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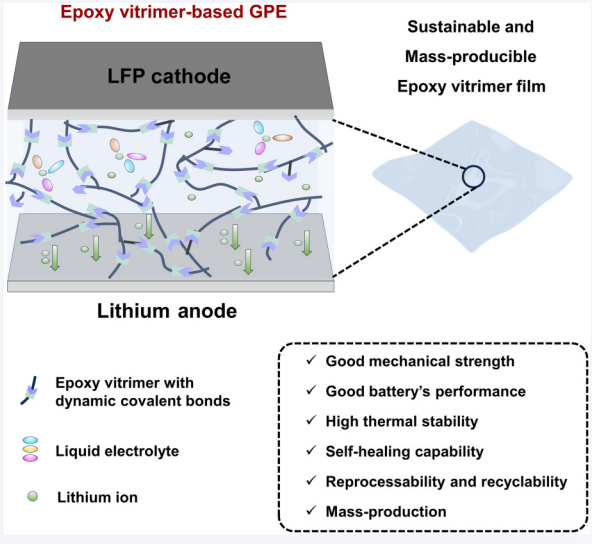
Sustainable and mass-producible epoxy vitrimer-based gel polymer electrolytes

Guojie Xu[1,2], Yuhang Tian[3], Yuting Wang[2], Mingrui Wu[2], Xinyu Zhao[2], Yen Wei[2], Jianlong Wang[1], Xiangming He[1], Eugene M. Terentjev[4], Pengfei Cao[3] ✉, and Yang Yang[1] ✉

[1] *Institute of Nuclear and New Energy Technology, Tsinghua University, Beijing 100084, China*
[2] *Key Laboratory of Bioorganic Phosphorus Chemistry & Chemical Biology of the Ministry of Education, Department of Chemistry, Tsinghua University, Beijing 100084, China*
[3] *School of Materials Science and Engineering, Beijing University of Chemical Technology, Beijing 100029, China*
[4] *Cavendish Laboratory, University of Cambridge, Cambridge CB3 OHE, UK*

Cite this article: *Nano Research*, **2026**, *19*, 94908453. https://doi.org/10.26599/NR.2026.94908453

ABSTRACT: With the growing demand for high-energy-density energy storage devices, lithium metal batteries (LMBs) using lithium metal as anodes have emerged as promising candidates. However, the issues of lithium anodes, such as dendrite growth, poor compatibility with liquid electrolytes, and safety concerns, pose challenges for their practical use. Therefore, gel polymer electrolytes (GPEs) become popular due to their good compatibility with lithium anodes. However, traditional GPEs suffer from poor mechanical strength, complex fabrication processes, lack of self-healing ability and recyclability, which hinder their large-scale and sustainable application. Here we present an epoxy vitrimer (cross-linked network with dynamic covalent bonds) as GPE for LMBs, synthesized from commercially available raw materials of epoxy resins by a simple technology of hot-pressing. The epoxy vitrimer-based GPE shows good electrochemical performance, with over 1000 h of stable cycling in Li symmetric cells and impressive cycling retention of 92.4% after 200 cycles in $LiFePO_4$||Li cells. Moreover, dynamic covalent bonds endow vitrimer with remarkable self-healing ability and recyclability, which would further improve the cycling and safety aspects of the battery. Therefore, due to its low material costs and simple manufacturing process, epoxy vitrimer-based GPE has a great potential for sustainable mass production in practical application in LMBs.



KEYWORDS: vitrimer, dynamic covalent bond, gel polymer electrolyte, epoxy, sustainable

1 Introduction

With the rapid development of society, the demand for high-energy-density energy storage devices has become increasingly urgent [1–7]. Due to the ultra-high theoretical specific capacity (3860 mAh·g^{-1}) and the lowest electrochemical potential (–3.04 V vs. standard hydrogen electrode) of lithium metal anodes, lithium metal batteries (LMBs), which use lithium metal as the anode, are considered the most promising device for achieving high energy density. However, during charge-discharge cycling, lithium dendrites are prone to form in LMBs. Excessive growth of lithium dendrites can penetrate the separator, causing internal short circuits and triggering a series of chain reactions that pose serious safety hazards [8–12]. Moreover, lithium metal anodes exhibit poor compatibility with liquid electrolytes, leading to the formation of numerous by-products and gas emission due to side reactions, which accelerate the degradation of capacity and even cause safety problems [13].

