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## Supplementary Information (SI)

### A High-Performance Li-O<sub>2</sub>/Air Battery System with Dual Redox Mediators in the Hydrophobic Ionic Liquid-Based Gel-Polymer Electrolyte

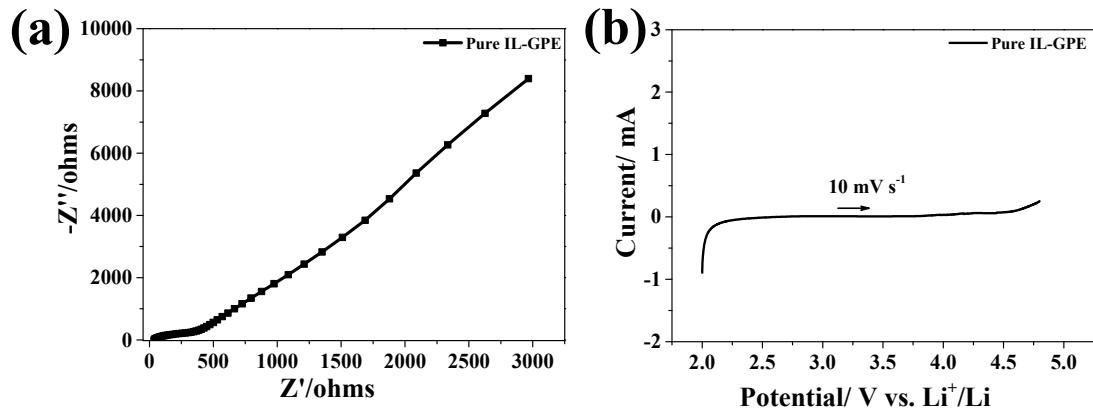
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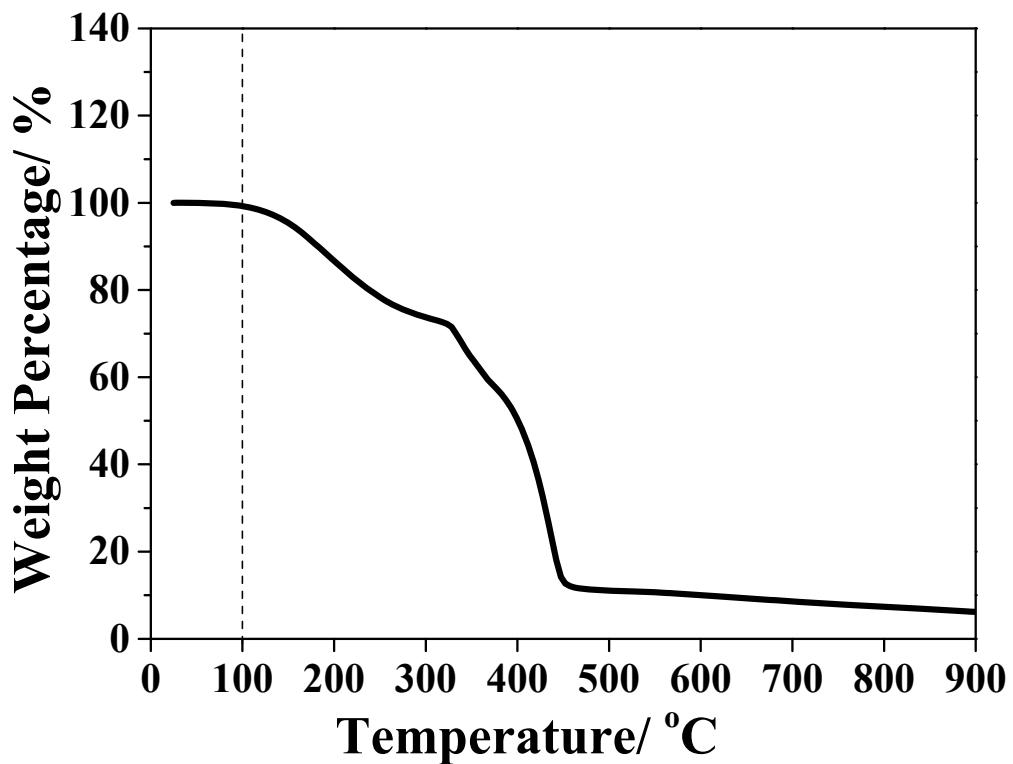
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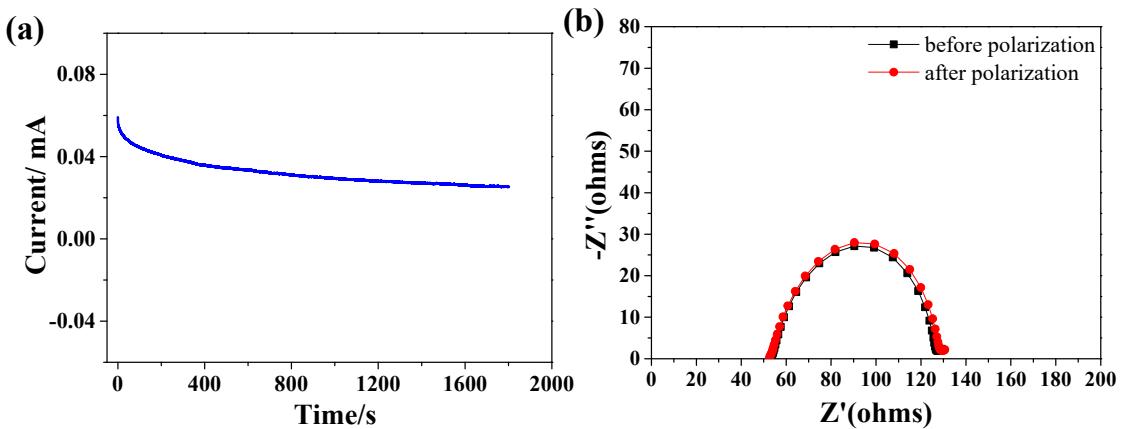


