

## **Supplementary Information**

# **A Novel Zwitterionic Ionic Liquid-based Electrolyte for More Efficient and Safer Lithium-Sulfur Battery**

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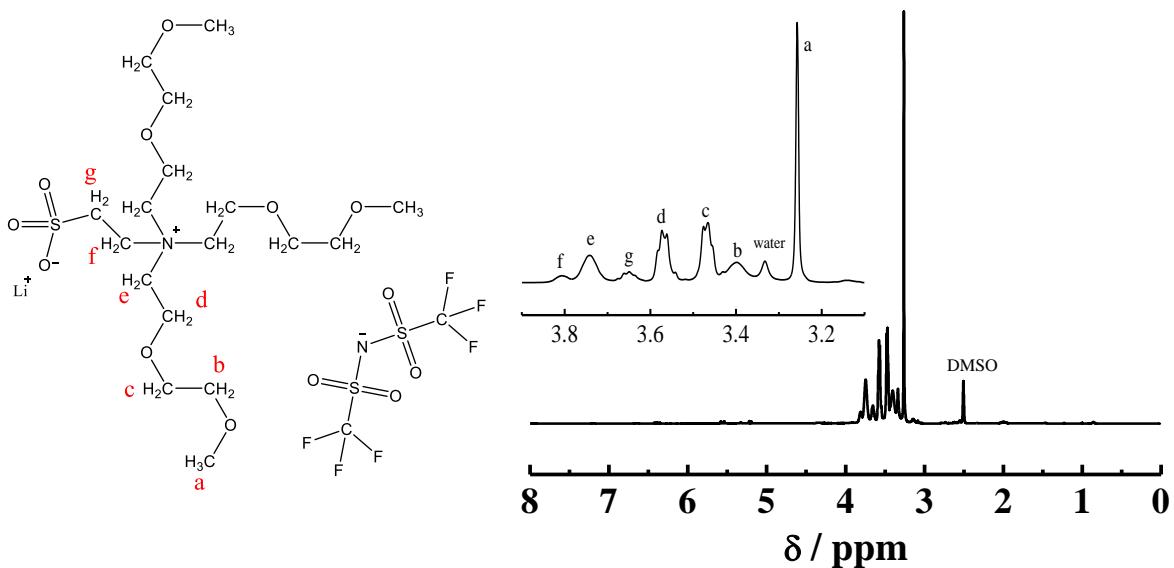
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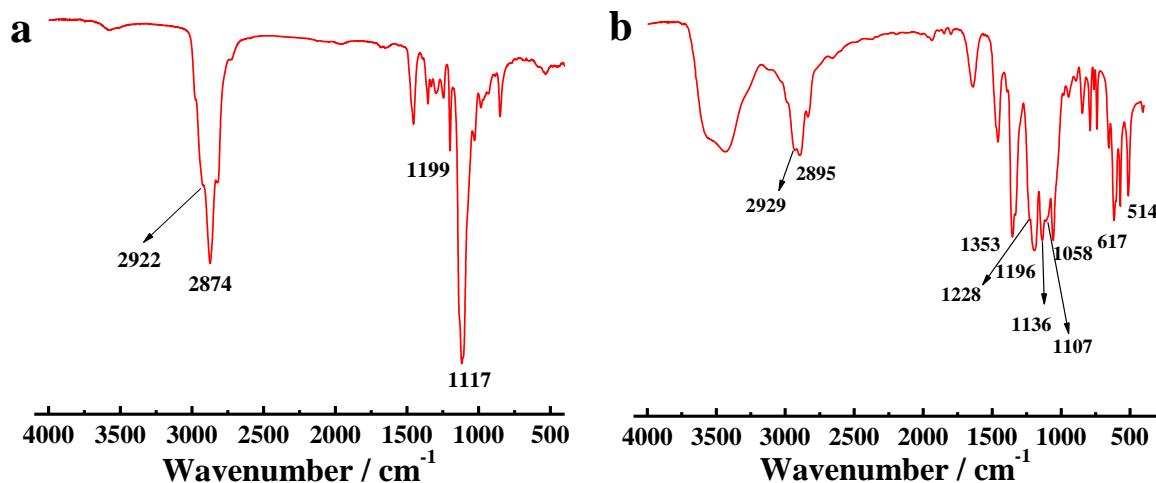
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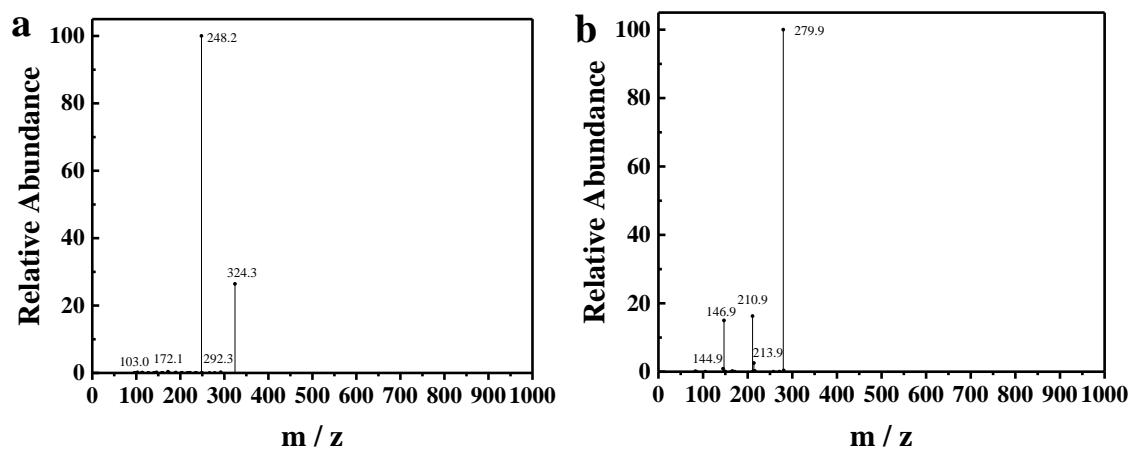
**Figure S1.**  ${}^1\text{H}$ -NMR spectrum of zwitterionic ionic liquid TLTFSI.



