Neuron-inspired self-healing PU-based hybrid solid-state electrolyte enable high-performance lithium metal batteries

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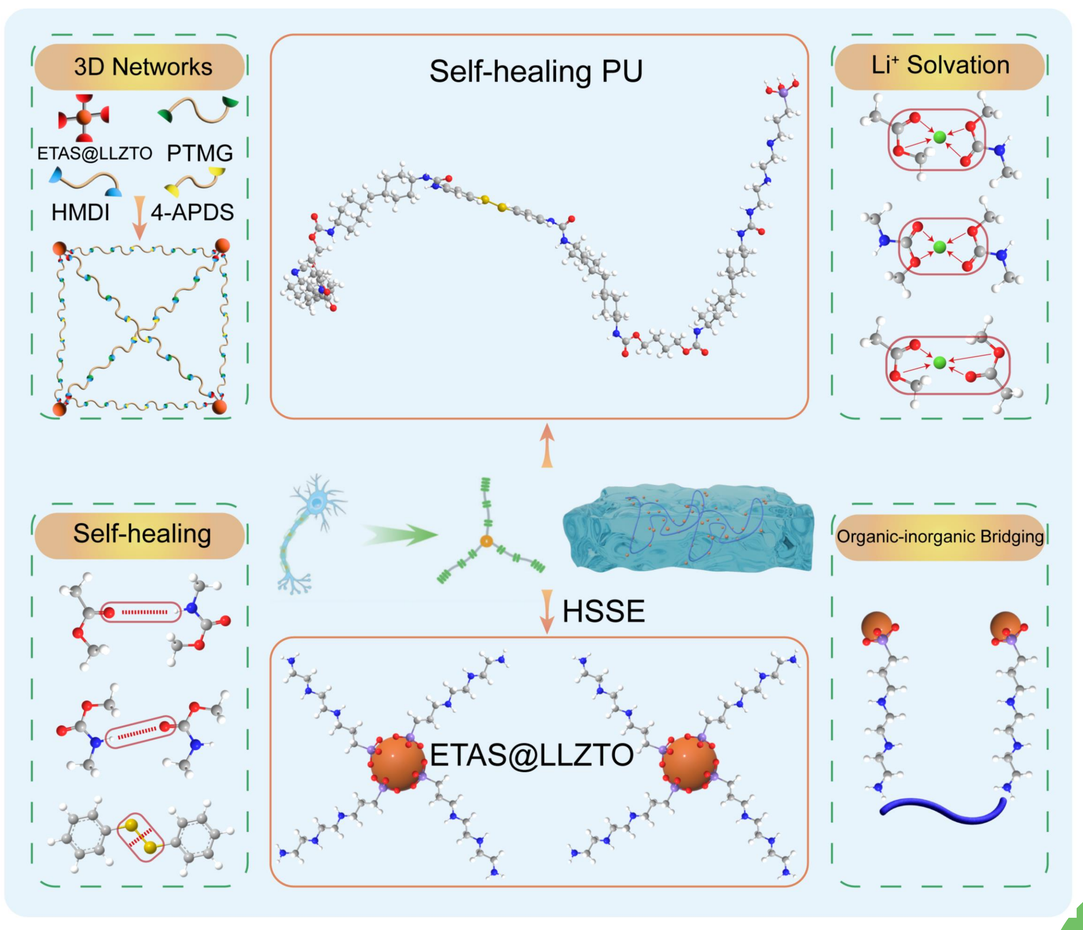
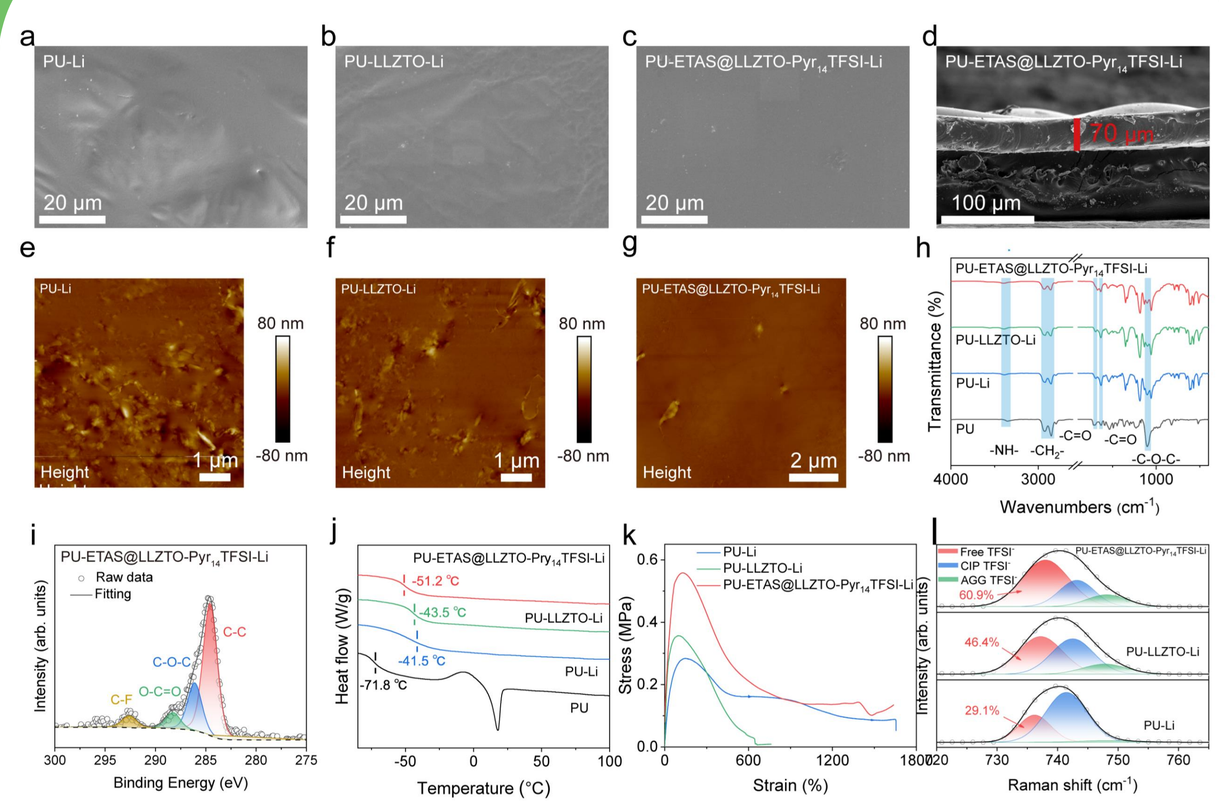
Background