Gel polymer electrolytes (GPE) consist of liquid electrolytes embedded within a polymer network [14–20]. GPE is an intermediate state between liquid electrolytes and all-solid-state

Received: December 9, 2025; **Revised:** January 12, 2026
Accepted: January 15, 2026

✉Address correspondence to Yang Yang, yangyang86@tsinghua.edu.cn; Pengfei Cao, caopf@buct.edu.cn

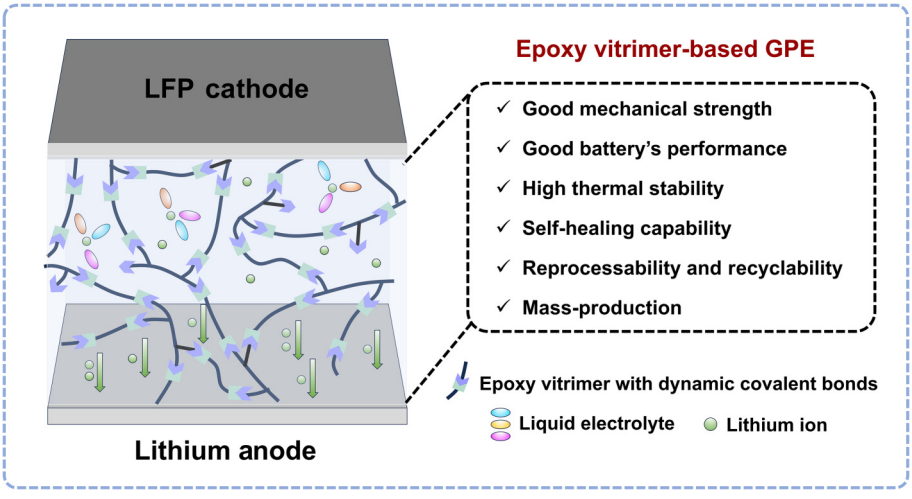
polymer electrolytes, so it combines the advantages of both, featuring high ionic conductivity, good electrolyte/electrode interfacial compatibility, and no risk of liquid leakage. Therefore, GPE has attracted much attention in exploration of high-specific-energy and high-safety LMBs [21–24]. However, the reported GPEs have some common drawbacks, which hinder their extensive practical applications. Firstly, most polymer matrices used in GPEs are based on linear thermoplastics, which have relatively poor mechanical strength and thermal stability. These properties, however, are critical for preventing short circuits caused by lithium dendrite penetration and thermal contraction. Till now, only a few of GPEs that based on thermosetting cross-linked networks have been developed, and they have demonstrated significantly improved mechanical and safety performance. Secondly, most of above thermosetting GPEs are constructed with irreversible covalent networks. These rigid networks lack the flexibility needed to accommodate the growth of lithium dendrites and the volume changes of electrodes during cycling, leading to interfacial instability between the electrode and electrolyte. As a result, the cycling performance of lithium metal batteries deteriorates gradually. Furthermore, due to the irreversible cross-linked network, these GPEs are generally non-reprocessable, non-recyclable, and non-degradable, posing challenges for their sustainable use. Thirdly, the commonly used methods for preparing GPEs—such as solution casting, phase inversion, *in situ* polymerization, UV curing, and electrospinning—often involve complex procedures and high manufacturing costs, making them unsuitable for large-scale production. Fourthly, most polymer matrices lack self-healing capability, which is essential for enhancing battery reliability and stability, and for extending the battery lifespan. Therefore, the development of GPEs that simultaneously exhibit high mechanical strength, recyclability, self-healing capability, and suitability for scalable manufacturing remains a critical goal in the field.

In this work, we developed a self-healable and recyclable GPE with not only high mechanical strength but also suitability for mass-production, based on the epoxy vitrimer concept [25]. Epoxy resin is one of the most common thermosetting polymers, and has been widely used in various industrial fields, due to its excellent mechanical strength, chemical and dimensional stability [26–32]. By introducing dynamic covalent bonds (e.g., ester bond or disulfide bond) into the cross-linked network of epoxy resin, it transforms into a "vitrimer": The covalently bonded network could be reprocessed, recycled, self-healed, and reprogrammed, as its topology could be changed by the stimulus-triggered exchange of its dynamic covalent bonds [33–39]. Therefore, here we use epoxy vitrimers to produce GPE, adopting the most common raw materials (such as diglycidyl ether of bisphenol A, carboxylic acids, etc.) and widely-applied technology of hot-pressing. Owing to its three-dimensional sparsely cross-linked network, the epoxy vitrimer has a good swelling in liquid electrolytes, forming a GPE. The epoxy vitrimer-based polymer electrolyte showed impressive electrochemical performance in both Li symmetric cells (cycling for over 1000 h) and in $LiFePO_4$ (LFP)||Li cells (maintaining a capacity retention of 92.4% after 200 cycles), outperforming traditional liquid electrolytes.

