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HIGHLIGHTS

Advancing electrolytes for sustainable cryogenic Li||Cl_2 batteries

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Rechargeable lithium||chlorine (Li||Cl_2) batteries represent a highly promising platform for high-energy-density storage [1]. Pioneering investigations, including heteroatom-doped bicontinuous carbon hosts [2], multiwalled carbon nanotubes [3], and functional metal-organic frameworks [4], have been devised to confine chlorine species and mitigate the sluggish mass and electron transport limitations. However, thionyl chloride ($SOCl_2$)-based electrolytes face a critical bottleneck of high reactivity accompanied by severe corrosivity. Accordingly, we explored ester-based electrolytes that mitigated the corrosion issue of conventional systems [5]. However, operating thionyl chloride ($SOCl_2$)-based batteries at cryogenic temperatures remains hindered by intrinsic interfacial instability. The highly reactive solvation environment, dominated by chloroaluminate anions, inevitably triggers solvent decomposition and results in the accumulation of insulating byproducts, leading to rapid cell failure.

To address this limitation, Zhi and co-workers [6] recently reported a ternary solvation sheath reconfiguration (TSSR) approach for cryogenic Li||Cl_2 batteries in *Nature Communications*. Rather than relying on physical confinement or trial-and-error additive screening, the authors for the first time modulated the electrolyte coordination chemistry by introducing lithium trifluoromethanesulfonate (LiOTf) as a primary solvation modifier, which is guided by the selection criteria established through correlating the donor numbers and HOMO energy levels of various conventional anions (Figure 1a). The OTf^- anion exhibits a distinctive thermodynamic profile that facilitates its intervention in the coordination environment. The resulting structural transition is depicted in the

