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Interfacial-tension-driven micro-emulsion electrolytes enable bipolar stability in Li-metal batteries

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Lithium metal batteries (LMBs), by replacing graphite with lithium metal anodes, significantly enhance energy density, offering a revolutionary solution for electric long-range vehicles and vertical take-off and landing aircraft. However, the ultra-low potential of lithium metal (-3.04 V vs. Standard hydrogen electrode) is thermodynamically incompatible with conventional carbonate-based electrolytes, necessitating the formation of a stable passivation layer at the electrode interface to enable reversible lithium deposition/stripping.[1]

To enable the formation of a protective passivation layer, extensive research has focused on designing advanced electrolytes through strategies such as high-concentration electrolytes, localized high-concentration electrolytes, and weakly solvating electrolytes. Among these approaches, a significant focus of current electrolyte design centers on modulating the Li^+-anion coordination structure to construct a lithium fluoride (LiF)-rich solid electrolyte interphase (SEI) via anion decomposition at the anode to suppress dendrite growth.[2] However, this strategy faces fundamental challenges at the high-voltage cathode: during charging, the unidirectional electric field coupled with the Li^+ concentration gradient creates an "anion-deficient and Li^+-enriched" environment at the cathode interface (Figure 1A), severely restricting anion decomposition to form a stable, LiF-rich cathode electrolyte interphase (CEI). This unstable CEI triggers continuous electrolyte decomposition and cathode material failure, ultimately leading to capacity decay in LMBs. This fundamental imbalance between the LiF-rich SEI achievable at the anode and the LiF-deficient CEI at the high-voltage cathode represents the central barrier preventing conventional electrolytes from concurrently stabilizing both electrodes, critically impeding the realization of Li-metal batteries with energy densities ≥ 500 Wh kg^{-1}.

In a recent research publication in *Nature*, Huang et al. introduced a novel micro-emulsion electrolyte design strategy.[3] This approach driven by liquid-liquid interfacial tension ($\gamma_{L\text{-}L}$) circumvents the influence of electric fields and concentration gradients on the solvation structure, enabling the simultaneous construction of stable SEI and CEI layers (Figure 1B). Based on this micro-emulsion electrolyte, the 7.5 ampere-hour (Ah) Li||$LiNi_{0.8}Co_{0.1}Mn_{0.1}$ pouch cell delivers 547 Wh kg^{-1} energy density with 79% capacity retention after 155 cycles.

The microemulsion electrolyte introduces immiscible (IM) 1,1,1,2,2,4,5,5,5-nonafluoro-4-(trifluoromethyl)-3-pentanone (PFP) and amphiphilic (AM)