**Figure S1.** (a) EIS measurement of pure IL-GPE, two stainless steel meshes (SSM) are used with pure IL-GPE to form an SSM/GPE/SSM symmetric cell, and the measurement frequency ranges from 0.1 Hz to 100 KHz. (d) Linear sweep voltammetry (LSV) profile of pure IL-GPE and the sweep rate is  $10 \text{ mV s}^{-1}$ .

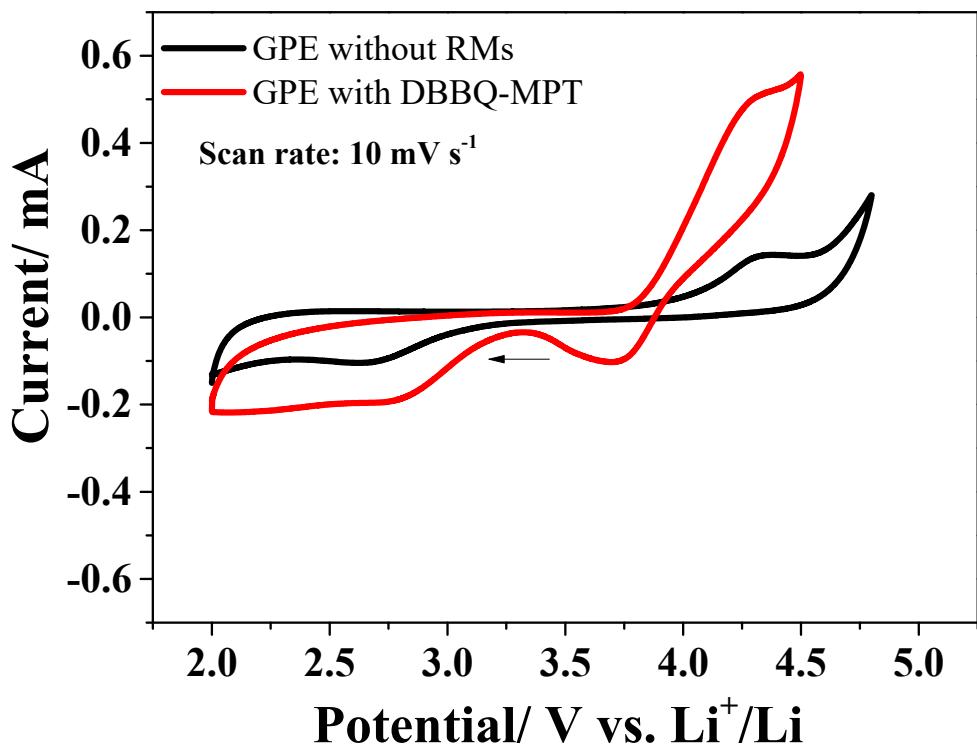


**Figure S2.** TGA curve of DBBQ-MPT-IL-GPE, the heating rate is  $10\text{ }^{\circ}\text{C min}^{-1}$ , and the protected atmosphere is airflow.

It can be found that the GPE undergoes huge weight loss at higher temperatures ( $>100\text{ }^{\circ}\text{C}$ ), which may be attributed to TEGDME volatilization, NMP vaporization, and PVDF-HFP decomposition.



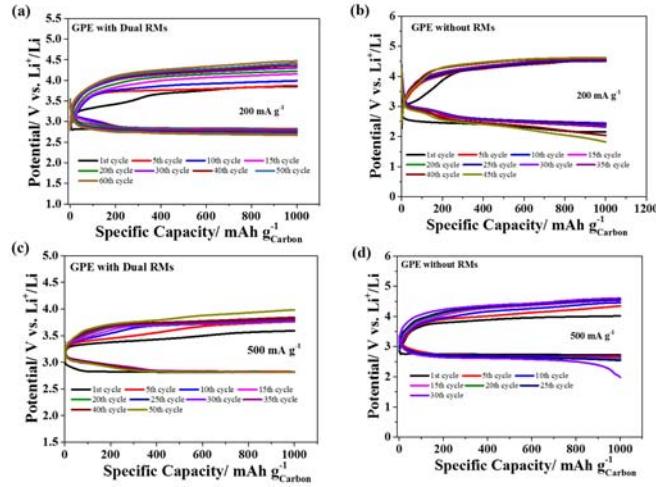
**Figure S3.** (a) Chronoamperometry of the Li/DBBQ-MPT-IL-GPE/Li cell at room temperature. (b) Impedance spectra of DBBQ-MPT-IL-GPE recorded before and after the polarization at room temperature.



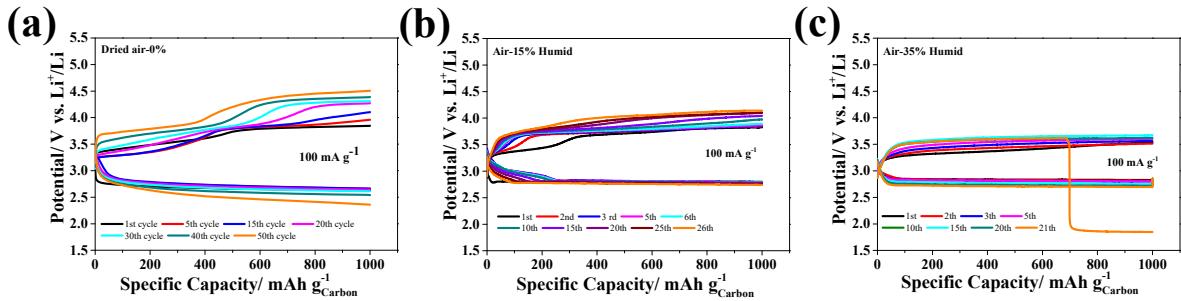
**Figure S4.** Cyclic voltammogram tested in the Li-O<sub>2</sub> cells using Pure IL-GPE (black line) or DBBQ-MPT-IL-GPE (red line) at a scan rate of 10 mV s<sup>-1</sup>.

