超越500 Wh/kg的电池专栏

聚环氧乙烷基电解质结构设计与离子传输机制研究进展

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摘 要：针对聚环氧乙烷基固态电解质离子电导率、机械强度与界面稳定性难以协同优化的瓶颈，本文从聚合物链段运动、离子解离-传输和界面化学的多尺度调控出发，系统评述了近年来的突破策略。在聚合物本体结构设计中，重点分析了深共晶溶剂竞争配位、离子液体限域交联网络、硫醇-烯半互穿网络耦合双盐原位富氟化锂界面，以及锂化有机纳米纤维增强等四种路径的离子传导与力学协同强化机制。在复合电解质层面，深入讨论了金属有机框架开放金属位点强配位固定阴离子、二维纳米片锚定阴离子并抑制聚环氧乙烷结晶，以及Janus界面层调控聚合物与功能填料化学相容性等策略，凝练出阴离子锚定、界面钝化与体相连续导锂的协同设计思路。针对硅碳负极界面，阐明了预浸润构建连续聚合物离子传导网络对活性材料利用率和循环稳定性的提升机理。在此基础上，提出了链段松弛、离子对解离、界面输运多级耦合调控的新观点，并展望了面向高比能全固态锂金属电池的超薄化、梯度化复合电解质结构及其连续制造技术。本综述为聚合物固态电解质的理性设计与产业化推进提供了理论指引和创新思路。

关键词：聚环氧乙烷；固态电解质；结构设计；离子传输机制；全固态锂金属电池
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Progress in structural design and ion transport mechanisms of PEO-based electrolytes

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Abstract: To address the bottleneck of synergistically optimizing ionic conductivity, mechanical strength, and interfacial stability in poly(ethylene oxide) -based solid-state electrolytes, this review systematically summarizes recent breakthroughs from the perspective of multiscale regulation of polymer segmental motion, ion dissociation-transport, and interfacial chemistry. In terms of polymer bulk structure design, we highlight four pathways including competitive coordination by deep eutectic solvents, ionic liquid confinement within

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cross-linked networks, thiol-ene semi-interpenetrating networks coupled with dual-salt *in-situ* formation of a LiF-rich interphase, and lithiated organic nanofiber-reinforced networks and analyze their synergistic mechanisms for enhancing ionic conduction and mechanical properties. At the composite electrolyte level, we discuss in depth the strategies of strongly immobilizing $TFSI^-$ through open metal sites in metal-organic frameworks, anchoring anions and suppressing crystallization using two-dimensional nanosheets, and regulating the chemical compatibility between polymer and fillers via Janus layers, thereby distilling a synergistic design concept of anion anchoring, interfacial passivation, and bulk continuous lithium-ion conduction. For the silicon-carbon anode interface, we elucidate the mechanism by which pre-infiltration constructs a continuous polymer ion-conducting network to improve active material utilization and cycling stability. On this basis, we propose a new perspective of multilevel coupled regulation involving segmental relaxation, ion pair dissociation, and interfacial transport, and envision ultrathin and gradient composite electrolyte architectures along with their continuous manufacturing technologies for high-energy-density all-solid-state lithium metal batteries. This review offers theoretical guidance and innovative ideas for the rational design and industrialization of polymer solid-state electrolytes.

Keywords: poly(ethylene oxide); solid-state electrolyte; structural design; ion transport mechanism; all-solid-state lithium-metal batteries

随着电动汽车与储能系统的快速发展，对二次电池的能量密度和安全性提出了更高要求。当前锂离子电池受石墨负极容量限制（372 mAh/g），难以满足长续航（350~500 Wh/kg）需求[1]。锂金属负极以超高理论比容量（3860 mAh/g）和低电位成为破局关键，但与液态电解液的热力学不稳定性易引发枝晶生长、界面失效及热失控，严重制约实用化[2-4]。

全固态电池以固态电解质替代液态电解液，从根本上消除易燃隐患，并凭借宽电化学窗口和本征阻枝晶潜力，成为高比能电池的理想途径[5-6]。在各类固态电解质中，聚合物基电解质因柔韧性好、易加工、界面兼容性佳，被认为最具实用前景[7]，但其长期受制于“离子电导率-机械强度-界面稳定性”难以协同提升的共性难题。其室温离子电导率通常仅为 10^{-5}~10^{-4} S/cm，远低于无机电解质，根源在于链段运动受限和离子-配位强相互作用[8-12]。提高非晶相比例虽可改善电导率，却以牺牲机械强度为代价，而足够的模量是抑制锂枝晶穿刺的必要条件[13-16]。同时，聚合物在正极侧因高压氧化和负极侧锂金属副反应导致界面动态接触失效，进一步恶化循环性能[17-20]。