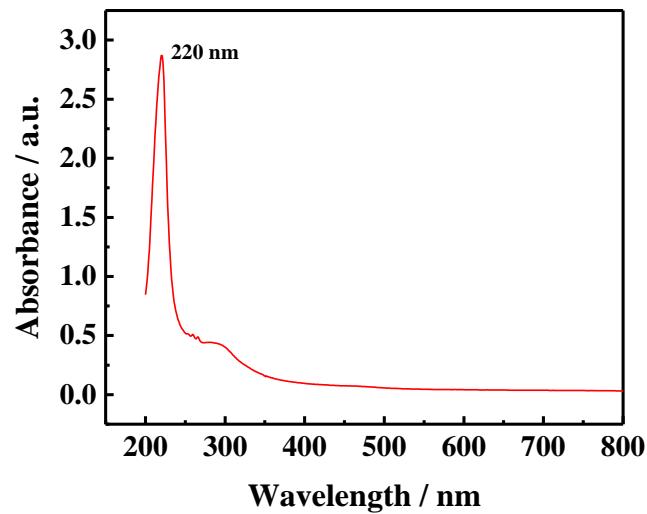
**Figure S2.** The FTIR spectra of (a) TDA and (b) TLTFSI.

The characteristic absorption peaks in **Figure S2a** represent for: v<sub>CH</sub> of  $\text{CH}_3$ & $\text{CH}_2$ : 2922  $\text{cm}^{-1}$ , 2874  $\text{cm}^{-1}$ ; v<sub>C-N</sub>: 1199  $\text{cm}^{-1}$ ; v<sub>C-O-C</sub>: 1117  $\text{cm}^{-1}$ .

