

ZIF-8-based Quasi-solid-state Electrolyte for Lithium Batteries

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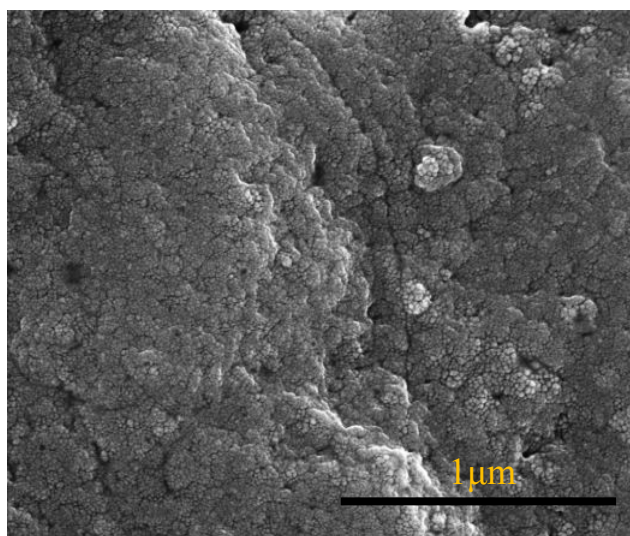


Figure S1. SEM image of ZIF-8 QSSE under 1.00 um.

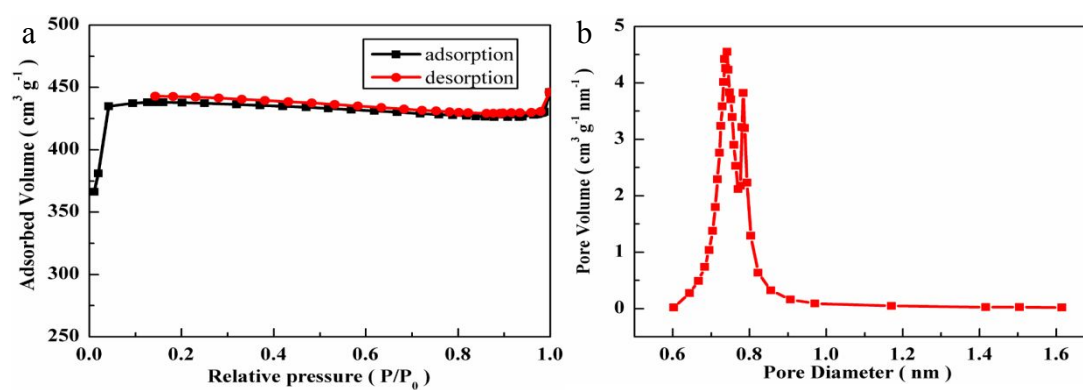


Figure S2. (a) Ar adsorption–desorption analysis of the ZIF-8 powder. (b) The corresponding Horvath-Kawazoe pore size distribution of the ZIF-8 powder.

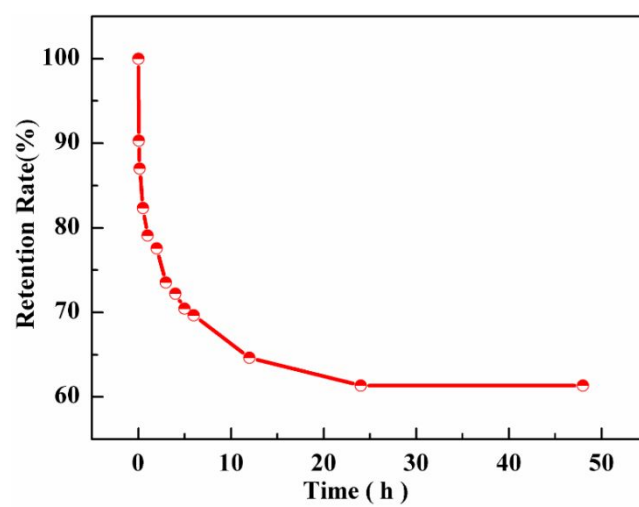


Figure S3. The liquid retention rate of the ZIF-8 QSSE.