在众多聚合物电解质中，聚氧化乙烯（PEO）基体系研究历史最长、认识最深入[21-23]。PEO 凭借链段柔顺、对锂盐溶解性好、成本低、易成膜等特点，成为核心研究对象，其离子传输遵循“链段运动协同模型”：锂离子与醚氧原子配位，随链段热运动迁移[24]。该体系集中体现了聚合物电解质的普遍瓶颈，如室温高结晶度导致电导率低、力学与离子传输相互制约、氧化稳定性不足等[25-26]。因此，PEO 基电解质的研究对突破整个领域的实用化障碍具有示范意义。近年来，通过动态键、多位点交联及共价功能化等策略，在协同提升 PEO 基电解质的离子电导率、机械强度与界面稳定性方面已取得显著进展[27-29]。本文立足作者团队研究积累，聚焦 PEO 基电解质核心瓶颈与突破路径，系统梳理结构设计与离子传输机制，凝练出“链段松弛-离子对解离-界面输运”多级耦合调控的新观点，旨在为高比能全固态锂金属电池的理性设计提供理论参考。

1　聚环氧乙烷基电解质结构设计与离子传输

面对室温链段运动受限引起的结晶度高、离子电导率与机械强度相互制约及氧化稳定性不足等瓶颈，研究者已从聚合物链结构层面提出了共聚[30-31]、

交联网络[32-33]、超分子滑动交联[34-35]、单离子导体[36-37]、添加剂[38-41]以及多种工艺[42-46]等调控策略，并开始从焓熵协同的热力学视角寻求突破[47-48]。上述进展推动PEO基电解质从单一参数优化走向多功能协同，但仍缺乏对“链段松弛-离子对解离-界面输运”多级耦合过程的系统性理解。基于此，本团队围绕聚合物本体的结构设计与工艺调控，从分子链配位环境、网络拓扑构筑到界面工艺优化，形成了系统研究体系，凝练出多级耦合调控的新思路。

1.1 深共晶溶剂增塑：竞争配位驱动离子对解离与链段活化

针对Li^+与醚氧强配位导致的离子对聚集、链段运动受限及低锂离子迁移数等核心瓶颈[49-50]，本团队首次报道了基于深共晶溶剂（DES）的原位增塑策略[图1(a_1)][51]。该DES由LiTFSI与*N*,*N*-二甲基甲磺酰胺（DMMSA）构成，其分子设计的关键在于磺酰胺基团提供竞争性配位位点，有效弱化醚氧与Li^+的相互作用[图1(a_2)]，从根源上解耦离子传输的动力学限制。

差示扫描量热（DSC）分析[图1(b)]显示，DES使PEO结晶度由40.5%骤降至25.7%，非晶区大幅增加。60℃下PEO-DES-3的离子电导率达2.1×10^{-4} S/cm，较PEO-LiTFSI体系提升约160%[图1(c)]。MD模拟进一步揭示微观机制：RDF[图1(d)]和配位数统计[图1(e)]表明，DMMSA的竞争配位使Li^+-$TFSI^-$平均配位数趋近于零，即高效解离离子对，释放自由Li^+。这直接驱动锂离子迁移数（t_{Li^+}）从0.28显著升至0.33，且为离子液体增塑体系的2.36倍[图1(f)]。因此，DES设计通过同时增强链段运动能力和促进离子对解离，实现了从链段松弛和离子对解离两个层级的协同调控。Li/Li对称电池测试[图1(g)]中，PEO-DES体系在0.2 mA/cm²下稳定循环超200 h，极化电压平稳，而PEO-LiTFSI仅约50 h即短路。$LiFePO_4$/Li全电池在60℃、0.2 C下首圈放电比容量达168 mAh/g，优于对比体系[图1(h)]。该工作通过竞争配位策略同时提升了电导率、迁移数与界面相容性，为本领域提供了离子-链段配位弱化的全新设计范式，深刻体现了离子对解离对体相输运的调控作用。

1.2 离子液体封装交联网络：双重传导路径与原位界面锚定

从聚合物拓扑结构入手，同样是打破电导率与机械强度制约的有效途径[52-53]。本团队设计了一种离子液体（$Pyr_{14}TFSI$）封装的交联聚合物电解质（C-SPE-IL），通过原位热聚合将离子液体均匀限域于三维交联PEO网络[图2(a)]，形成聚合物相与离子液体相纳米级互穿的复合结构[54]。

原子力显微镜（AFM）与扫描电镜（SEM）证实离子液体在基体中纳米级均匀分散，无相分离[图2(b)，(c)]。PALS[图2(d)]和DFT计算[图2(e)]表明，Pyr^+_{14}与聚合物的结合能（1.36 eV）高于Li^+（0.99 eV），优先占据醚氧位点，迫使Li^+从链段解离并进入离子液体相。^{7}Li NMR[图2(f)]出现双峰，明确证实存在聚合物链跳跃和离子液体相自由扩散两条独立传导路径。MSD[图2(h)]定量显示，离子液体相中Li^+扩散系数极高，交联聚合物的链段运动亦优于线性体系，双重通路使C-SPE-IL在30℃离子电导率达3.98×10^{-4} S/cm。该策略巧妙地通过交联网络提供力学支撑，离子液体相承担快速传质，成功解耦了机械强度与离子电导的竞争。同时，原位聚合工艺实现了电解质与电极的共形接触，对称电池极化电压仅57 mV，稳定循环超200 h[图2(g)]；XPS[图2(i)]证实诱导形成了富含LiF和Li_3N的界面层，有效抑制枝晶。固态$LiFePO_4$电池在60℃、0.5 C下循环150圈容量保持率95.6%[图2(j)]。该工作提出了构筑高速离子通道的同时原位锚定稳定界面，印证了界面输运调控对长循环稳定性的关键作用。