- Although rechargeable lithium-ion batteries have been widely used in electronic products, their traditional liquid electrolytes are not optimal since they present problems such as easy leakage, flammability, narrow operating temperature range, and poor safety.
- In order to solve the above problems and at the same time increase the energy density of the battery, solid-state lithium-ion batteries have received more and more attention[1-3].
- Here, inspired by cross-linked neurons, we developed a self-healing polyurethane (PU)-based hybrid solid-state electrolyte (HSSE) based on ETAS-modified LLZTO as a multi-site crosslinking center and applied it to solid-state lithium-ion batteries.

1. HSSE synthesis



2. HSSE characterization



- HSSE has a smooth surface, less crystallization area, good elongation at break, and excellent lithium salt dissociation capacity.

3. Electrochemical performance

- High ionic conductivity, wide electrochemical window, large exchange current density, low interface impedance, and good performance of Li/Li symmetric cells have been obtained.

4. SEM/XPS characterization

- In the Li/PU-ETAS@LLZTO-Pyr$_{14}$TFSI-Li/Li symmetric cells, the surface of the lithium sheets is smooth, and the solid electrolyte interface is rich in inorganic substances such as LiF and Li_2O, which is extremely beneficial for forming a stable interface.

Conclusions

- Inspired by cross-linked neurons, a self-healing PU-based HSSE was successfully synthesized, and an ionic conductivity of 3.4×10^{-4} S cm^{-1} was obtained at 30°C.
- The organic-inorganic interface hybridisation greatly reduces interface impedance and increases Li^+ flux, while forming an inorganic interface rich in LiF, Li_2O and Li_3N, thereby inhibiting the growth of lithium dendrites. As a result, the Li/Li symmetric cell cycles for over 2000 h.

Reference

[1] Fei Pei, Yimeng Huang, et al. Adv. Mater. 2024, 2409269.

[2] Sha Li, Fei Pei, et al. Adv. Funct. Mater. 2024, 2415495.

[3] Jing Chen, Xuetian Deng, et al. Angew. Chem. 2023, 135, e202307255.

Acknowledgements

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