Furthermore, epoxy vitrimer-based GPE offers following significant advantages (Scheme 1). (1) Due to the thermosetting cross-linked network, epoxy vitrimer-based GPE exhibits high mechanical strength and excellent thermal stability, which effectively suppresses lithium dendrite growth and enhances both cycling life and battery safety. (2) The presence of dynamic covalent bonds enables intrinsic self-healing capability, further improving the reliability and longevity of the GPE. Additionally, the reprocessability and recyclability enabled by dynamic covalent bonds greatly facilitate the recycling of spent batteries, contributing to a more sustainable society. (3) Most importantly, the raw materials and fabrication processes for epoxy vitrimer-based GPE are nearly identical to those used in industrial epoxy resin production. The only difference is the addition of a small amount of base catalyst for transesterification. Given the widespread availability of raw materials, the simplicity of the process, and the low cost, this approach is highly scalable for the cost-effective manufacture of GPE. (4) In addition, the epoxy vitrimer-based GPE is compatible with multiple commercial liquid electrolytes, including ester-based and ether-based electrolytes. Therefore, epoxy vitrimer-based GPE holds great promise for large-scale manufacturing applications in lithium metal batteries.

2 Results

The epoxy vitrimer utilized in this study was widely available epoxy resin in the industrial, fabricated by the ring-opening reaction of epoxy groups from diglycidyl ether of bisphenol A (BADGE) and dodecanedioic acid (DC) with a molar ratio of 1:1, using 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD) as catalyst for the



Scheme 1 Schematic diagram of the advantages of epoxy vitrimer-based GPE.

transesterification reaction (Fig. 1(a)). It is worth noting that the fabrication process only involved two simple steps (Fig. 1(b)): (1) uniformly mixing DC and BADGE at 160 °C and adding TBD catalyst, then stirring the mixture to obtain a solid prepolymer; (2) then hot-pressing the solid prepolymer within a designed mold at 180 °C for 4 h under a pressure of 4 MPa and yielding an epoxy vitrimer film with desired thickness. Therefore, this simple fabrication process is quite conducive to mass-production.

The obtained epoxy vitrimer is a cross-linked network, which is confirmed by the results of Fourier transform infrared (FTIR) spectra and swelling tests. From the FTIR spectra shown in Fig. 1(c), the stretching vibration of epoxy group from BADGE located at 915 cm^{-1} disappeared, the peak of –OH at 3500 cm^{-1} in vitrimer generated after ring-opening reaction, and the vibration peak of –C=O at 1698 cm^{-1} from carboxyl group in DC transferred to 1730 cm^{-1} from ester group in epoxy vitrimer, unequivocally demonstrating the successful reaction between the epoxy and carboxyl groups. From the results of swelling tests shown in Fig. 1(d), the obtained epoxy vitrimer film swelled but did not dissolve in N-methylpyrrolidone (NMP), also suggesting a cross-linked network.

In addition, the epoxy vitrimer exhibits excellent mechanical property and thermal stability. As shown in the stress–strain curve in Fig. 1(e), the epoxy vitrimer has a fracture elongation rate exceeding 400%, and withstands a stress of 13.5 MPa. And from Fig. 1(f), it is evident that the epoxy vitrimer has good thermal stability, with almost no weight loss even at 300 °C. Compared to Celgard 2325, the epoxy vitrimer has much higher thermal stability. This can be seen from Fig. S1 in the Electronic Supplementary Material (ESM), Celgard 2325 curls up above 80 °C, while the epoxy vitrimer remains unchanged at 140 °C even for 30 min. Both high mechanical strength and high thermal stability are of great significance for the application of epoxy vitrimer as GPE in LMBs, which will highly contribute to the safety of the battery.

In addition, this epoxy vitrimer has excellent wettability and swelling capacity for liquid electrolyte, further showing potential application as GPE. Firstly, the epoxy vitrimer showed good electrolyte affinity. It could be seen that the contact angle of the liquid electrolyte (1.0 M $LiPF_6$ in a mixture of ethylene carbonate (EC)/dimethyl carbonate (DMC)/ethyl methyl carbonate (EMC) (1:1:1, weight ratio) with 1 wt.% vinylene carbonate (VC) as an additive) on the vitrimer interface is small, with the value of 58.9° (see Fig. S2 in the ESM). Moreover, when immersing epoxy vitrimer in liquid electrolyte, the liquid electrolyte could be effectively filled into the cross-linked network, resulting in a GPE. After being immersed in liquid electrolyte for 12 h, the uptake capacity of epoxy vitrimer has reached saturation, and the value was 178% (Fig. 2(a)). At this level of electrolyte swelling, we considered