1.3 点击化学构建半互穿网络：短链增塑与双盐界面化学协同

针对线性PEO结晶度高、链段运动与力学强度难以兼顾的问题[55-56]，本团队利用硫醇-烯点击化学制备了高度均匀的半互穿聚合物网络电解质[57]。以三羟甲基丙烷三（3-巯基丙酸酯）与聚乙二醇二丙烯酸酯的点击反应构建三维交联骨架，线性聚乙二醇二甲醚（PEGDME）短链均匀嵌入，形成连续的纳米增塑域[图3(a)]。

PEGDME使T_g显著下降，自由体积增大，SIPE-1-1在30℃离子电导率达1.78×10^{-4} S/cm，t_{Li^+}高达0.53[图3(b)]。DFT计算表明，PEGDME对Li^+的吸收能（−6.02 eV）弱于C-PEO（−6.89 eV），更易释放自由Li^+，且MSD[图3(d)]证实其扩散更快。这揭示出短链增塑剂不仅活化了链段松弛，更作为高效的离子对解离与传输介质，协同提升了传

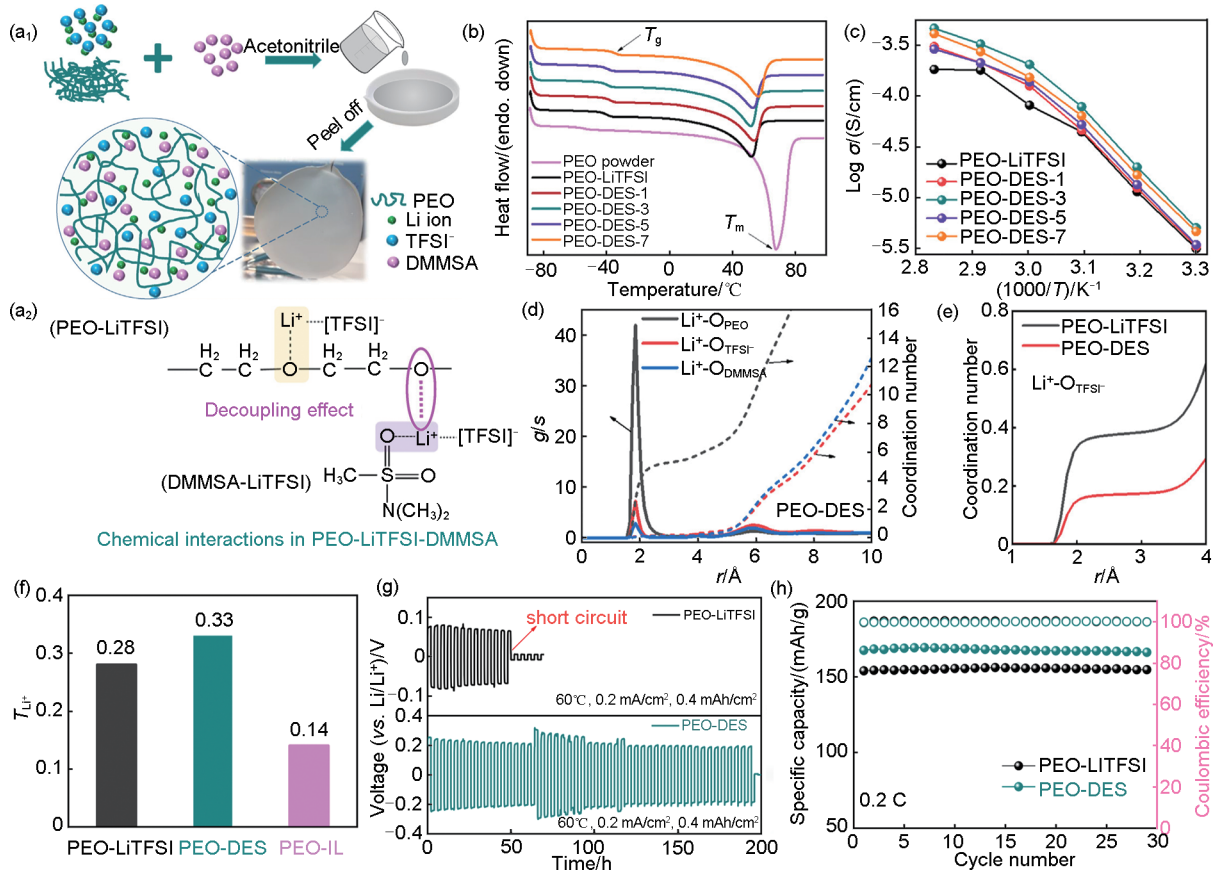

图1　(a_1) PEO-DES电解质膜制备示意图；(a_2) DMMSA竞争配位弱化EO-Li^+作用的化学机制；(b) DSC曲线；(c) 离子电导率；(d) Li^+与不同O原子的径向分布函数（RDF）；(e) Li^+-$TFSI^-$配位数对比；(f) 锂离子迁移数对比；(g) Li/Li对称电池循环测试；(h) Li||SPE||LFP电池的循环性能