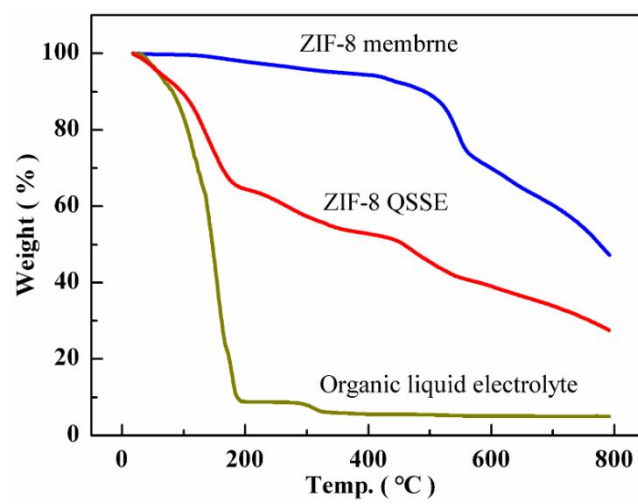


Figure S4. TG thermograms for ZIF-8 membrane, ZIF-8 QSSE and organic liquid electrolyte.

Table S1. Chemical composition content of the ZIF-8 QSSE.

Composition	ZIF-8	PTFE	Organic liquid electrolyte			
			LiPF ₆	EC	DMC	EMC
Melting point (C)	-	327	200	35	2	-14
Boiling point (C)	-	400	-	248	90	107
Content (wt. %)	~ 48%	~ 7%	~ 45%			

In an Ar glovebox, ZIF-8 QSSE was assembled into CR2025 coin cells (SS |ZIF-8 QSSE| SS) for the ionic conductivity test. The electrochemical impedance could be obtained by fitting the Nyquist plot to the model circuit, which the model circuit was RQ. R represents a resistor and Q is a constant phase elements. The ionic conductivity is calculated by using equation (1), as follows:

$$\sigma = \frac{L}{AR} \quad (1)$$

where σ is ionic conductivity, A is the electrode area, R is the resistance, L is the thickness of the electrolyte membrane¹.

Table S2. The fitting data of the SS |ZIF-8 QSSE| SS battery at different temperatures.

Temperature(°C)	R($\Omega \cdot \text{cm}^{-2}$)	Q(S·secn·cm ⁻²)	$\sigma(\text{S} \cdot \text{cm}^{-1})$
25	64.98	1.60E-06	1.05E-04
40	18.14	3.55E-06	3.74E-04
55	12.00	6.66E-06	5.66E-04
70	6.584	7.14E-06	1.03E-03

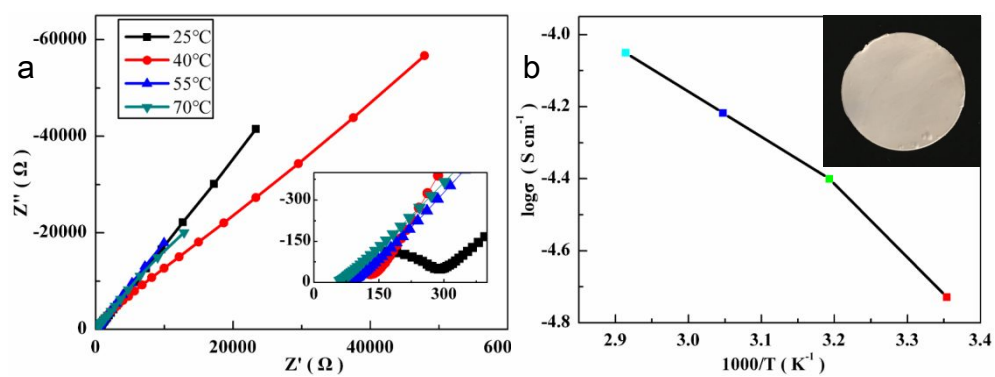


Figure S5. (a) The impedance spectra of the TiO_2 QSSE at various temperature. (b) Ionic conductivity as a function of inverse temperature for the TiO_2 QSSE (inset: the photograph of TiO_2 membrane).

