0.8 × 3.0 cm with sulfur coated).

Cell assembling

Lithium ribbon (thickness: 0.75 mm, Aldrich, 99.9% trace metals basis) was cut into 0.3 cm × 7.0 cm as the anode. A glass cup with a circular truncated cone (r1 = 0.8 cm, r2 = 0.5 cm and h = 4.5 cm) was used as the reactor. Sulfur cathode and lithium anode were placed opposite to each other in the cup and the bottom was fixed by a filter paper ball. 4 mL of electrolyte containing 0.5 M lithium bis(trifluoromethanelsulfonyl)imide (LiTFSI, Alfa Aesar, 98+%) in tetraethylene glycol dimethyl ether (TEGDME, Aldrich, ≥99%) with 1.0 wt% of $LiNO_3$ additive (Sigma, ≥99%) was added into the glass cup to start the measurement. All procedures were carried out in an Ar-filled glove box (Mbraun). The used lithium salts, LiTFSI and $LiNO_3$, were dried at 180 °C for 15 hours before transferred into the glove box.

In-situ electrochemical test

Electrolyte viscosity change in a full discharge-charge cycle was monitored by a Sine-wave vibro viscometer (SV-1A, AND). Electrochemical test was performed on the BTS3000 battery cycler (Neware) at a constant current of 1.0 mA in a voltage window of 1.5-2.8 V (*vs.* Li/Li^+). The data of cell voltage, electrolyte viscosity and temperature were collected in every 5 seconds. The time-lapse video was recorded by GC-P100 (JVC) at 5-second intervals and edited by the software of Movie Maker (Microsoft).

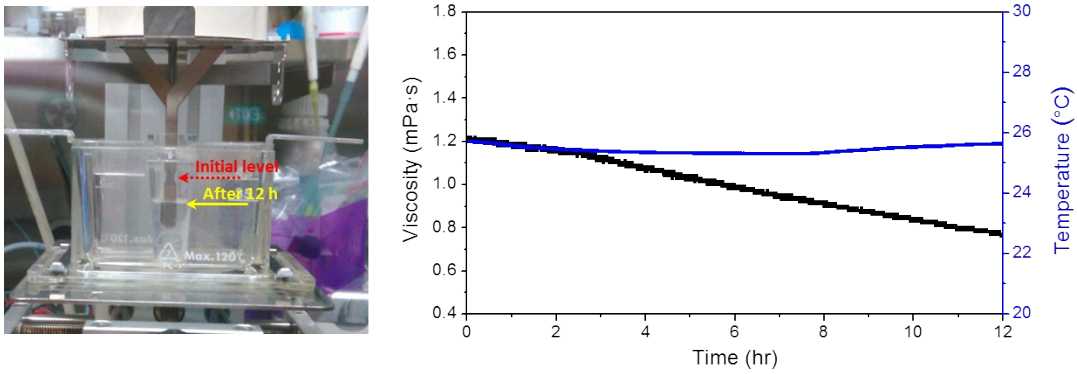

Fig. S1 Viscosity test of the electrolyte with 1.0 M LiTFSI dissolving in a mixture solvent of 1,2-dimethoxyethane (DME, Sigma-Aldrich, anhydrous, 99.5%) and 1,3-dioxolane (DOL, Sigma-Aldrich, anhydrous, 99.8%, v:v=1:1). After 12 hours, around 1/3 volume of electrolyte in the cup disappeared and the drawdown resulted in a false viscosity.

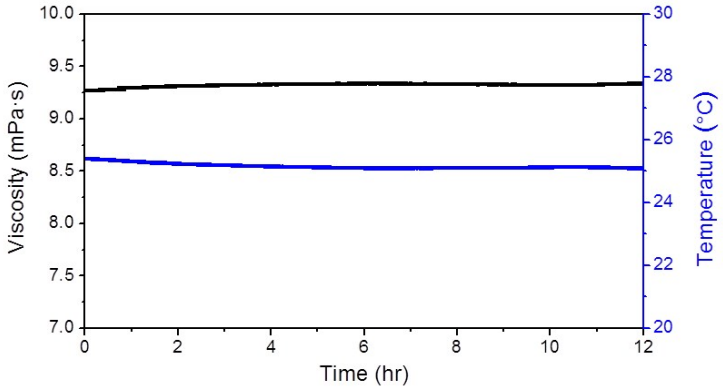

Fig. S2 Viscosity test of the electrolyte containing 1.0 M LiTFSI in TEGDME. There was no clear evaporation and the viscosity remained a constant value.