**Table S1** Summary of cycle performance and discharge-charge voltage gap in various GPE-based Li-O<sub>2</sub> batteries.

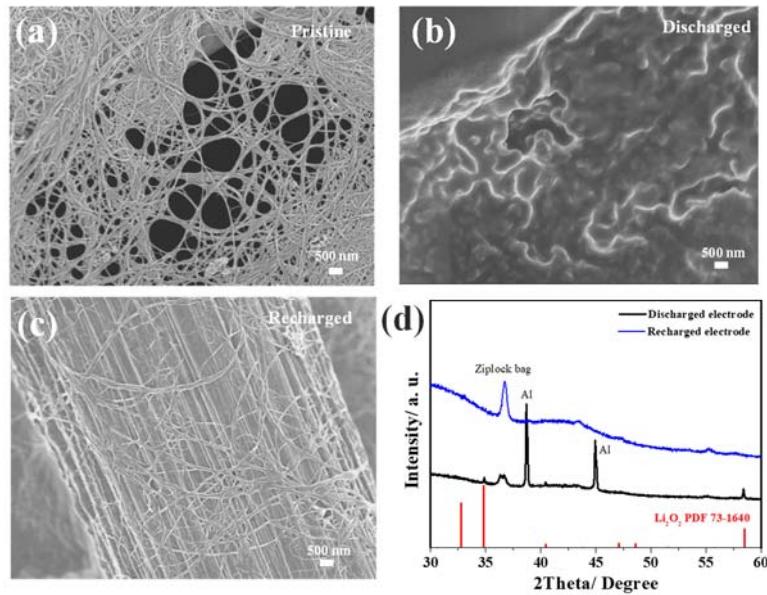
Polymer matrix	Lithium salt	Other components	Testing temperature (°C)/atmosphere e	Discharge-charge voltage gap	Round-trip efficiency	Cycle performance	Reference
PTFE/PFSA-Li	DMSO	25/O <sub>2</sub>	1.56 V	63%	1 A g <sup>-1</sup> , 1000 cycles	mAh g <sup>-1</sup> , 95	S1
PMMA/cross-linked PSt	LiTFSI	SiO <sub>2</sub> /TEGDME	25/O <sub>2</sub>	1.30 V	66.7%	0.25 mA cm <sup>-2</sup> , 2, 1 mAh cm <sup>-2</sup> , >60 cycles	S2
PE/P(MMA-st)	LiTFSI	LAGP/SiO <sub>2</sub> /TEGD ME	50/O <sub>2</sub>	1.45 V	65%	200 mA g <sup>-1</sup> , 1000 mAh g <sup>-1</sup> , >350 cycles	S3
PVDF-HFP	LiTFSI	PYR <sub>14</sub> TFSI	140/Air	1.50 V	64.3%	10 A g <sup>-1</sup> , 500 cycles	mAh g <sup>-1</sup> , S4
PVDF-HFP/TMPET	LiTFSI	PP13TFSI+TEGD ME (volume ratio: 3:1)/DBBQ/MPT	25/O <sub>2</sub>	1.02 V	72%	100 mA g <sup>-1</sup> , 1000 mAh g <sup>-1</sup> , >380 cycles	This work, k