The lithium transference number was calculated using equation (2) as proposed

by Vincent and co-workers,
$$T_{Li+} = \frac{I_{ss} (\Delta V - I_0 R_0)}{I_0 (\Delta V - I_{ss} R_{ss})} \quad (2)$$

the Li /ZIF-8 QSSE/ Li cell was polarized using a dc pulse of 5 mV. The initial (I_0) current and the steady state (I_{ss}) current flowing through the cell during polarization were measured, while R_0 and R_{ss} , represent the resistance values before and after perturbation of the system, respectively. The values of R_0 and R_{ss} were measured before and after perturbation².

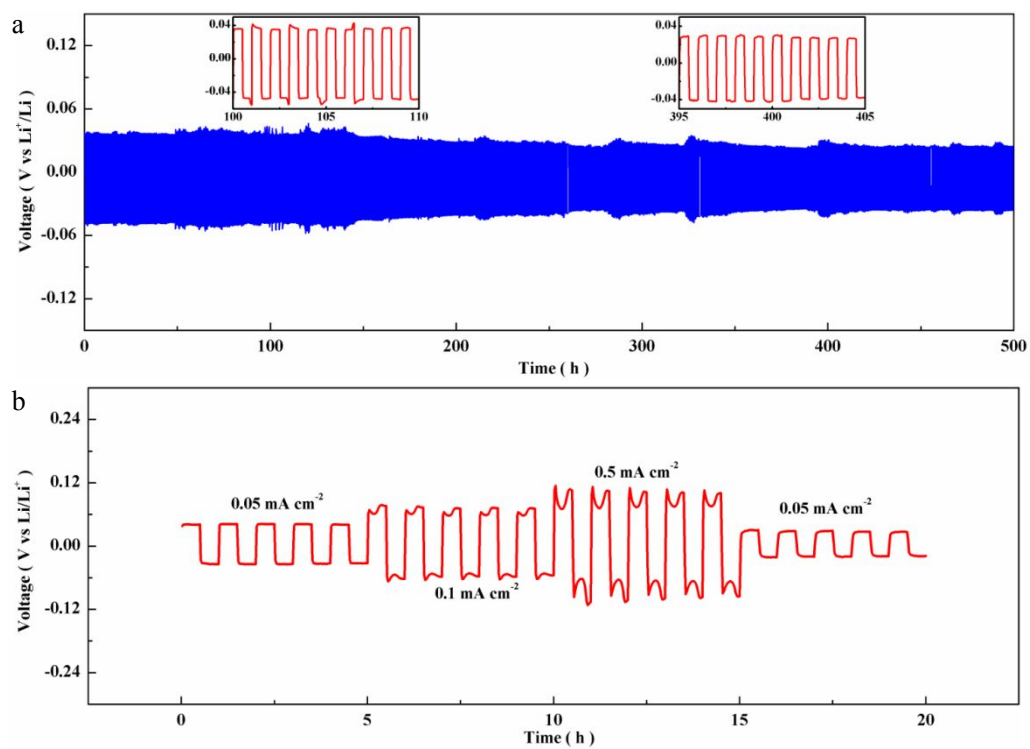


Figure S6. (a) Voltage profiles for the Li | organic liquid electrolyte | Li symmetric cell at the current density of 0.05 mA cm⁻² (inset: the corresponding voltage profiles of the cell at 100-110 cycles, and 395-405 cycles). (b) Voltage profiles for the Li | organic liquid electrolyte | Li symmetric cell at current densities of 0.05 mA cm⁻², 0.1 mA cm⁻² and 0.5 mA cm⁻².

