COMMENTARY

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Thermal runaway prevented by instantaneous polymerization

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Thermal runaway of lithium metal battery arising from elevated temperature remains a major challenge for their application in energy storage systems [1,2]. Separator melting upon thermal abuse conditions is considered as a common cause of short circuits, which further leads to thermal runaway [2–4]. In a recent study published in *Nature Energy*, You and coworkers [5] demonstrate a thermal-responsive electrolyte being able to solidify within 5 s once the temperature reaches the melting point of the separator (T_m=140 °C). While various types of thermal-responsive electrolytes have been reported to address short circuits utilizing thermal-induced polymerization of electrolyte [6–8], few have demonstrated the potential to respond in such a fast manner (Figure 1a).

The as-designed electrolyte (TDT electrolyte) is prepared with a composition of lithium bis(trifluoromethane) sulfonimide, $LiNO_3$ and $LiPF_6$ (2:0.5:0.01, M/M) in a mixture of triethyleneglycol divinyl ether (TEGDVE), 1,2-difluorobenzene and triglyme (3:2:5, v/v). TDT electrolyte remains as a liquid below T_m, providing sufficient ionic conductivity and compatibility for the battery to operate at normal conditions. Ultrafast liquid-to-solid transition of electrolyte occurs once the temperature reaches T_m (Figure 1b), therefore preventing heat accumulation generated by short circuits. Instantaneous phase transition is enabled by polymerization with both fast initiation and chain growth. As the rate-determining step, rapid initiation is ensured by low activation energy, which is achieved by cationic polymerization using Lewis acid ($LiPF_6$) as initiator. High degree of functionality (DOF) and low steric hindrance (SH) of monomer (TEGDVE) synergistically promote fast chain growth, further contributing to the rapid polymerization kinetics (Figure 1c).

At temperatures below T_m, LFP||Li (LFP = $LiFePO_4$) cell with TDT electrolyte exhibits high capacity retention at both room (86.5%, 25 °C) and elevated temperature (93.9%, 55 °C) for 300 cycles (Figure 1d). When the temperature rises above T_m, TDT electrolyte solidifies within seconds, securing a normal open-circuit voltage for LFP||Li cell (Figure 1e). In comparison, LFP||Li cell with EDV ($LiPF_6$/ethylene carbonate/dimethyl carbonate/vinylene carbonate) electrolyte delivers much lower capacity retention at 55°C and experiences thermal short circuit at 160 °C. Pouch-cell level test further confirms the enhanced safety of lithium metal battery using TDT electrolyte under thermal abuse conditions (Figure 1f).

The research team shows that their electrolyte outperforms commercial electrolytes in terms of thermal

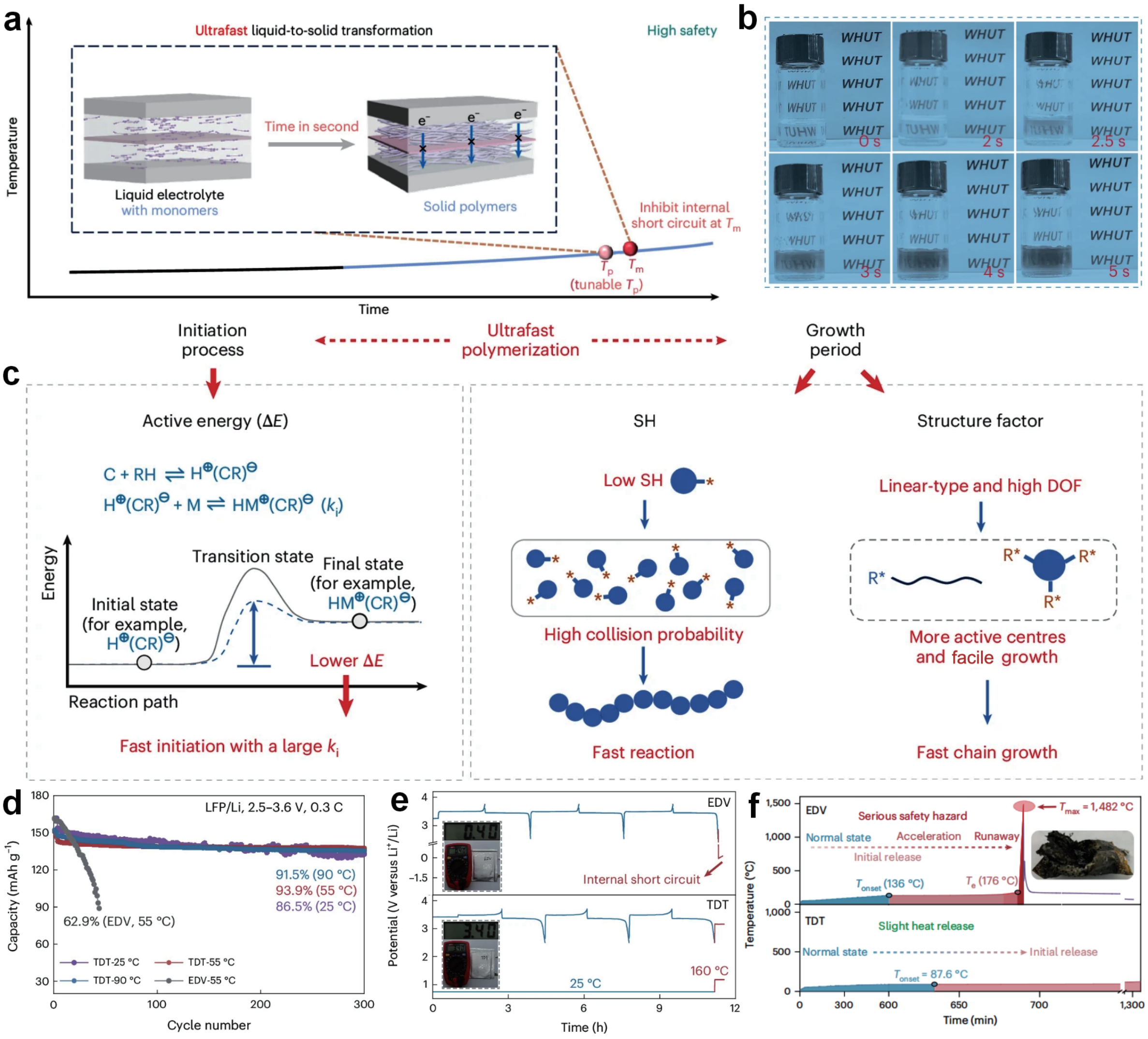

Figure 1 (a) Schematic illustration showing the design of an ultrafast thermal responsive electrolyte to prevent thermal runaway in lithium metal battery. (b) Images showing the solidification of TDT electrolyte by polymerization occurs within seconds at 140 °C. (c) Schematic illustration showing the determining factors of polymerization kinetics. (d) Cycling stability of LFP||Li cells with TDT and EDV electrolytes under different temperatures at 0.3 C. (e) Voltage-time curves of LFP||Li cells with TDT and EDV electrolytes at temperature below and above the melting point of separator. (f) Temperature-time curves of LFP||Li pouch cells with TDT and EDV electrolytes of accelerating rate calorimeter tests. TDT electrolyte shows no obvious heat release during the test, while the temperature of EDV electrolyte exceeds 1000 °C. Adapted with permission from Ref. [5].

runaway prevention. The findings provide valuable insights for improving battery safety through rational electrolyte design.

Conflict of interest

The authors declare no conflict of interest.

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