Fig. 1　(a_1) Preparation of PEO-DES electrolytes, (a_2) Competitive coordination of DMMSA with Li^+ weakening EO-Li^+ interaction, (b) DSC curves, (c) Ionic conductivity, (d) RDFs curves, (e) Coordination numbers of Li^+-$TFSI^-$, (f) Li-ion transference numbers, (g) Cycling tests of Li/Li symmetric cells, (h) Cycling performance of Li||SPE||LFP cells

质动力学。进一步引入LiDFOB/LiTFSI双盐，氧化稳定性提升至4.5 V[图3(e)]，对称电池稳定循环长达900 h[图3(f)]，XPS确认原位生成了富含LiF的SEI[图3(g)]。全电池在60℃、0.5 C下经300次循环容量保持率88%[图3(h)]。该体系通过半互穿网络设计，在体相构建了力学骨架与快速传质域的分级结构，并在界面通过双盐化学原位钝化，将链段松弛、离子对解离与界面输运的多级调控落实到同一材料平台。

1.4　热压层合构建锂化有机纳米纤维增强网络：界面氢键与快速离子通道

针对溶液浇铸法填充纳米骨架时存在的缺陷和界面剥离问题[58-59]，本团队发展了真空辅助热压层合技术，将锂化有机纳米纤维膜（LOF）与PEO基体复合，制备了结构致密的LOF-CPE[图4(a)~(d)][60]。LOF膜通过静电纺丝PAN、bPEI接枝及Li^+动态吸附制得，表面胺基与PEO醚氧形成氢键，显著抑制结晶[图4(e)，(f)]。30℃下LOF-CPE离子电导率7.41×10^{-5} S/cm，较SPE提升逾一个数量级，活化能降至0.83 eV[图4(g)，(h)]。基于此LOF-CPE的对称电池稳定循环超900 h[图4(i)，(j)]，固态$LiFePO_4$电池在0.5 C、60℃下500圈容量保持率82%[图4(k)]。该工作阐明了界面氢键抑制结晶-界面通道促进传输的协同机制，将界面输运调控从电极界面拓展到电解质内部增强相界面，为多级调控提供了新维度。