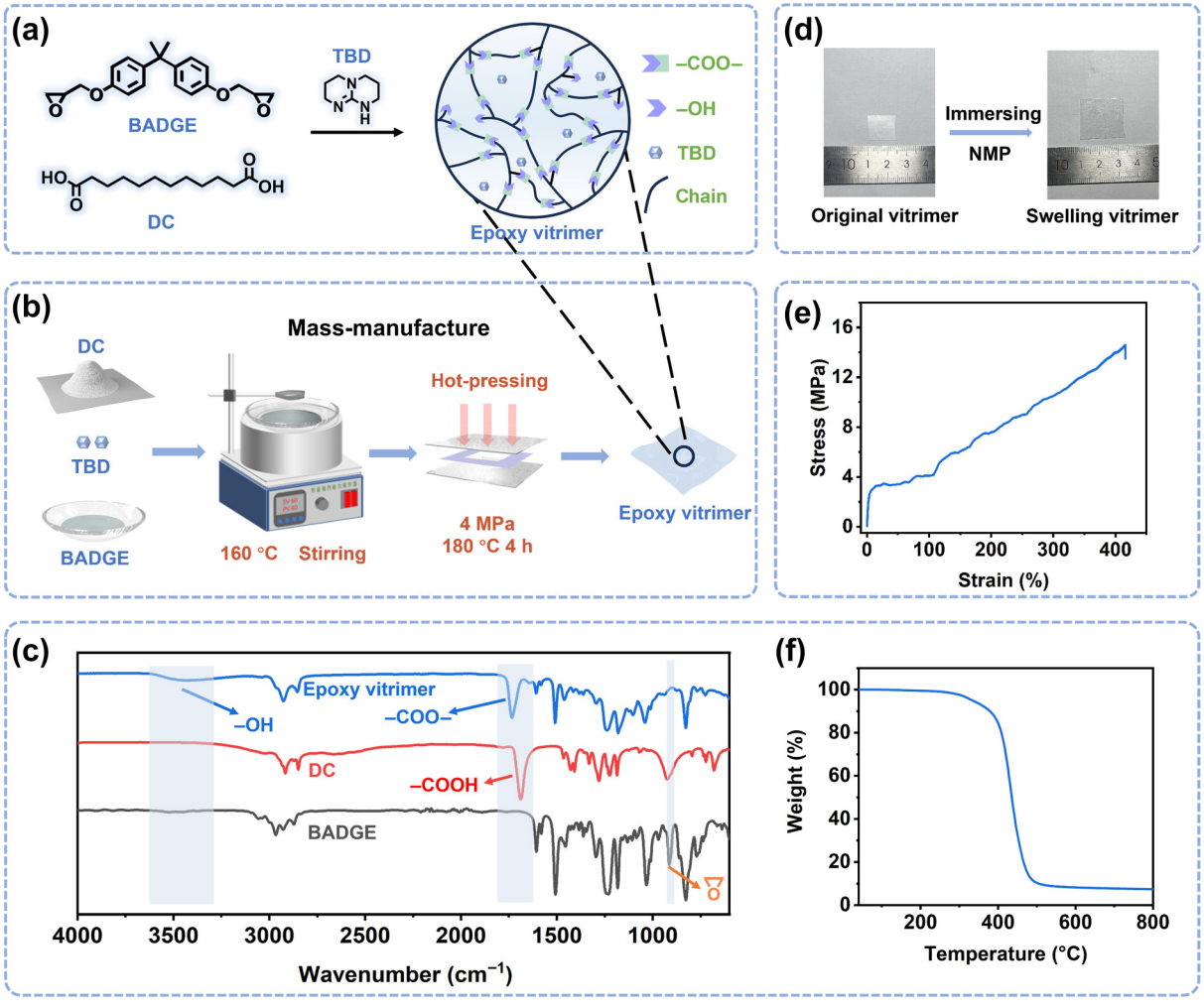


Figure 1 (a) Synthesis of epoxy vitrimer. (b) Mass-manufacture diagram of epoxy vitrimer. (c) FTIR spectra of epoxy vitrimer (blue), DC (red) and BADGE (black). (d) Swelling test of epoxy vitrimer in NMP. (e) Stress–strain curve of epoxy vitrimer. (f) Thermogravimetric curve of epoxy vitrimer.

the epoxy vitrimer-based GPE ready for the subsequent electrochemical performance tests and battery cycling.