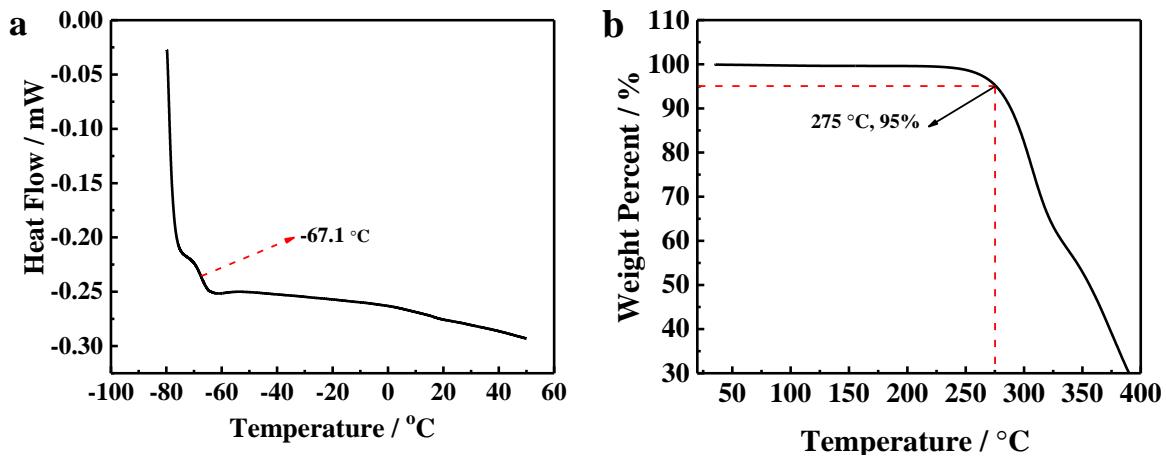
The characteristic absorption peaks in **Figure S2b** represent for: v<sub>CH</sub> of  $\text{CH}_3$ & $\text{CH}_2$ : 2929  $\text{cm}^{-1}$ , 2895  $\text{cm}^{-1}$ ; v<sub>S=O</sub> of the  $\text{SO}_2$  in  $\text{TFSI}^-$ : 1353  $\text{cm}^{-1}$ ; v<sub>CF<sub>3</sub></sub> of the  $\text{SO}_2$  in  $\text{TFSI}^-$ : 1228  $\text{cm}^{-1}$ ; v<sub>S=O</sub> of the  $\text{SO}_3^-$ : 1196  $\text{cm}^{-1}$ , 1058  $\text{cm}^{-1}$ ; v<sub>S=O</sub> of the  $\text{SO}_3^-$ : 617  $\text{cm}^{-1}$ , 514  $\text{cm}^{-1}$ ; v<sub>C-N</sub>: 1136  $\text{cm}^{-1}$ ; v<sub>C-O-C</sub>: 1107  $\text{cm}^{-1}$ .



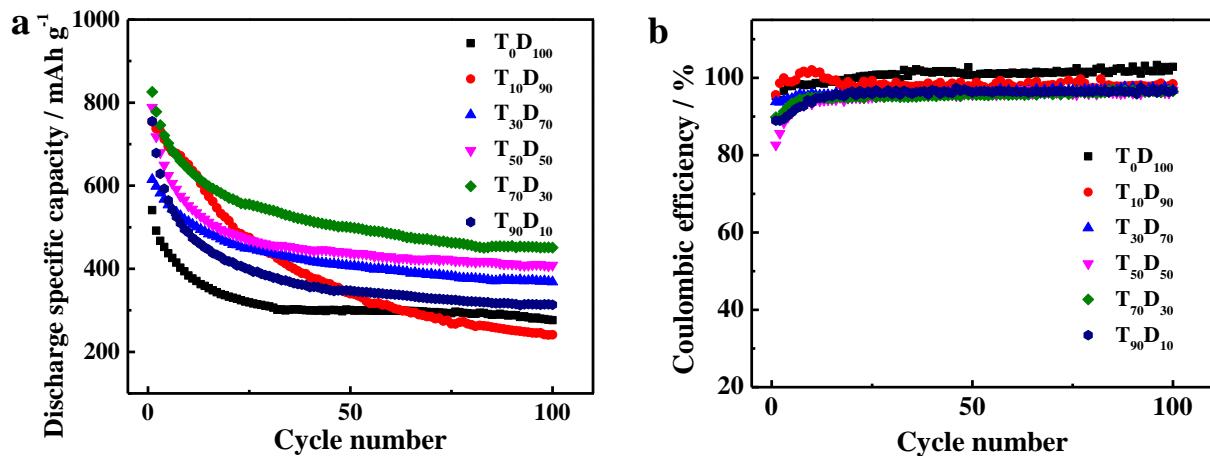
**Figure S3.** The mass spectra of TLTFSI, (a) positive mode and (b) negative mode.