1.5　预浸润构建硅碳负极连续离子传导网络：体相机械稳定性的主导作用

微米硅负极面临巨大的体积变化和固-固界面接触难题[61-62]。本团队系统研究了PEO基全固态电

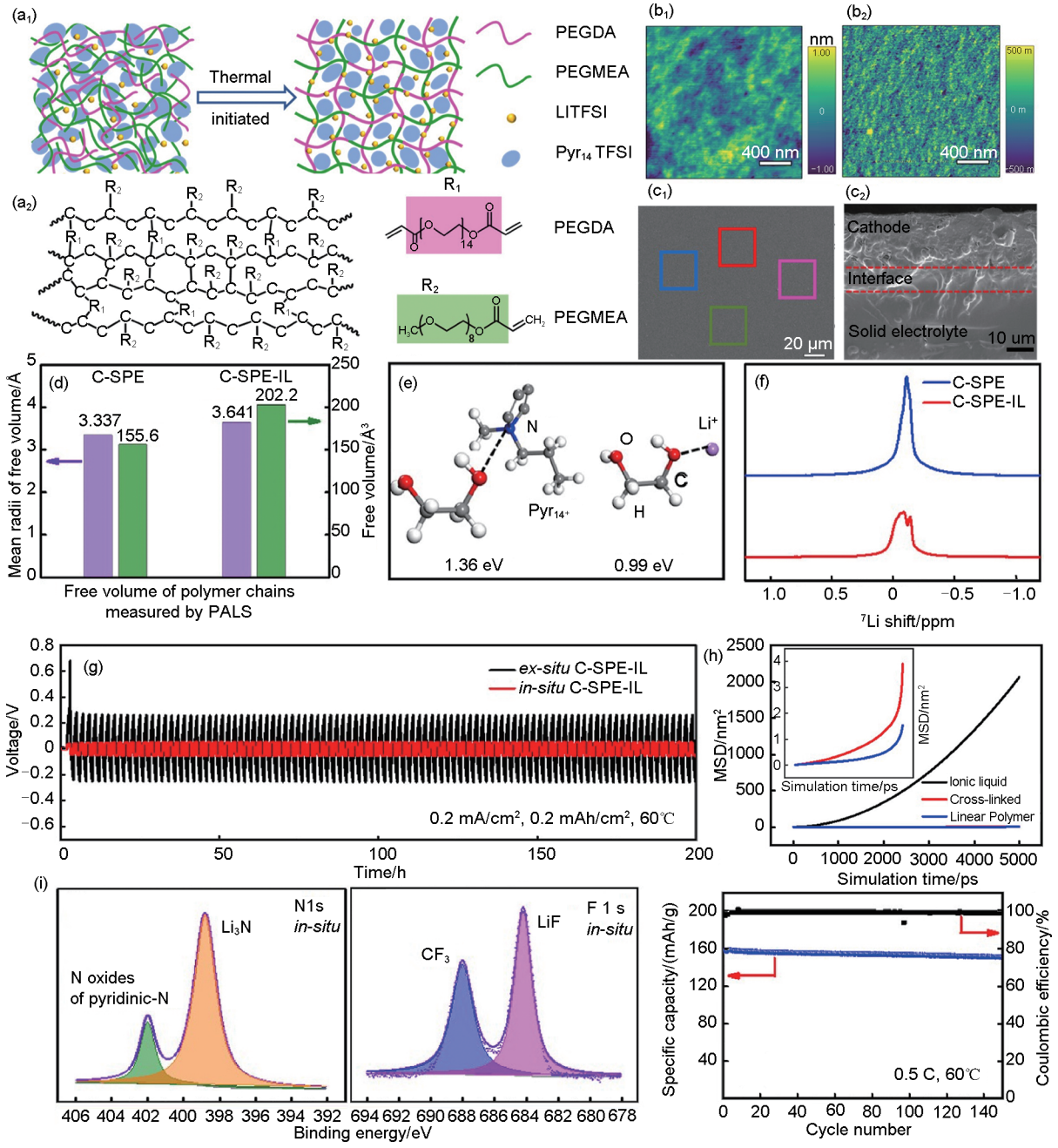

图2 (a_1) C-SPE-IL原位热聚合制备过程；(a_2) 三维交联网络化学结构；(b_1) (b_2) AFM形貌与相图；(c_1) (c_2) SEM表面图像及区域EDS；(d) C-SPE与C-SPE-IL的自由体积；(e) DFT计算；(f) 固态^{7}Li NMR谱；(g) 锂对称电池循环测试；(h) 不同体系中Li^+的均方位移；(i) 循环后锂负极N 1*s*与F 1*s* XPS谱；(j) $LiFePO_4$||原位C-SPE-IL||Li电池的循环性能

Fig. 2 (a_1) *In-situ* thermal polymerization process, (a_2) Cross-linked network structure, (b_1) (b_2) AFM morphology and phase images, (c_1) (c_2) SEM and EDS, (d) Free volume parameters, (e) DFT binding energies, (f) Solid-state ^{7}Li NMR, (g) Cycling tests of symmetric lithium cells, (h) MSD of Li^+, (i) XPS of cycled Li anode, (j) Cycling performance of $LiLiFePO_4$||in-situ C-SPE-IL||Li

池中Si/C负极的界面工程，对比了原始、液态预锂化和聚合物预浸润三种电极结构[图5(a_1)(a_2)][63]。研究发现，60℃下PEO-IL虽离子电导率更高，但机械强度不足导致界面脱黏[图5(b_3)(b_4)]，高倍率性能急剧衰减。PEO-LiTFSI凭借更好的力学稳定性，即使电导率较低，仍获得更优的倍率性能[图5(c_2)]，揭示出在该温度下机械稳定性对固-固界面输运起主导作用。预浸润策略通过溶液浇铸使聚合物

