COMMENTARY

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A thermo-responsive electrolyte paradigm for safer lithium metal batteries: From passive defense to active prevention

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Lithium metal batteries (LMBs) represent a frontier of next-generation energy storage technology owing to the ultrahigh theoretical specific capacity and low redox potential of lithium metal anodes [1]. However, safety concerns remain the most formidable obstacle to their practical deployment. Existing safety strategies, including flame-retardant additives [2,3] and thermally stable separators [4], are fundamentally passive in nature, activating only after the onset of failure. This post-failure intervention inherently limits its efficacy in preventing fatal thermal runaway. Addressing this long-standing challenge necessitates a paradigm shift from reactive mitigation to proactive, predictive safety mechanisms capable of intervening at the earliest stage of thermal instability.

Recently, Yang *et al*. [5] introduced an ultrafast thermo-responsive electrolyte composed of 2 M lithium bistrifluoromethanesulfonimidate (LiTFSI), 0.5 M $LiNO_3$ and 10 mM $LiPF_6$ in a mixed solvent of triethylene glycol divinyl ether (TEGDVE), 1,2-difluorobenzene (1,2-DFB) and triglyme in a volume ratio of 3:2:5 (denoted as TDT) that fundamentally redefines the role of electrolytes in LMBs, transforming them from inert ion-transport media to intelligent safety regulators (Figure 1a, b). This work represents the first successful translation of such smart electrolyte concepts into practical LMB systems without compromising electrochemical performance.

A defining characteristic of the TDT electrolyte is its sub-second thermal response kinetics. When heated to a temperature slightly below the melting point of the separator with a tunable trigger temperature between 100 and 150 °C, the electrolyte undergoes cationic polymerization initiated by $LiPF_6$ at 140 °C. This reaction forms a cross-linked network, driving transforms from a liquid conductor to an insulating and flame-retardant solid within 5 s (Figure 1c). The resulting solid phase is electrically insulating and flame-retardant, effectively terminating ionic transport pathways at the precise moment when separator softening begins. By instantaneously arresting ion conduction, the TDT electrolyte suppresses internal short-circuit formation and prevents the initiation of thermal runaway. In contrast to traditional electrolytes that accelerate failure once separator integrity is lost, the TDT system intervenes at the origin of instability, maintaining electrode separation and mitigating heat accumulation before escalation.