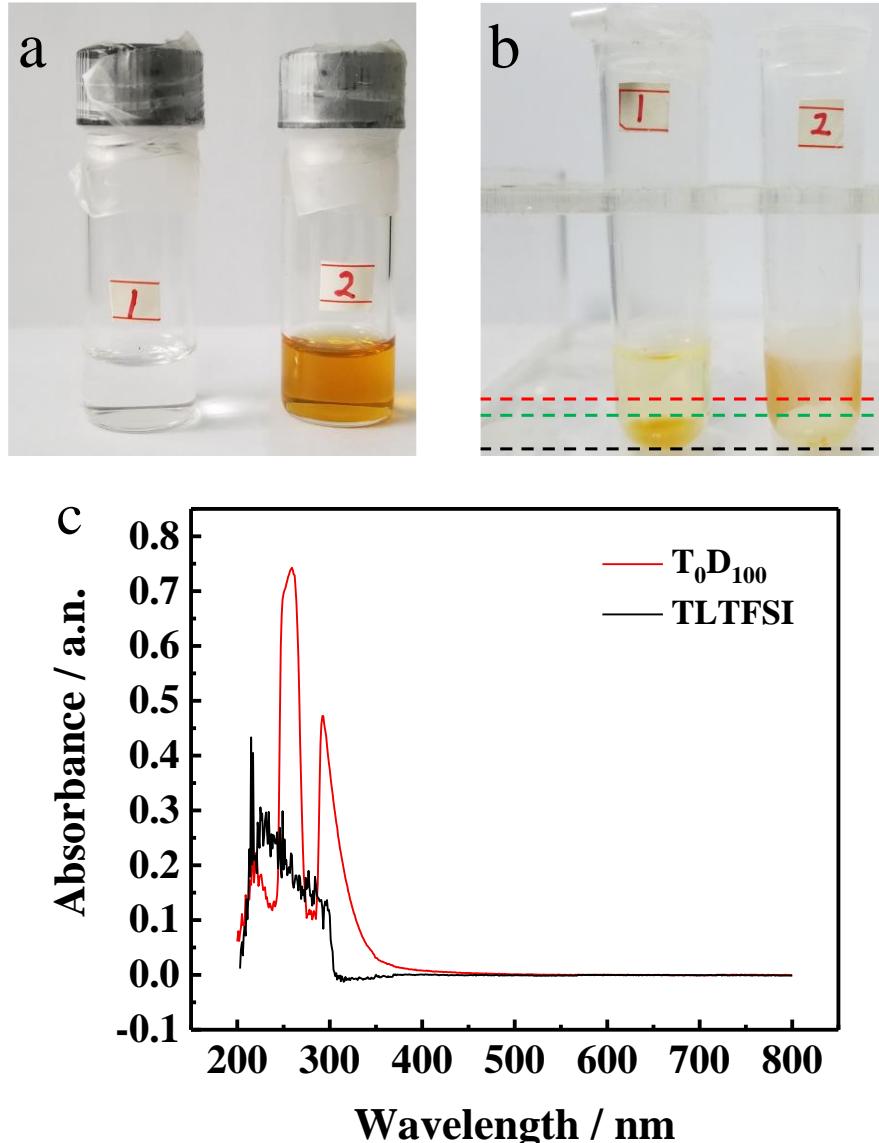
**Figure S4.** The UV-vis spectrum of TLTFSI.