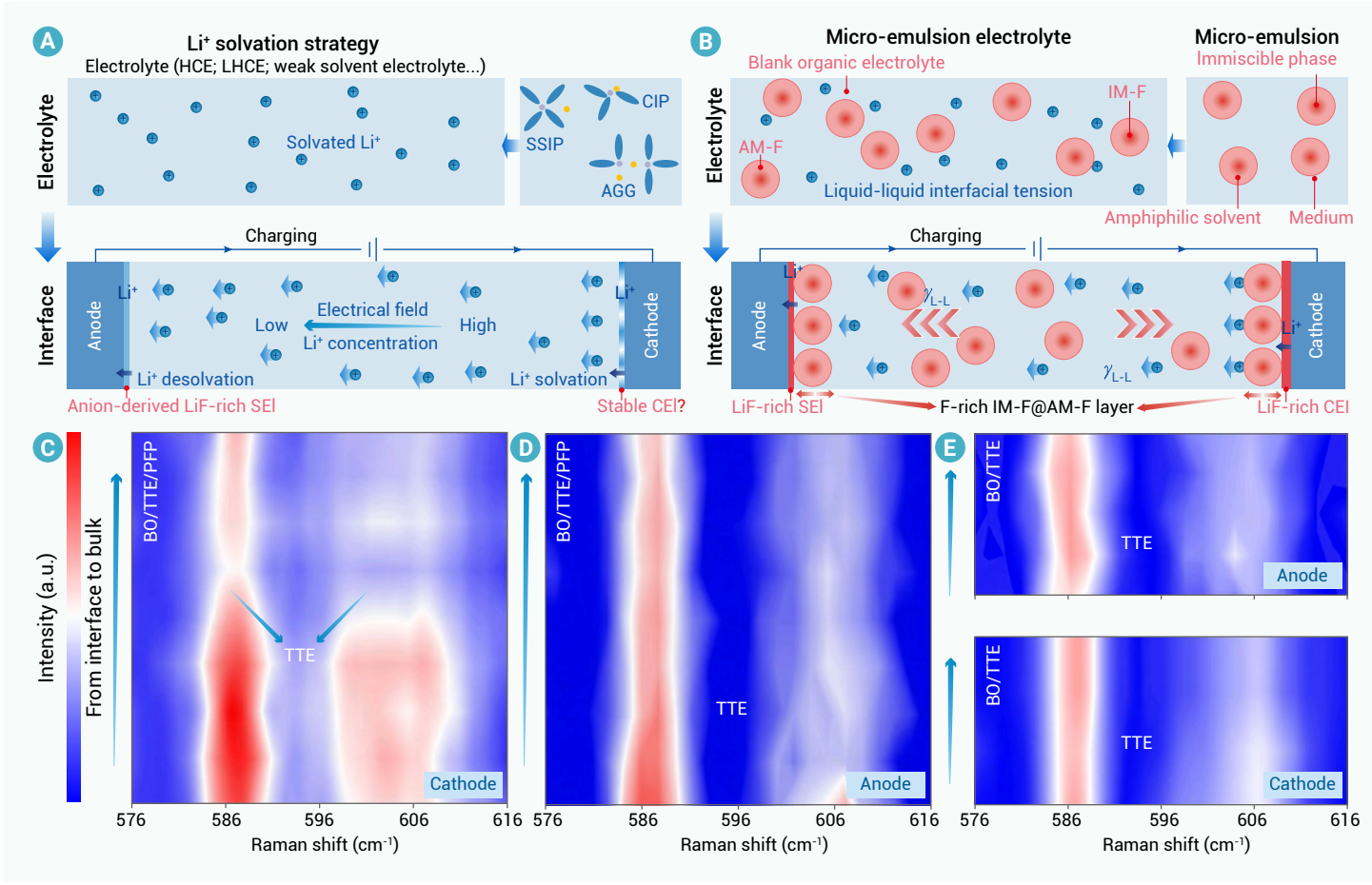

Figure 1. Micro-emulsion engineering approach. Schematic diagrams contrasting Li^+ ion-transport behavior under charging conditions with resultant interfacial transformations at electrodes (A) conventional solvation electrolyte and (B) the proposed micro-emulsion electrolyte. Raman spectra from the interface between the electrolyte and the NCM811 cathode (C) and Li-metal anode (D) to the bulk of the micro-emulsion electrolyte (colour scale: red, high intensity; blue, low intensity). (E) Raman spectra from the interface between the electrolyte and the cathode and the anode to the bulk of conventional solvation electrolyte. Reproduced with permission.[3] Copyright 2025, Springer Nature.

1,1,2,2-tetrafluoroethyl-2,2,3,3-tetrafluoropropyl ether (TTE) into the base electrolyte (BO) comprising 1 M lithium difluoro(oxalato) borate (LiDFOB)-0.2 M lithium tetrafluoroborate ($LiBF_4$) in diethyl carbonate (DEC)-fluoroethylene carbonate (FEC) (2:1, v/v). Owing to the differential solubility of PFP and TTE within the BO, ultrafine (50-120 nm) insoluble PFP@TTE emulsion droplets form in the electrolyte, whose addition does not alter the original solvation structure of the BO electrolyte. These PFP@TTE emulsion droplets exhibit substantially lower liquid-liquid interfacial tension than the base organic electrolyte (BO), enabling their spontaneous migration to both the cathode and anode interfaces under the tension-driven force.

Through cryo-electron microscopy (cryo-EM), the authors directly observed the morphology of ultrafine PFP@TTE emulsion droplets in the BO/TTE/PFP system. Complementary ultraviolet-visible spectroscopy semi-quantitatively confirmed the adsorption behavior of PFP@TTE at electrode surfaces. Building on these observations, Raman characterization revealed that within the BO/TTE/PFP micro-emulsion electrolyte, PFP@TTE emulsion droplets exhibited significant enrichment at both the cathode and anode interfaces (Figures 1C-D) compared to the conventional BO/TTE electrolyte (Figure 1E). During the charging/discharging process, the TTE signal intensity remained stable at the cathode/anode interface, distinct from the DEC solvent, which migrated with Li^+. This observation provides direct evidence that the droplet migration is governed by the interfacial tension-driven force ($\gamma_{L\text{-}L}$), rather than being coupled to the external electric field or Li^+ concentration gradients. Consequently, the PFP@TTE droplets continuously enrich and decompose at the interfaces, forming a LiF-rich protective layer that enables persistent protection of the electrodes.