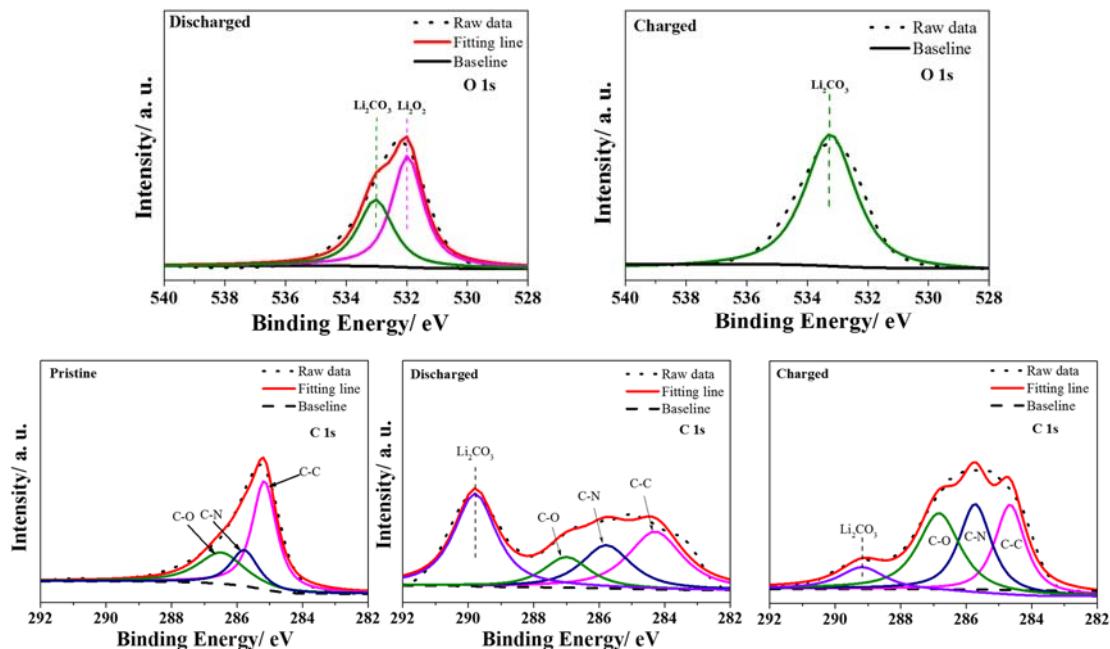
**Figure S5.** Discharge–charge voltage profiles of the DBBQ-MPT-IL-GPE-based (a and c) or pure IL-based Li–O<sub>2</sub> batteries (b and d) at different current densities (200 and 500 mA g<sup>−1</sup>).



**Figure S6.** Discharge–charge voltage profiles of the DBBQ-MPT-IL-GPE-based Li–O<sub>2</sub> batteries with different humidities at the applied current density: 100 mA g<sup>−1</sup> and the fixed specific capacity (1000 mAh g<sup>−1</sup>).



**Figure S7.** Ex situ SEM images of the SWCNT cathodes in the pure IL-GPE-based  $\text{Li-O}_2$  batteries at different stages, (a) pristine, (b) discharged, and (c) recharge. (d) Ex situ XRD pattern of the SWCNT cathodes at different stages.



**Figure S8.** Ex situ XPS spectra of O 1s and C 1s of the SWCNT cathodes in the DBBQ-MPT-IL-GPE-based  $\text{Li-O}_2$  batteries at different stages.

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## References

- Ref. S1: Shi, Y.; Wu, C.; Li, L.; Yang, J. A Lithiated Perfluorinated Sulfonic Acid Polymer Electrolyte for Lithium–Oxygen Batteries, *J. Electrochem. Soc.* **2017**, *164*, A2031-A2037.
- Ref. S2: Woo, H. S.; Moon, Y. B.; Seo, S.; Lee, H. T. Semi-Interpenetrating Polymer Network Composite Gel Electrolytes Employing Vinyl-Functionalized Silica for Lithium–Oxygen Batteries with Enhanced Cycling Stability, *ACS Appl. Mater. Interfaces* **2018**, *10*, 687–695.
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- Ref. S4: Pan, J.; Li, H.; Sun, H.; Zhang, Y.; Wang, L.; Liao, M.; Sun, X. M.; Peng, H. A Lithium–Air Battery Stably Working at High Temperature with High Rate Performance, *Small* **2018**, *14*, 1703454.