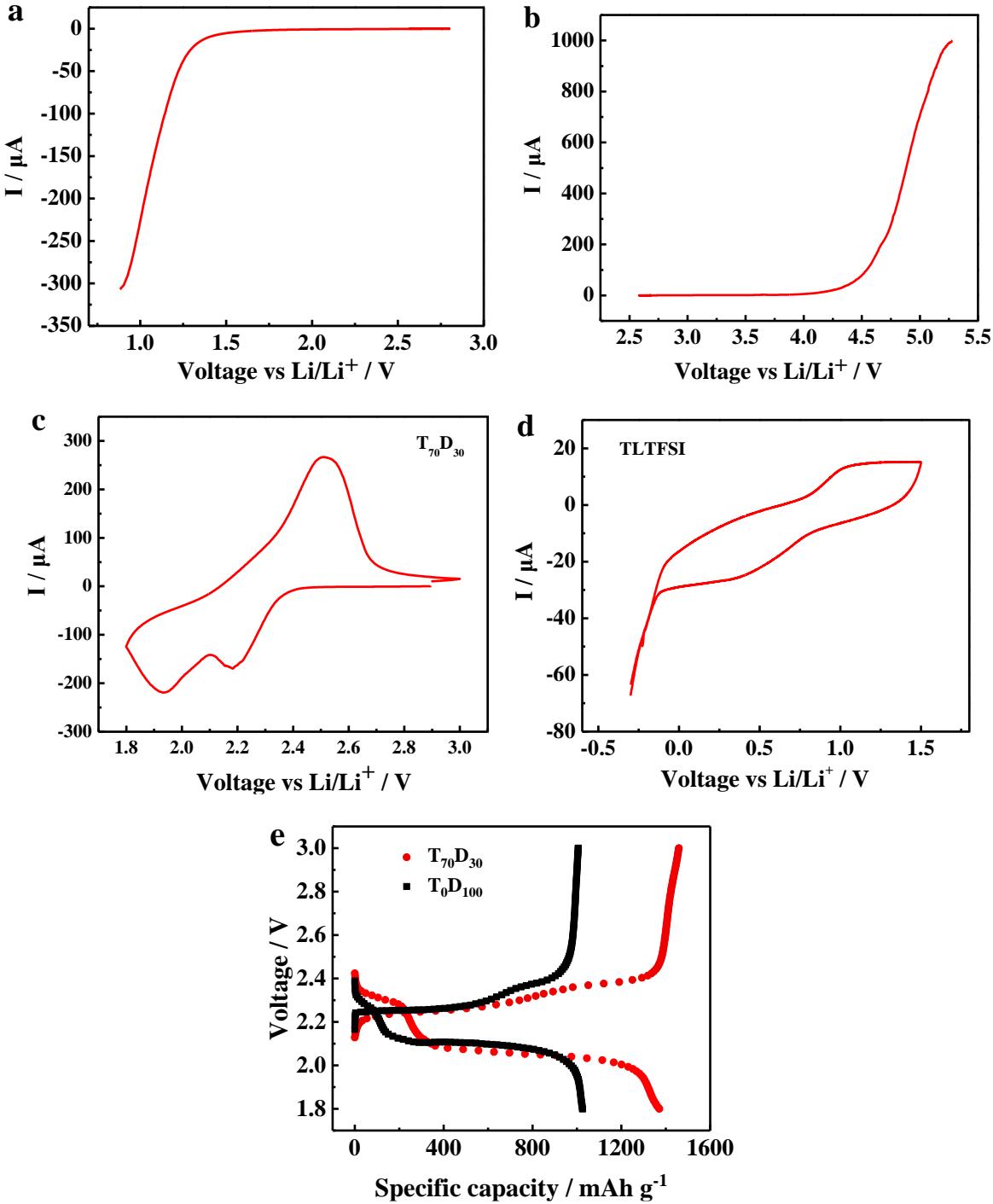
**Figure S5.** (a) DSC and (b) TGA curves of zwitterionic ionic liquid TLTFSI