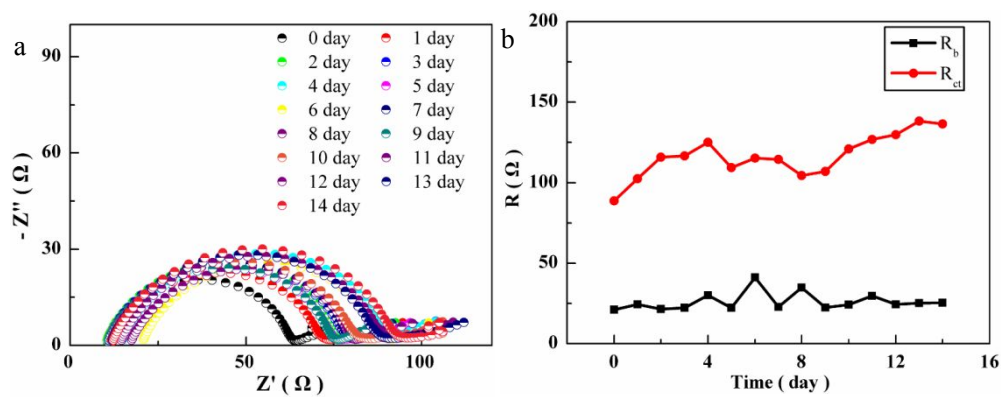


Figure S7. (a) The impedance spectra for the Li | ZIF-8 QSSE | Li symmetric cell with different storing time at 25°C. (b) The variation of bulk resistance (R_b) and interfacial resistance (R_{ct}), as a function of time for the Li | ZIF-8 QSSE | Li symmetric cells at 25 °C.

The CV curves of the liquid lithium battery and the quasi-solid-state lithium battery in **Figure S8** shows a main cathodic peak at near 3.8 V and a anodic peak at 4.0 V in the first cycle. During charging, Lithium ions are removed from the cathode while Co undergoes oxidation from Co^{3+} to Co^{4+} , and in the discharging process, the reverse reaction occurs. At the same time, the CV curves of the quasi-solid-state lithium battery after five cycles nearly completely correspond to the first cycle, indicating that the ZIF-8 QSSE has good reversibility. It can be inferred that the quasi-solid-state lithium battery will show good electrochemical stability. Besides, the CV curves of the quasi-solid-state lithium battery are similar to that of the conventional batteries with organic liquid electrolyte, which indicates the similar mechanism for lithium storage.

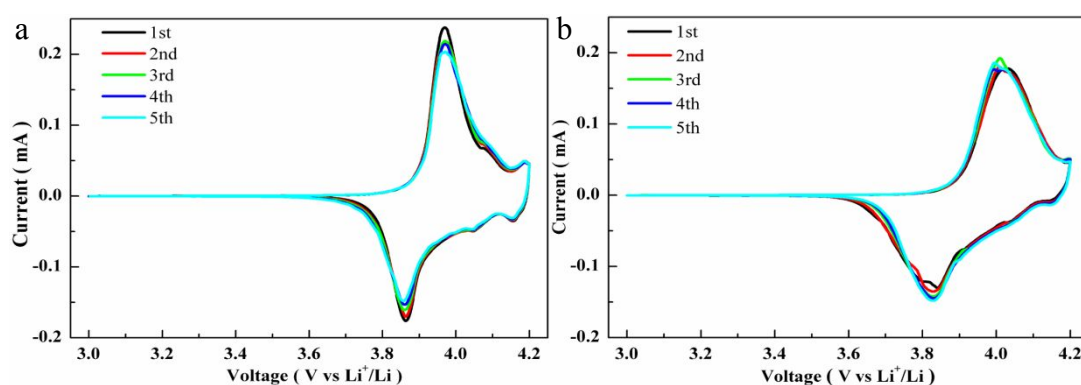


Figure S8. (a)The cyclic voltammetry curve of liquid lithium battery at a scan rate of 0.05 mV s^{-1} in the potential range of 3.0 V~4.2 V. (b) The cyclic voltammetry curve of the quasi-solid-state lithium battery at a scan rate of

0.05mV s⁻¹ in the potential range of 3.0 V~4.2 V.