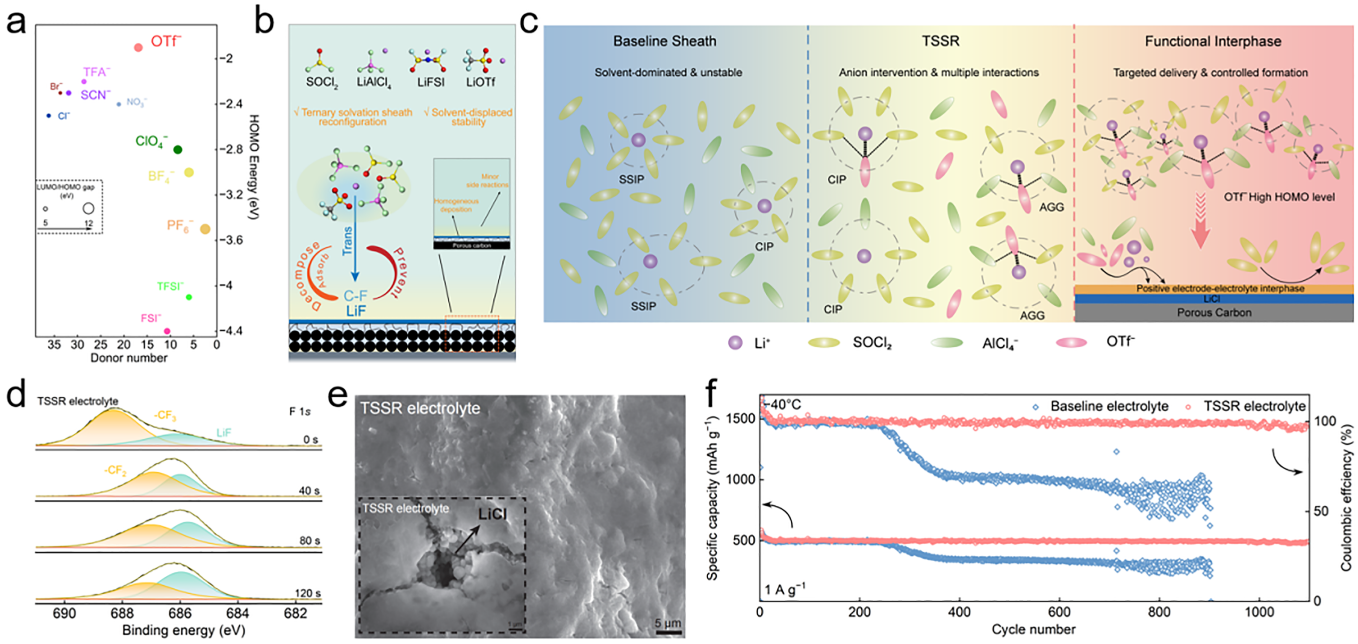

Figure 1 (Color online) (a) Donor number *versus* HOMO energy for various anions commonly used in battery electrolytes. Each colored bubble denotes a different anion (*e.g.*, OTf^-, TFA^-, SCN^-, Br^-, NO_3^-, Cl^-, ClO_4^-, BF_4^-, PF_6^-, $TFSI^-$, FSI^-). The bubble size indicates the corresponding LUMO-HOMO gap (5 to 12 eV). (b) Schematic of the electrolyte solvation structure and positive electrode reaction in ternary solvation sheath reconfiguration (TSSR) electrolytes. (c) Conceptual illustration of the TSSR mechanism. The TSSR mechanism leverages the OTf^- anion to interact with all electrolyte components, reconfiguring the Li^+ solvation sheath from a solvent-dominated state to an anion-rich aggregate (AGG) structure and enabling the controlled *in-situ* formation of a functional positive electrode-electrolyte interphase. (d) F 1s X-ray photoelectron spectroscopy (XPS) spectra under sequential sputtering 0, 40, 80, 120 s of positive electrodes in the the TSSR electrolyte. (e) Scanning electron microscopy (SEM) images of positive electrodes cycled in the TSSR electrolyte. (f) Sustained cryogenic cycling stability of the TSSR electrolyte at −40 °C and 1 A g^{-1}. Adapted from Ref. [6]. Copyright©2026, Springer Nature.

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schematics of Figure 1b, c. The TSSR mechanism leverages the OTf$^-$ anion to interact with all electrolyte components, competitively displacing $SOCl_2$ solvent molecules from the Li$^+$ primary solvation shell. This reconfiguration transforms the system from a solvent-dominated state to an anion-rich aggregate (AGG) structure. This fundamental shift effectively lowers the chemical activity of the free solvent, mitigating its spontaneous decomposition.

Furthermore, this reconfigured AGG state governs the interfacial chemistry. X-ray photoelectron spectroscopy (XPS) depth profiling (Figure 1d) confirms the *in-situ* construction of a functional positive electrode-electrolyte interphase, characterized by a robust, depth-dependent accumulation of LiF. Scanning electron microscopy (SEM) (Figure 1e) further visualizes the structural integrity of the positive electrode after cycling, avoiding the thick byproduct passivation observed in conventional systems. Consequently, this interphase effectively suppresses parasitic side reactions while maintaining rapid ion transport kinetics. As shown in Figure 1f, the Li||Cl_2 cells utilizing the TSSR electrolyte demonstrate sustained cryogenic cyclic stability, achieving over 1100 cycles at a specific current of 1 A g^{-1} at −40 °C.

The work by Zhi and co-workers [6] provides a coordination chemistry strategy to address $SOCl_2$-induced interfacial instability, representing a significant breakthrough in this field. This specific coordination intervention effectively suppresses the intrinsic corrosivity and mitigates the thermodynamic instability of $SOCl_2$, providing a distinct physical chemistry framework for stabilizing the highly reactive halogen-based liquid components. However, to facilitate the practical deployment, the concentration optimization of the introduced salts is essential for balancing electrochemical performance with scalable cost-effectiveness [7]. Furthermore, the interfacial evolution at the Li metal anode under ultra-low temperatures warrants systematic investigation, since sluggish Li diffusion kinetics can aggravate dendrite formation during long-term cycling.

Conflict of interest

The authors declare no conflict of interest.

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