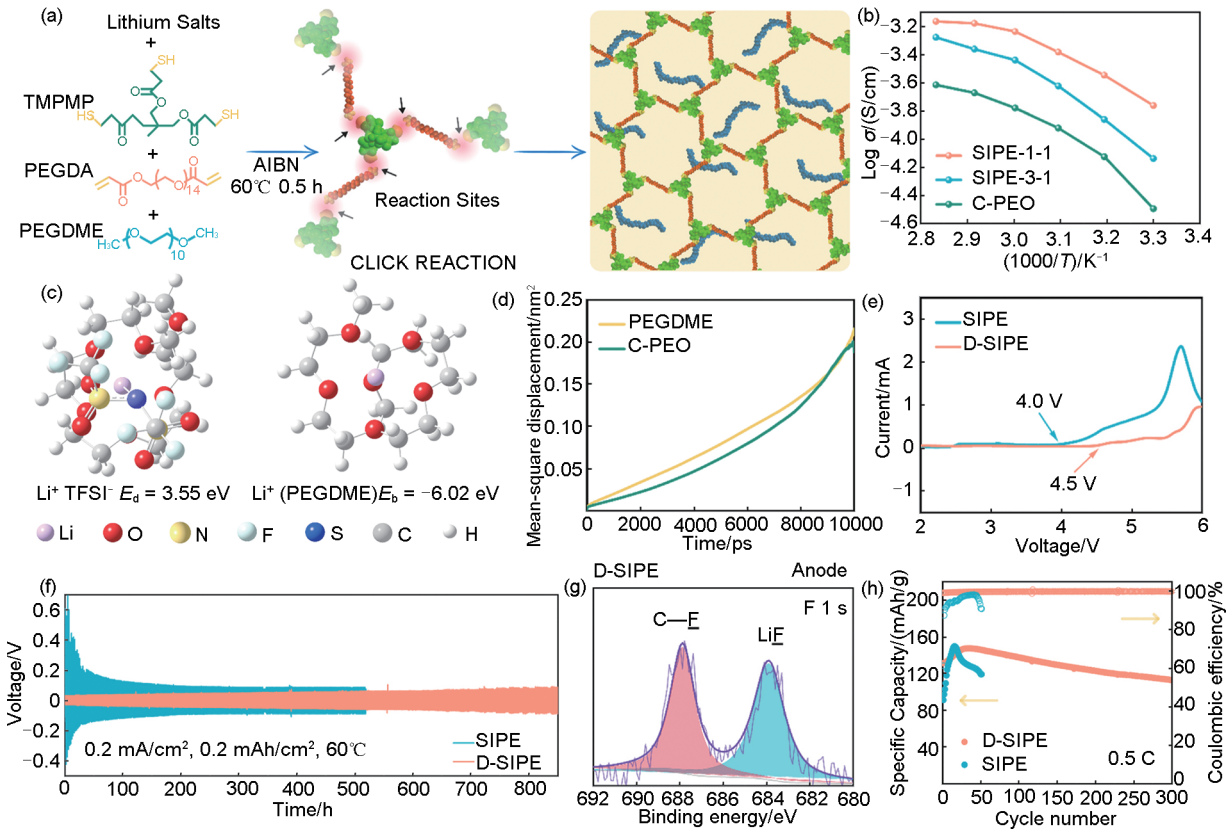

图 3　(a) SIPE 合成策略与结构示意图；(b) 离子电导率；(c) DFT 计算；(d) MSD 曲线；(e) LSV 曲线；(f) 对称电池循环测试；(g) 循环后锂负极 F 1*s* XPS；(h) 固态 Li/LiFePO$_4$ 电池循环性能

Fig. 3　(a) Synthesis and structure of SIPE, (b) Ionic conductivity, (c) DFT calculation of LiTFSI/PEGDME complex, (d) MSD curves, (e) LSV profiles, (f) Cycling tests of symmetric lithium cells, (g) F 1*s* XPS of cycled Li anode, (h) Cycling performance of Li/LiFePO$_4$ cells

电解质渗透入电极孔隙，预先构建三维连续的离子传导网络，将界面离子输运从依赖外压接触转变为内在连通。预浸润电极在 800 mA/g 下初始比容量超 2000 mAh/g，100 圈后仍保持约 1000 mAh/g，极化电压最低，而原始和预锂化电极容量衰减迅速[图 5(e$_2$)]。倍率测试后容量可逆恢复，证实了连续网络的结构稳定性[图 5(e$_1$)]。该工作从电极尺度揭示了界面输运连续性的构建与体相机械完整性的协同关系，丰富了多级耦合调控中界面输运的内涵。

2　聚环氧乙烷基复合电解质结构设计与离子传输

近年来，通过将 PEO 基体与填料复合，利用各组分协同效应实现性能调控[64]。例如无机惰性填料通过 Lewis 酸碱作用促进锂盐解离并抑制 PEO 结晶[65-66]，活性填料自身的高离子电导率可在填料/聚合物界面形成快速渗流通道[67-68]。在形貌设计上，零维纳米填料提供高比表面积界面相互作用位点[69]；一维填料构建长程连续离子通道并力学增强[70]；二维纳米片沿平面方向提供低能垒迁移路径[71]；三维互穿网络则实现体相均匀的快速传导与力学协同[72]。制备工艺方面，溶液混合-浇铸法操作简便但填料易团聚[73]，熔融共混可改善分散[74]，界面修饰层策略可有效缓解电极/电解质界面不稳定性[75-76]。总之，PEO 基复合电解质正从单一填料共混走向多组分、多尺度、多功能设计，填料/聚合物界面的精准调控是突破性能瓶颈的核心。

2.1　金属有机框架金属活性位点配位阴离子：锚定阴离子驱动锂盐高效解离

PEO 基电解质的离子电导率与 t_{Li^+} 长期受限于聚合物链段的高结晶度以及锂盐阴离子的自由迁移[77-78]。针对上述瓶颈，本团队提出利用金属有机框架（MOF）中金属活性位点作为路易斯酸中心，通过吸附 TFSI$^-$，实现自由 Li$^+$ 的高效释放与快速迁