Further, we conducted a series of tests to gain deeper insights into the electrochemical performances of epoxy vitrimer-based GPE. In this study, all assembled cells were rested for 12 h prior to testing. First, the electrochemical stability window was evaluated by linear sweep voltammetry (LSV) curves using Li||stainless steel (SS) cells, revealing that the epoxy vitrimer-based GPE remained stable up to 4.1 V without undergoing redox reactions (Fig. 2(b)). So, the epoxy vitrimer-based GPE was suitable for LMBs using LFP as cathode. Second, electrochemical impedance spectroscopy (EIS) measurements in SS||SS cells demonstrated a good ionic conductivity of this epoxy vitrimer-based GPE, helping to the smooth migration of Li^+ in batteries (Fig. 2(c)). Third, the impact of resting time on cell impedance was investigated in LFP||Li cells, showing a gradual decrease in impedance over time (Fig. 2(d)), which exactly matched the results of uptake amount of liquid electrolyte shown in Fig. 2(a). Therefore, thorough immersion helped improve the interfacial contact between the epoxy vitrimer-based GPE and the electrodes, thereby promoting more stable battery cycling. In addition, the epoxy vitrimer had a relatively low glass transition temperature (T_g) of 22.7 °C (Fig. 2(e)), demonstrating high flexibility and chain-segment mobility at room temperature, which facilitated the migration of lithium ions within the polymer segments [19, 40, 41]. Furthermore, we examined the cycling stability of Li||Li symmetric cell by using epoxy vitrimer-based GPE (Fig. 2(f)), where the results indicated that the cell incorporating epoxy vitrimer-based GPE maintained stable for over 300 h at the current density of 0.1 mA·cm^{-2} with a capacity of 0.1 mAh·cm^{-2} (Fig. 2(g)). Epoxy vitrimer-based GPE was found compatible with commercial ether-based electrolytes. The epoxy vitrimer-based GPE that swelled to 1.0 M LiTFSI in a mixture of DME/DOL (1:1 volume ratio) demonstrated a good ionic conductivity, and the Li||Li symmetric cell could cycle for over 300 h with high stability (Figs. S3 and S4 in the ESM). All these results indicated the great potential of epoxy vitrimer as a GPE.

To verify the feasibility of epoxy vitrimer-based GPE in LMBs, we assembled LFP||Li cells to assess the electrochemical