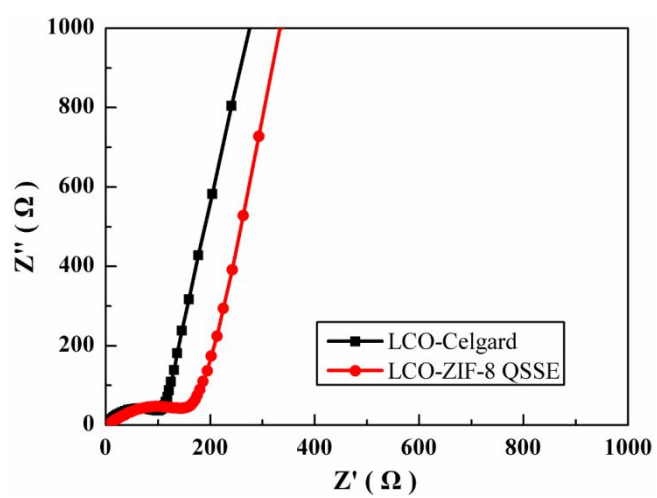


Figure S9. The impedance spectrum of liquid lithium battery and the quasi-solid-state lithium battery at 25 °C.

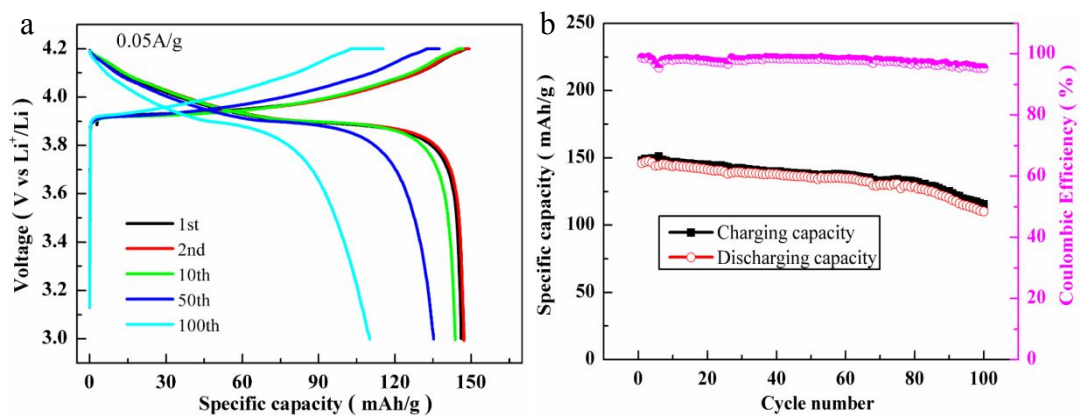


Figure S10. (a) The voltage capacity profile of the liquid lithium battery at 0.05 A g⁻¹. (b) Cycling performance and coulombic efficiency of the liquid lithium battery at 0.05 A g⁻¹.

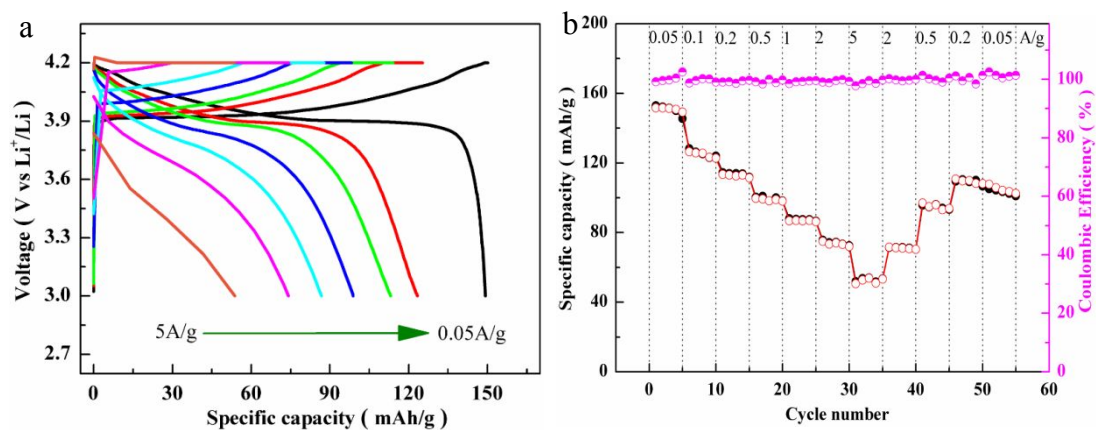


Figure S11. (a) The voltage capacity profiles of the liquid lithium battery at increasing current densities from 0.05 A g⁻¹ to 5 A g⁻¹. (b) The rate capability of the liquid battery at increasing current densities from 0.05 A g⁻¹ to 5 A g⁻¹.