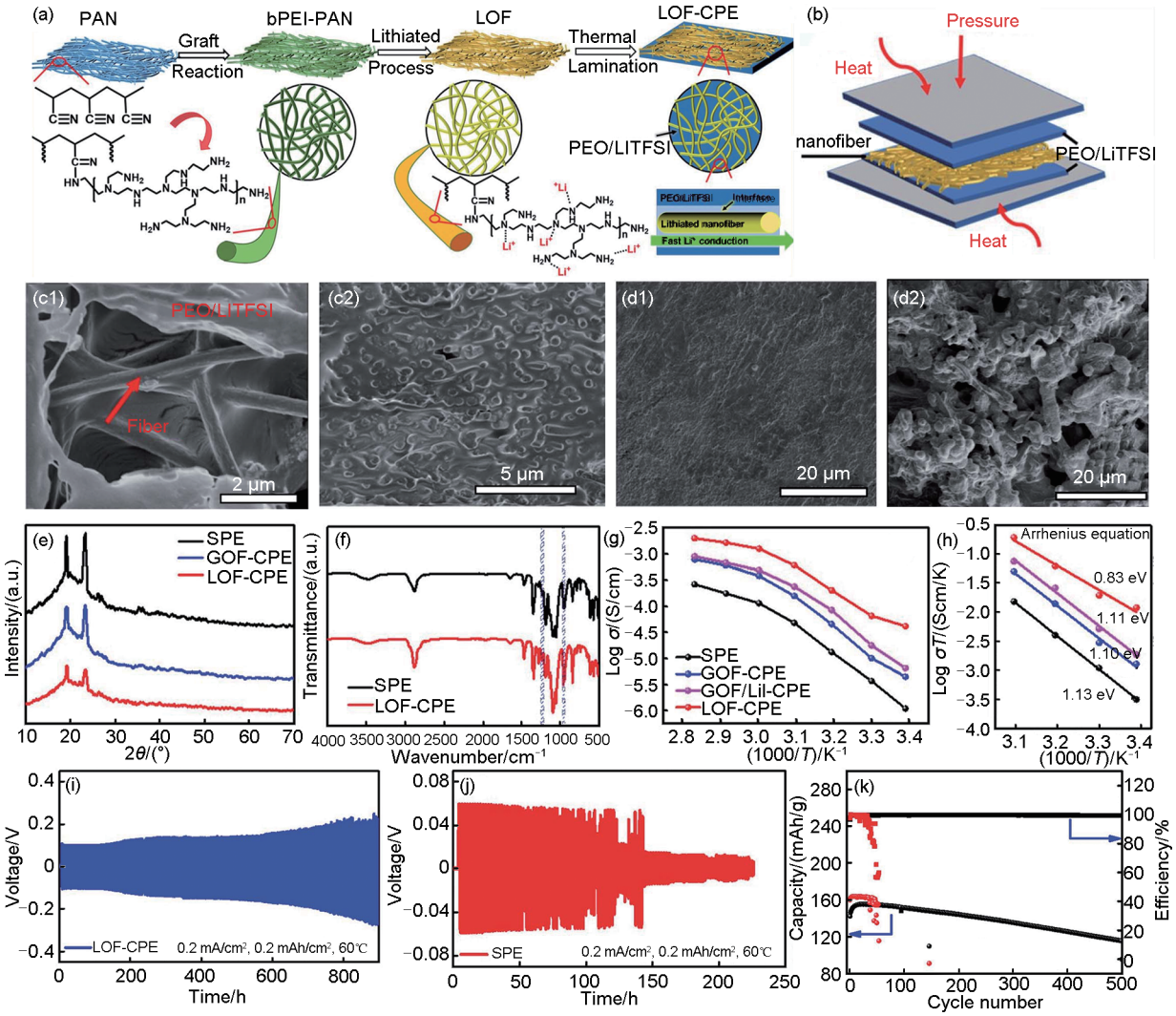

图 4 (a) LOF-CPE制备流程；(b) 热压层合示意图；(c_1) (c_2) LOF-CPE表面及截面SEM；(d_1) (d_2) 循环后锂负极表面SEM；(e) XRD图谱；(f) FTIR光谱；(g) 离子电导率；(h) 活化能；(i), (j) 锂对称电池的循环性能；(k) $LiFePO_4$||Li电池循环性能

Fig. 4 (a) Preparation of LOF-CPE, (b) Thermal lamination, (c_1) (c_2) Surface and cross-sectional SEM, (d_1) (d_2) SEM of Li anodes after cycling, (e) XRD, (f) FTIR, (g) Ionic conductivity, (h) Activation energies, (i), (j) Cycling tests of symmetric lithium cells, (k) Cycling performance of $LiFePO_4$||Li cells

移[图6(a_1)~(a_3)][79]。

Cu-MOF-74经热活化后暴露出丰富的Cu^{2+}活性位点，可与$TFSI^-$阴离子进行选择性配位。DFT计算表明，$TFSI^-$在金属活性位点的吸附能为−1.679 eV，远低于DMF，热力学上优先占据金属位点[图6(a_4)(a_5)]，从根源上抑制阴离子迁移并高效解离离子对。40% Li-MOF/PEO的t_{Li^+}从0.14跃升至0.36，30℃离子电导率达5.5×10^{-5} S/cm，较纯PEO提高一个数量级以上，结晶度由32.7%降至18.3%。此外，Li-MOF/PEO在0.2 mA/cm²、60℃下稳定循环超800 h[图6(c_1)]，而纯PEO体系200 h内即短路[图6(b_1)]。循环后锂负极SEM显示Li-MOF/PEO保护下锂表面致密光滑，纯PEO体系出现大量枝晶[图6(c_2)(c_3)]。$LiFePO_4$/Li电池在0.5 C、60℃下初始比容量161 mAh/g，300圈后容量保持率95%。该工作首次系统揭示了MOF中金属活性位点对$TFSI^-$的强配位效应，为从阴离子锚定角度提升迁移数与电导率提供了明确的设计准则。

2.2 热塑性聚氨酯共混与二维锂镁硅酸盐协同增强：多组分协同一体化

针对离子电导率、机械强度与界面稳定性难以兼顾的瓶颈[80]，本团队设计了热塑性聚氨酯（TPU）与二维锂镁硅酸盐（LMS）纳米片协同改性的PTLL复合电解质[图7(a)][81]。TPU的无定形域