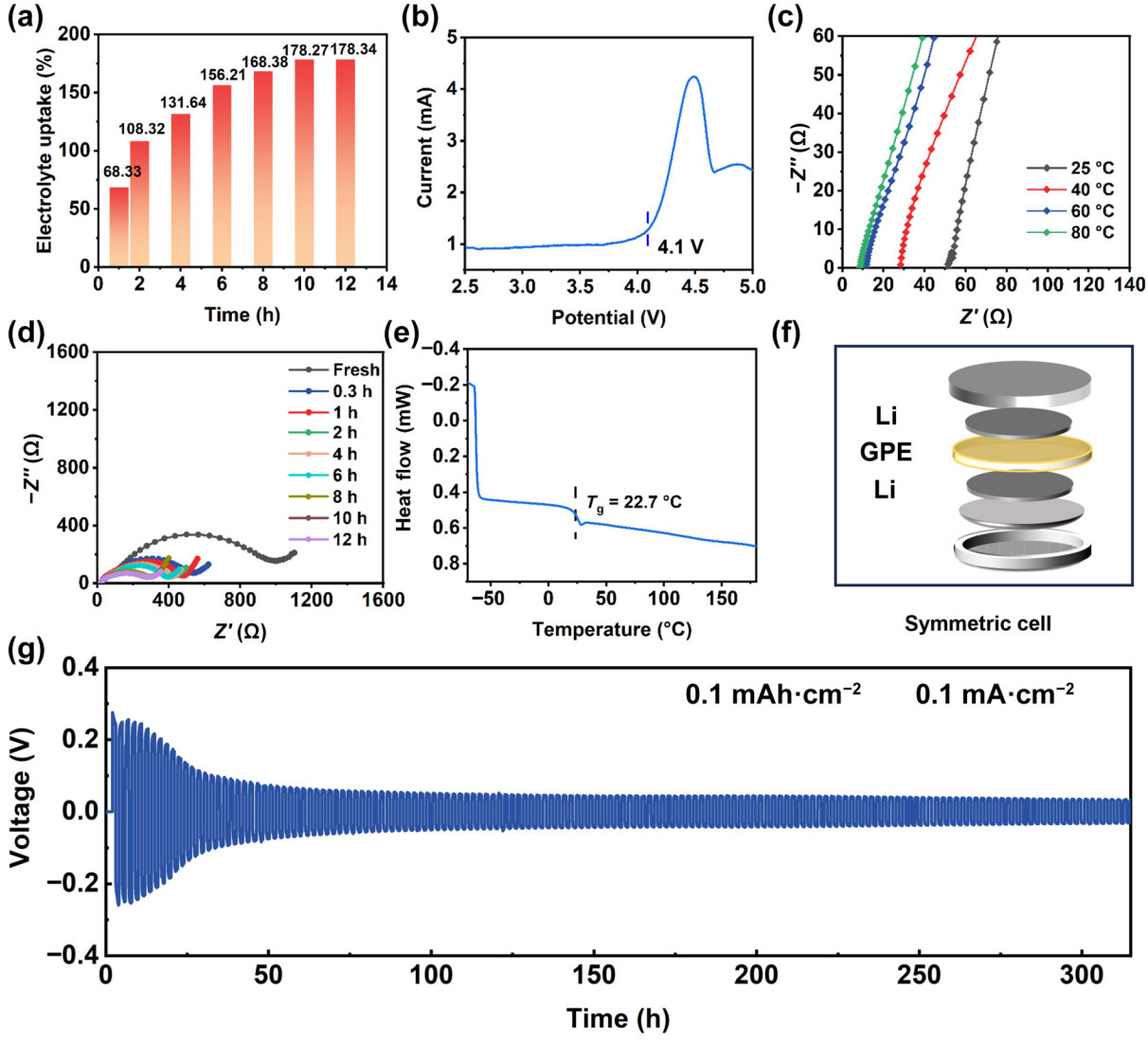


Figure 2 (a) Electrolyte uptake of epoxy vitrimer in 1.0 M $LiPF_6$ in a mixture of EC/DMC/EMC (1:1:1, weight ratio) with 1 wt.% VC. (b) LSV curve of epoxy vitrimer-based GPE in Li||SS cells. (c) EIS curves of epoxy vitrimer-based GPE in SS||SS cells under different temperature. (d) EIS curves of epoxy vitrimer-based GPE in LFP||Li cells under room temperature. (e) Differential scanning calorimetry (DSC) traces of epoxy vitrimer. (f) Schematic diagram of Li||Li cell. (g) Voltage profiles of symmetric Li||Li cell using epoxy vitrimer-based GPE at the current density of 0.1 mA·cm^{-2} with a capacity of 0.1 mAh·cm^{-2}.

performance. As shown in Fig. 3(a), the LFP||Li cell using epoxy vitrimer-based GPE exhibited excellent cycling stability at 25 °C under 0.5 C, maintaining 92.4% of its initial capacity after over 200 cycles. In contrast, the cell with liquid electrolyte and commercial separator (Celgard 2325) showed significant capacity degradation after 150 cycles. Figure 3(b) demonstrated that as the charge-discharge cycles progressed, the LFP||Li cell with epoxy vitrimer-based GPE maintained a stable voltage platform. Moreover, epoxy vitrimer-based GPE also exhibited commendable rate performance. As shown in Fig. 3(c), the cell delivered an initial capacity of 152.8 mAh·g^{-1} at 0.1 C. As the rate increased, both capacity and discharge voltage platform decreased (Figs. 3(c) and 3(d)), and the capacity decreased to 123.1 mAh·g^{-1} at 1 C. While the capacity had almost returned to its initial value (152.2 mAh·g^{-1}) when the current was reduced back to 0.1 C. These results indicated that the rate performance of epoxy vitrimer-based GPE was better than that of liquid electrolytes. Therefore, cells using epoxy vitrimer-based GPE exhibited excellent cycling stability and rate performance in LMBs, demonstrating great promising potential for practical applications.

In addition, epoxy vitrimer exhibits exceptional self-healing ability, reprocessability and recyclability compared to traditional epoxy resins, due to the rearrangement of the topological network caused by the transesterification of dynamic ester bonds (Fig. 4(a)). First, epoxy vitrimer demonstrated remarkable thermal healing properties (Fig. 4(b)), with damaged cuts visibly restoring after being maintained at 180 °C for 2 h, resulting in only a slight reduction in mechanical performance (Fig. 4(f)). Moreover, epoxy vitrimer could be self-healed by liquid electrolyte. As shown in Fig. 4(c), after immersing epoxy vitrimer in the commercial liquid electrolyte for one hour, surface scratches on the epoxy vitrimer underwent significant healing. This was quite useful and important, as the epoxy vitrimer-based GPE could be self-healed when damaged during charge/discharge processes, or accommodate volume changes at the electrode interface during charge/discharge processes, which was conducive to improve the cycling and safety performance of battery. Second, it also possessed excellent recyclability. On the one hand, the fragments of epoxy vitrimer could be reprocessed into a continuous film through hot-pressing (Fig. 4(d)), with minimal loss in mechanical strength compared to the original material (Fig. 4(g)), endowing with the ability to be reused multiple times. The reprocessed films exhibit a similar thickness comparable to that of the original samples. On the other hand, epoxy vitrimer could be efficiently recycled and degraded by immersion and agitation in ethylene glycol at 50 °C (Fig. 4(e)). After removing the solvent, the compounds could be re-polymerized and reused, highlighting its potential for sustainable applications.