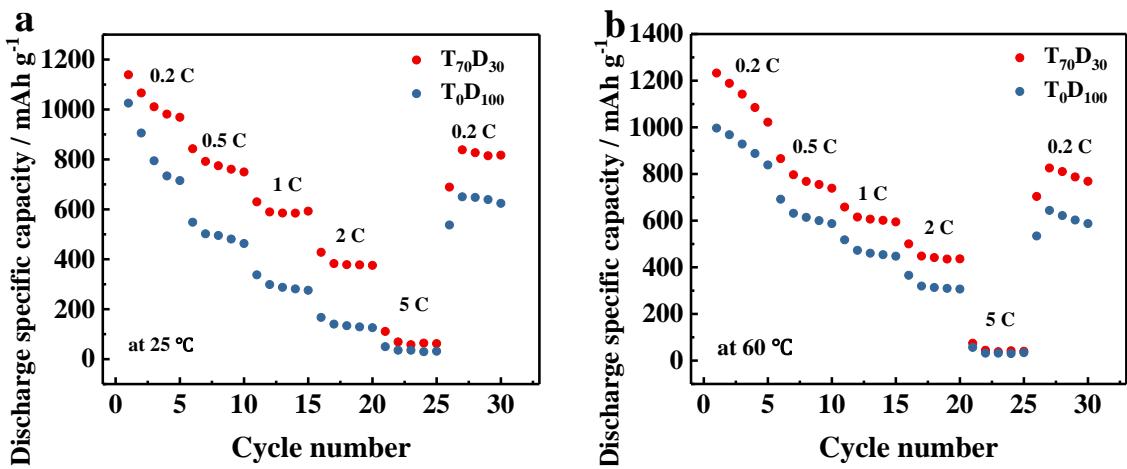
**Figure S6.** The cycle performances of LSBs with T<sub>n</sub>D<sub>m</sub> as the electrolytes recorded with a 0.5 C current density at 25 °C.