To validate the universality of the micro-emulsion strategy, the authors systematically constructed eight micro-emulsion formulations based on five categories of IM-F solvents (perfluorohexanoyl fluoride, 1,1,1,2,2,4,5,5,5-nonafluoro-4-(trifluoromethyl)-3-pentanone, perfluorohexane, perfluorohexyl sulfonyl fluoride, pentadecafluorotriethylamine) and three categories of AM-F solvents (TTE, bis(2,2,2-trifluoroethyl), fluorobenzene). The Li||NCM811 coin cells employing these electrolytes all demonstrated superior cycling performance at 4.5 V high voltage compared to both the base electrolyte (BO) and the commercial electrolyte (1.0 M lithium hexafluorophosphate ($LiPF_6$) in ethylene carbonate (EC)-DEC (1:1, v/v) with 5 vol% FEC), confirming the universal stabilizing effect of the liquid-liquid interfacial tension-driven mechanism in LMBs. Furthermore, the authors proposed a dual criterion for solvent selection: the fluorophilic-organicphilic balance (FOB) number and the $\gamma_{L\text{-}L}$ value. The base organic solvent requires both a high FOB number and high $\gamma_{L\text{-}L}$, the IM-F solvent needs both a low FOB number and low $\gamma_{L\text{-}L}$, while the physicochemical properties of AM-F solvents lie between these two. This criterion achieves liquid-liquid phase separation through graded disparities, which provides a quantitative framework for selecting solvent components that extends the micro-emulsion principle beyond purely fluorinated systems.

Huang et al. pioneered the micro-emulsion strategy driven by liquid-liquid interfacial tension. By designing micelles formed from the fluorinated solvent (PFP@TTE) that migrate to and subsequently decompose at both electrode interfaces, this strategy reduces at the anode to generate a LiF-rich SEI layer and oxidizes at the cathode to construct a stable LiF-rich CEI layer, thereby synergistically enabling stable cycling of high-voltage LMBs. The innovation of this strategy lies in replacing the traditional electric field-dependent mechanism with the physical internal driving force $\gamma_{L\text{-}L}$. Without disrupting the bulk solvation structure of the base electrolyte, this enables the fluorinated components to overcome the limitations of concentration gradients and the electrochemical potential field.

Critically, the micro-emulsion strategy driven by $\gamma_{L\text{-}L}$ may superficially resemble the solvation-mediated micelle-like approach due to mesoscale self-assembled structures in both systems.[4] However, the two strategies are fundamentally distinct in mechanism. The micelle-like electrolyte achieves anion-rich solvation structures through molecular design, resulting in anion-derived LiF-rich interphases. In contrast, the microemulsion strategy exploits physical tension between components for droplet migration, with interphase formation decoupled from solvation processes. This provides a paradigm-shifting electrolyte design approach for developing high-energy-density batteries.

The development of micro-emulsion electrolytes validates the significance of mesostructure regulation. By modulating electrolyte microdomains' composition and properties, this strategy synergistically optimizes the evolution of passivation layers at both electrodes, achieving bipolar electrochemical stability unattainable in traditional homogeneous systems. This strategy has demonstrated significant potential in $Li||LiNi_{0.8}Co_{0.1}Mn_{0.1}$ cells, while its interface-tailored optimization for sulfide/organic cathodes requires further electrochemical investigation.

Elucidating the stabilization mechanism of the micro-emulsion strategy necessitates molecular-level insights into several critical questions: Starting from molecular property, it is necessary to quantitatively establish relationships between solvent molecular polarity and the assembly thermodynamics of IM@AM micelles; delving deeper into principles of micro-emulsion electrolyte, intensive investigation is required on the coupling effect between solvation entropy changes and interfacial tension in phase separation kinetics. Concurrently, more in-depth characterization methods, such as in situ neutron diffraction technology, need to be employed to study the migration and decomposition process of the IM@AM micelles.

For future industrialization and application, exploration in three key dimensions remains necessary: Foremost, solvent system innovation requires molecular engineering to develop low-cost IM/AM solvents with reduced fluorine content or even fluorine-free alternatives (as demonstrated by the initial feasibility of fluorobenzene for reduced fluorine),[5] while investigating the influence of heteroatoms like sulfur/nitrogen on micelle formation mechanisms. Additionally, refinement of screening criteria is needed; beyond the existing FOB number and $\gamma_{L\text{-}L}$ standards, methods such as machine learning can be combined to clearly define the effects of factors such as fluorine atom distribution and molecular configuration on this strategy.[5] Finally, the multi-scale coupling mechanism remains to be elucidated, particularly how the base electrolyte governs the interfacial tension-driven effect through liquid-liquid phase separation dynamics. Simultaneously, practical deployment of micro-emulsion electrolytes requires overcoming metastability-derived engineering constraints. This necessitates ensuring persistent storage homogeneity with thermo-mechanical resilience under operational extremes (e.g., -20°C to 60°C thermal cycling, transport-induced agitation), alongside exercising exacting micellar configuration control within scalable manufacturing regimes.

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AUTHOR CONTRIBUTIONS

All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.