3 Conclusion

In summary, we have developed an epoxy vitrimer as gel polymer electrolyte for lithium metal batteries. This vitrimer is synthesized

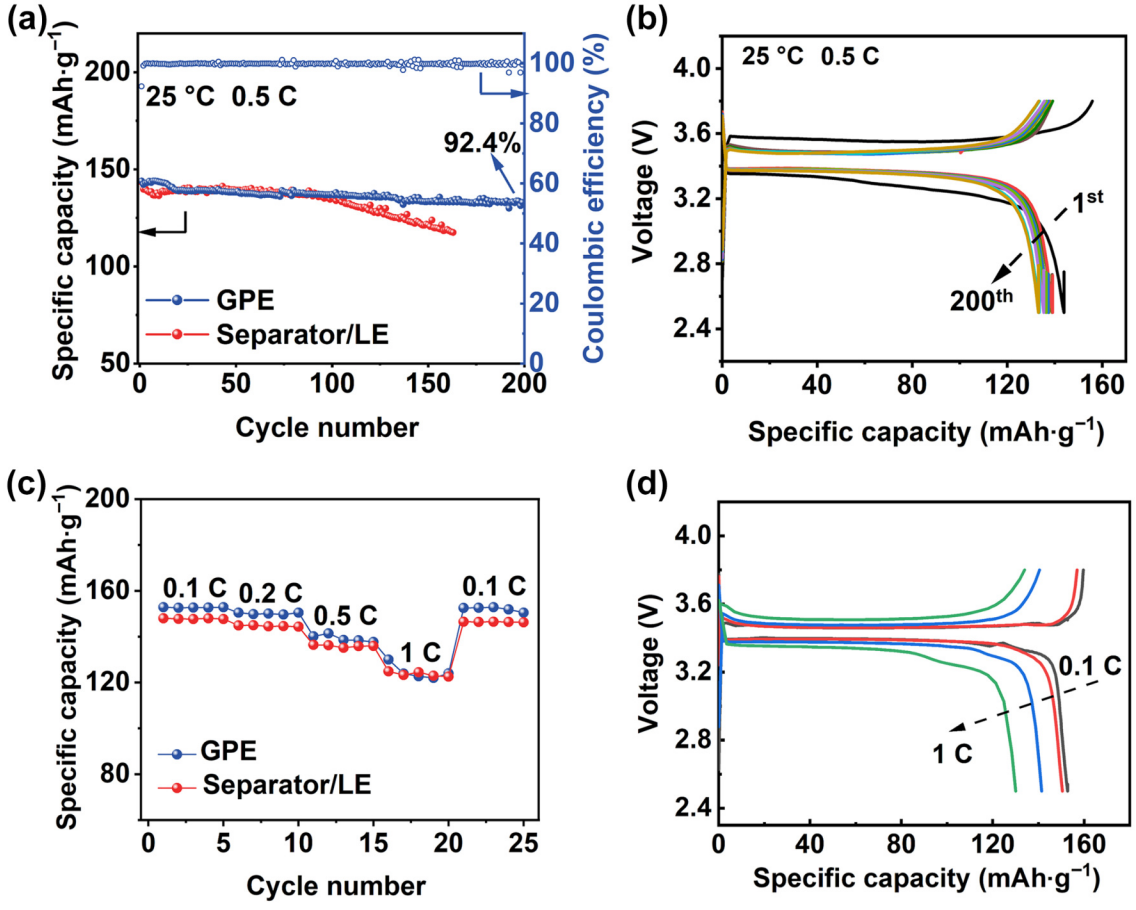


Figure 3 (a) The cycling performance of LFP||Li cells using epoxy vitrimer-based GPE or separator/liquid electrolyte (LE) at 0.5 C under 25 °C. (b) The discharge/charge curves of a LFP||Li cell using epoxy vitrimer-based GPE at 0.5 C under 25 °C. (c) Rate performance of LFP||Li with epoxy vitrimer-based GPE or separator/LE. (d) Capacity–voltage curves of LFP||Li with epoxy vitrimer-based GPE at different rates.