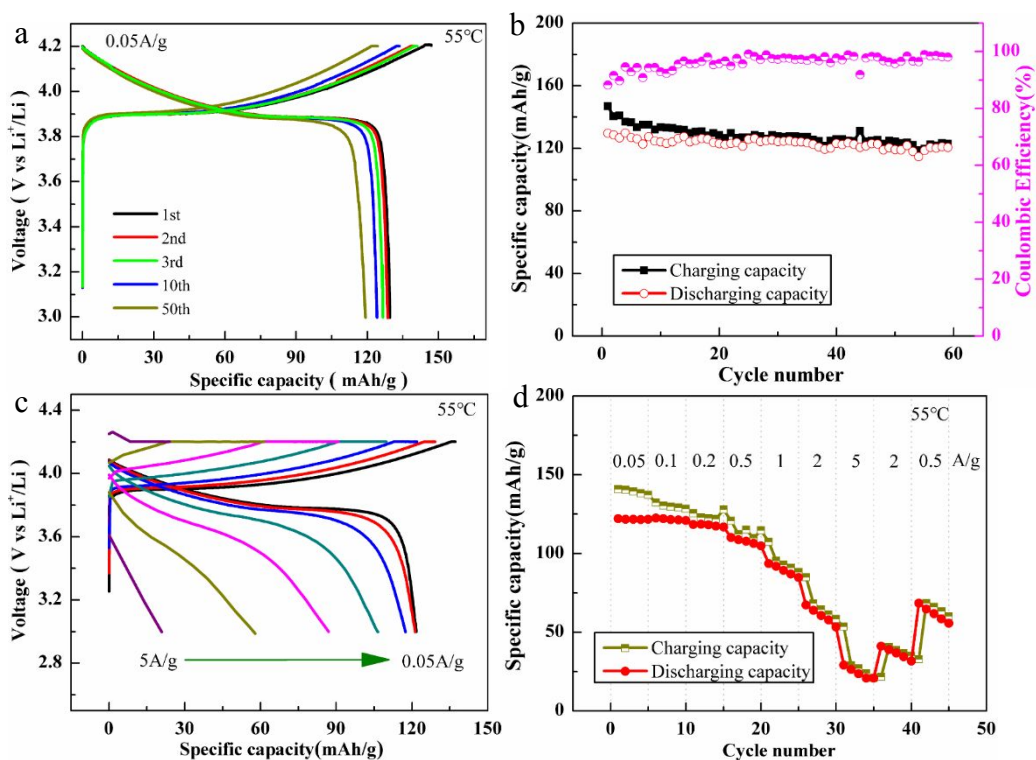


Figure S12. The electrochemical properties of the quasi-solid-state battery at 55°C. (a) The voltage capacity profile of the quasi-solid-state lithium battery at 0.05 A g⁻¹. (b) Cycling performance and coulombic efficiency of the quasi-solid-state lithium battery at 0.05 A g⁻¹. (c) The voltage capacity profiles of the quasi-solid-state lithium battery at increasing current densities from 0.05 A g⁻¹ to 5 A g⁻¹. (d) The rate capability of the quasi-solid-state lithium battery at increasing current densities from 0.05 A g⁻¹ to 5 A g⁻¹.

References

1. Pożyczka, K.; Marzantowicz, M.; Dygas, J. R.; Krok, F., IONIC CONDUCTIVITY AND LITHIUM TRANSFERENCE NUMBER OF POLY(ETHYLENE OXIDE):LiTFSI SYSTEM. *Electrochimica Acta* 2017, 227, 127-135.
2. J. Evans; C.A. Vincent; Bruce., P. G., Electrochemical measurement of transference numbers in polymer electrolytes. *Polymer* 1987, 28, 2324–2328.