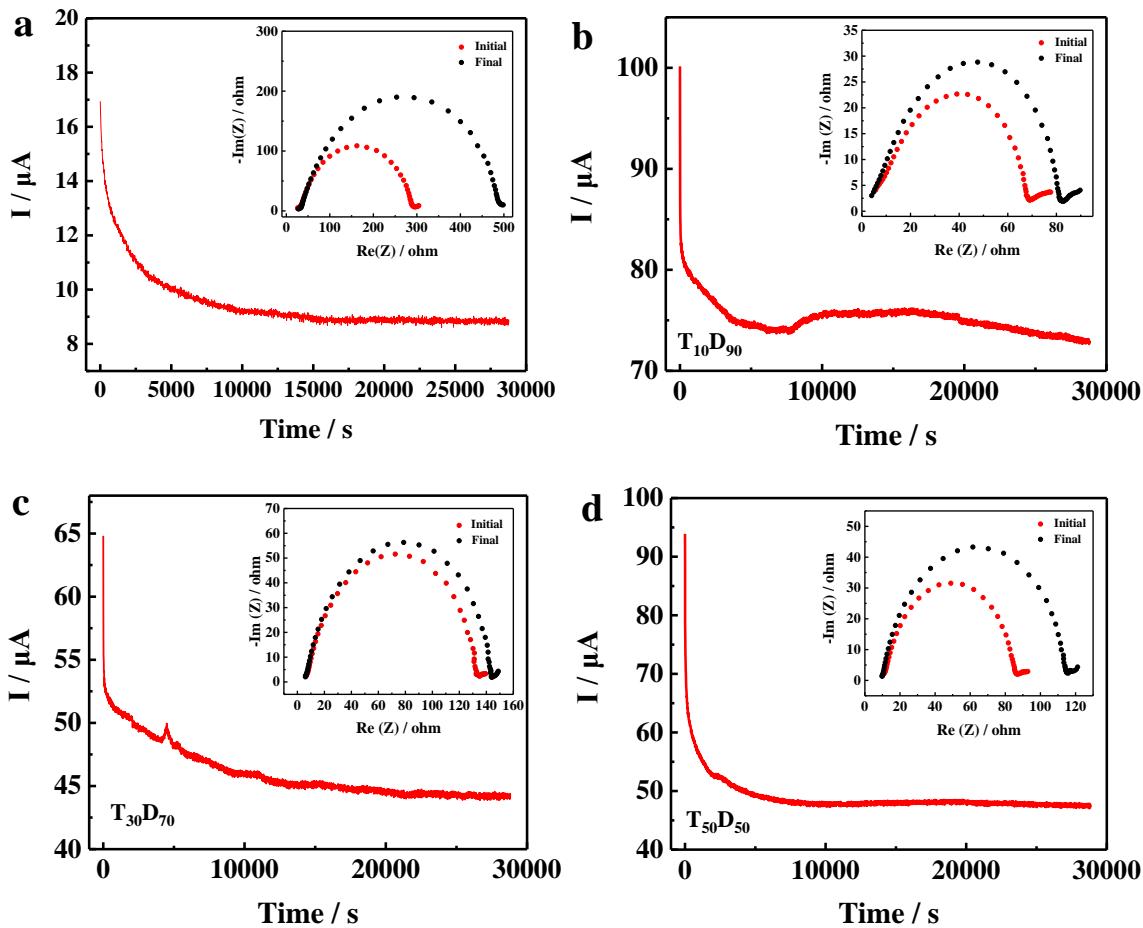
**Figure S7.** (a) The photo of T<sub>0</sub>D<sub>100</sub> and TLTFSI. The labels 1 and 2 represent for T<sub>0</sub>D<sub>100</sub> and TLTFSI, respectively. (b) the photo of T<sub>0</sub>D<sub>100</sub> and TLTFSI with Li<sub>2</sub>S and sulfur powder mixtures. The labels 1 and 2 represent for T<sub>0</sub>D<sub>100</sub> and TLTFSI with Li<sub>2</sub>S and sulfur powder mixtures, respectively. (c) the UV-vis spectra of T<sub>0</sub>D<sub>100</sub> and TLTFSI electrolytes with dissolved Li<sub>2</sub>S<sub>x</sub>.