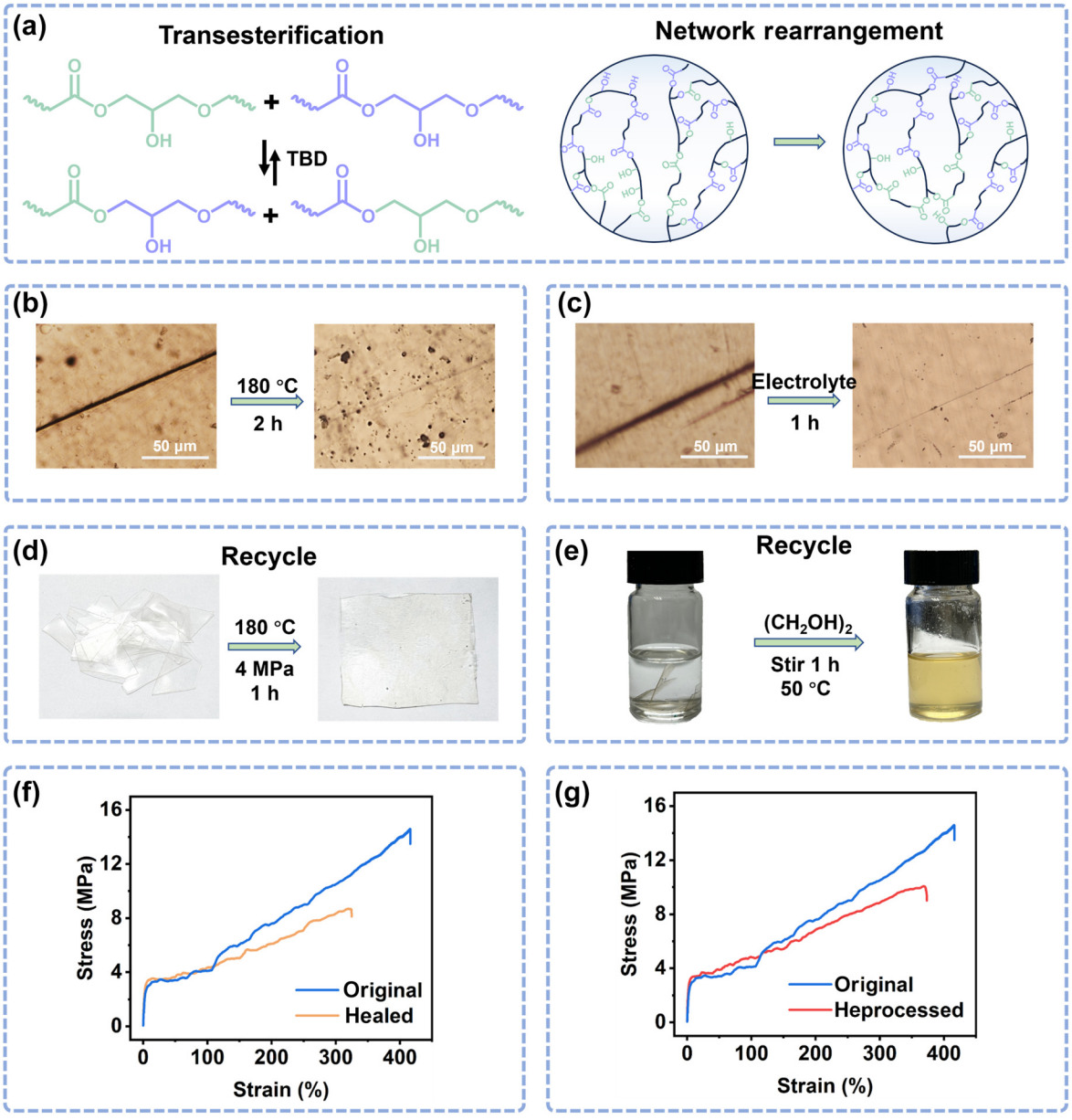


Figure 4 (a) Schematic diagram of transesterification and network rearrangement of epoxy vitrimer. (b) Self-healing ability of epoxy vitrimer in commercial liquid electrolyte (1.0 M $LiPF_6$ in a mixture of EC/DMC/EMC (1:1:1, weight ratio) with 1 wt.% VC). (c) Self-healing ability of epoxy vitrimer under heat. (d) Recyclability of epoxy vitrimer by hot-pressing. (e) Recyclability of epoxy vitrimer by degradation. (f) Stress–strain curves of original epoxy vitrimer and epoxy vitrimer after healing. (g) Stress–strain curves of original epoxy vitrimer and epoxy vitrimer after reprocessing.

using commercially available low-cost materials and simple preparation technology of hot-pressing. This makes epoxy vitrimer suitable for large-scale production. Notably, epoxy vitrimer-based GPE also shows excellent electrochemical performance, exhibiting good cycling stability and rate performance in lithium metal batteries. Additionally, epoxy vitrimer exhibits excellent mechanical property and thermal stability, as well as remarkable self-healing ability, reprocessability and recyclability, which may greatly improve the cycling and safety performance of LMBs. Based on these advantages, epoxy vitrimer-based polymer electrolyte holds promising potential for large-scale commercial applications in lithium metal batteries.

Electronic Supplementary Material: Supplementary material (materials, methods, characterizations, and additonal images and figures) is available in the online version of this article at https://doi.org/10.26599/NR.2026.94908453.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

Acknowledgements

This work was supported the National Natural Science Foundation of China (No. 22175106), the Beijing Nova Program (No.

20220484126), Tsinghua University Initiative Scientific Research Program, Young Elite Scientists Sponsorship Program by CAST (No. YESS20230470), and Tsinghua–Cambridge Joint Research Initiative Fund.

Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

G. J. X.: Data curation, project administration, validation, writing manuscript, experimental design. Y. H. T.: Data curation. Y. T. W.: Writing manuscript. M. R. W.: Writing manuscript. X. Y. Z.: Writing manuscript. Y. W.: Project administration. J. L. W.: Project administration. X. M. H.: Funding acquisition. E. M. T.: Writing manuscript, funding acquisition. P. F. C.: Project administration. Y. Y.: Project administration, funding acquisition, writing manuscript, experimental design. All the authors have approved the final manuscript.

Use of AI statement

None.

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