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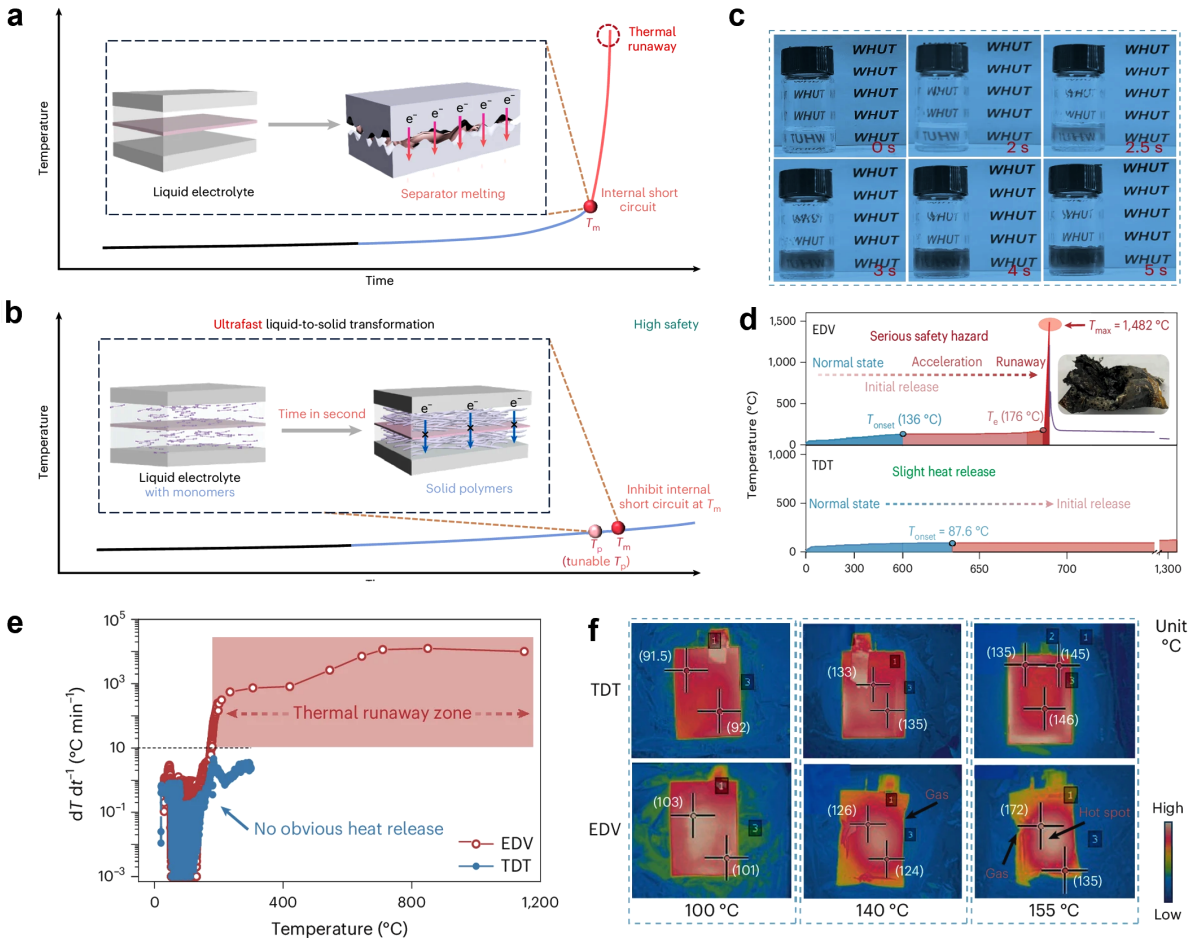


Figure 1 (a) Schematic illustration of the thermal runaway process in LMBs employing conventional carbonate electrolytes under thermal abuse conditions. (b) Conceptual design of an ultrafast thermo-responsive intelligent electrolyte in LMB that proactively prevents thermal runaway through rapid phase transformation. (c) The high-speed camera image capturing the state evolution of the TDT electrolyte during the heating process. (d) Temperature-time profile of a fully charged $LiFePO_4$||Li pouch cell in an adiabatic accelerating rate calorimetry test. (e) Self-heating rate as a function of temperature for pouch cells employing different electrolytes following the onset of self-heat generation. (f) Infrared thermal images of pouch cells with TDT and traditional carbonate electrolytes. Reproduced with permission from Ref. [5].

Direct visualization via high-speed camera imaging confirms the rapid phase transition characteristic of the TDT electrolyte. Comprehensive safety evaluations further validate its efficacy under realistic abuse scenarios. Accelerating rate calorimetry and infrared thermography measurements reveal that pouch cells using TDT electrolyte exhibit negligible heat release under thermal abuse, maintaining stable maximum temperatures without detectable hot spots. In contrast, cells employing conventional carbonate electrolytes undergo thermal runaway at 176 °C, reaching catastrophic peak temperatures of up to 1482 °C (Figure 1d). In addition, the self-heating rate (SHR) measurements indicate no origin inducing thermal runaway in the TDT system (Figure 1e). Under mechanical abuse conditions, TDT-based cells sustain voltages above 2.3 V even after electrolyte solidification, underscoring the structural and functional resilience afforded by this active protection mechanism (Figure 1f).

This work establishes a new safety paradigm for high-energy LMBs, shifting protection strategies from delayed and passive defense to ultrafast and autonomous intervention grounded in the fundamental physics of

thermal failure. Beyond mitigating safety concerns, the development of thermo-responsive intelligent electrolytes provides a broadly applicable design strategy for reconciling energy density with safety. These advances represent a critical step toward the reliable and practical deployment of high-energy LMB technologies.

Conflict of interest

The authors declare no conflict of interest.

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