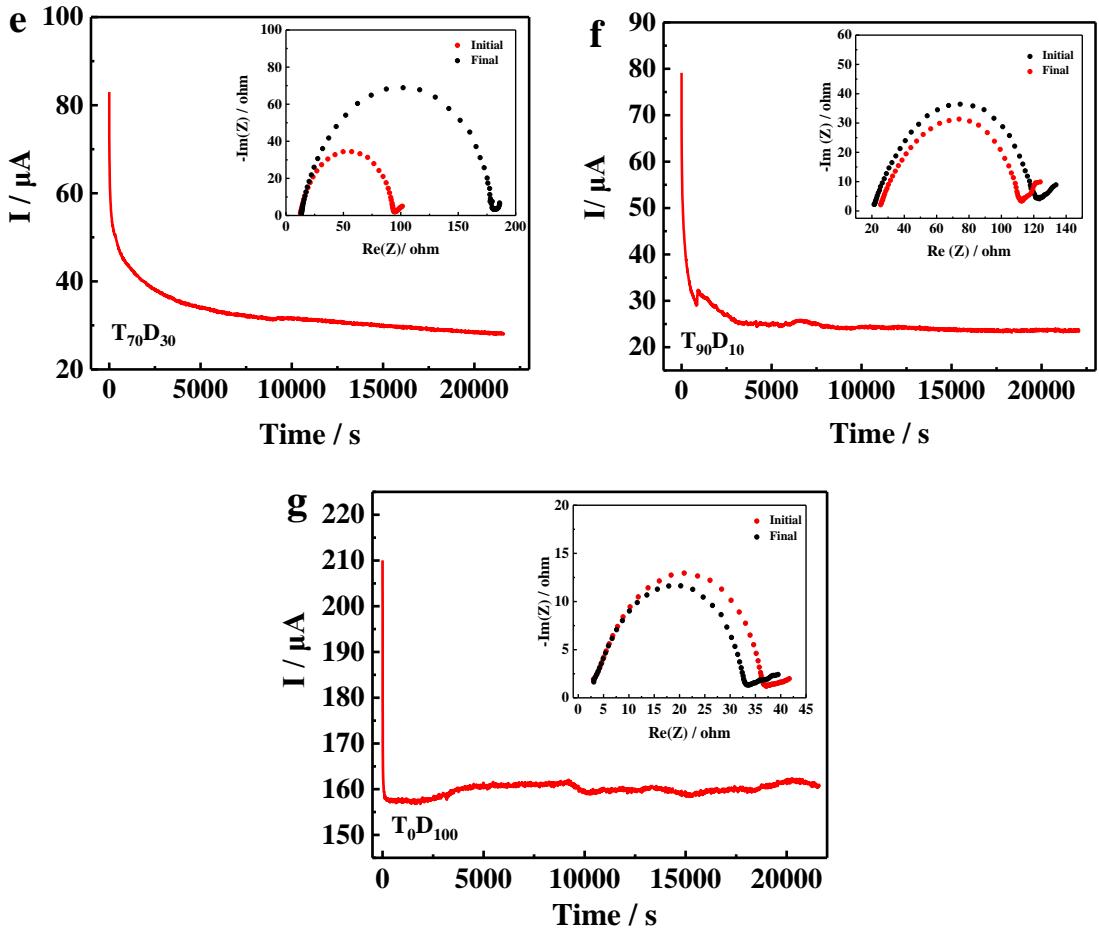


**Figure S8.** (a) LSV curve of  $T_{70}D_{30}$  from 2.75 V to 0.75 V. (b) LSV curve of  $T_{70}D_{30}$  from 2.60 V to 5.25 V. (c) The CV curve of LSB with  $T_{70}D_{30}$  as electrolyte. (d) the CV curve of TLTFSI from -0.3 to 1.5 V. (e) the charge-discharge curves of LSBs with  $T_{70}D_{30}$  and  $T_0D_{100}$  as electrolyte, respectively. The scanning speed for CV is 0.1 mV s<sup>-1</sup> and the current density for charge-discharge test is 0.2 C.



**Figure S9.** The rate performances of LSBs with  $T_{70}D_{30}$  and  $T_0D_{100}$  electrolyte, respectively, measured under various current densities at (a) 25 °C, (b) 60 °C.





**Figure S10.** The current of a [Li metal anode | TLTFSI-based electrolyte | Li metal cathode] cell under a polarization voltage of 10 mV at 25 °C as a function of time. The inset shows the AC impedance spectra of the symmetrical cell before and after polarization. (a) TLTFSI, (b) T<sub>10</sub>D<sub>90</sub>, (c) T<sub>30</sub>D<sub>70</sub>, (d) T<sub>50</sub>D<sub>50</sub>, (e) T<sub>70</sub>D<sub>30</sub>, (f) T<sub>90</sub>D<sub>10</sub> and (g) T<sub>0</sub>D<sub>100</sub>.

The lithium-ion transference number ( $t_{\text{Li}^+}$ ) can be calculated by the following equation:

$$t_{\text{Li}^+} = \frac{I_{ss}R_e^{ss}(\Delta V - I_0R_0)}{I_0R_e^0(\Delta V - I_{ss}R_f)}$$

where  $I_0$ ,  $R_e^0$ , and  $R_0$  are the initial current, electrolyte resistance and lithium interfacial resistance, respectively.  $I_{ss}$ ,  $R_e^{ss}$ , and  $R_f$  are the steady current, electrolyte resistance and lithium interfacial resistance, respectively.  $\Delta V$  is the polarization voltage.

**Video S1.** The degreased cotton after dipping into the T<sub>70</sub>D<sub>30</sub> electrolyte can hardly be lighted by a lighter.

**Video S2.** The degreased cotton after dipping into the T<sub>0</sub>D<sub>100</sub> electrolyte can be easily lighted by a lighter and burned to ashes.