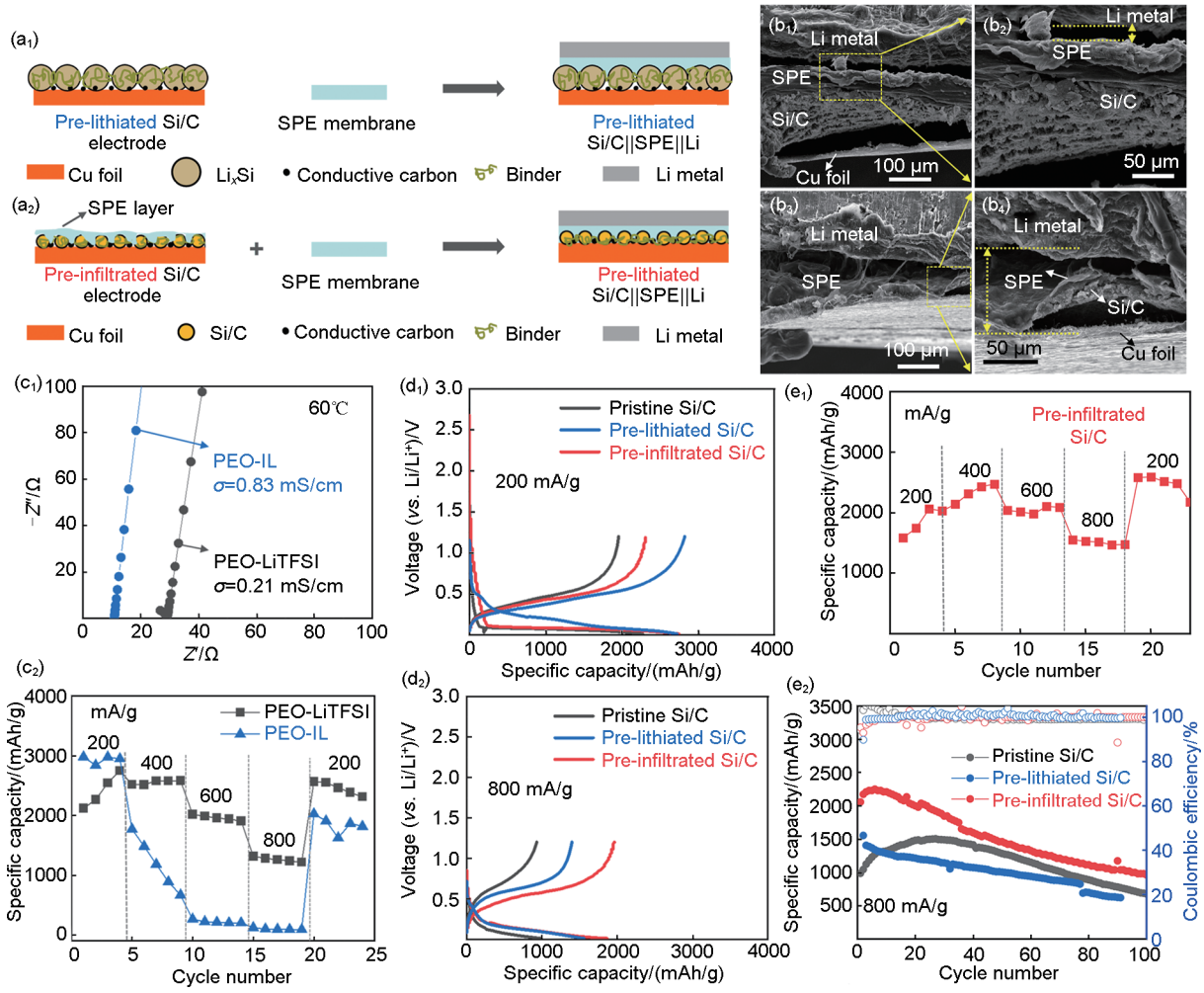

图 5　(a_1) (a_2) 预锂化与预浸润Si/C电极的全电池结构示意；(b_1)~(b_4) PEO-LiTFSI与PEO-IL电解质下截面SEM；(c_1) Nyquist图；(c_2) 原始电极的倍率性能；(d_1) (d_2) 不同电流密度下的电压曲线；(e_1) 预浸润电极倍率性能；(e_2) 三种电极在800 mA/g下的循环性能

Fig. 5　(a_1) (a_2) Schematics of cells with prelithiated and preinfiltrated Si/C electrodes, (b_1)—(b_4) Cross-sectional SEM with different electrolytes, (c_1) Nyquist plots, (c_2) Rate performances of pristine electrodes, (d_1) (d_2) Voltage profiles, (e_1) Rate performance of preinfiltrated electrode, (e_2) Cycling performance at 800 mA/g

有效抑制PEO结晶，其极性基团（—NH、C═O）与$TFSI^-$及PEO链段产生相互作用；二维LMS纳米片利用表面Lewis酸位点（Mg^{2+}）进一步固定$TFSI^-$。XRD显示PTLL结晶度最低[图7(b)]；FTIR与Raman证实LMS表面Lewis酸位点与$TFSI^-$存在强相互作用，游离$TFSI^-$比例仅8.5%。PTLL在30℃离子电导率达2.48×10^{-4} S/cm，t_{Li^+}为0.42，显著优于PTL和PL[图7(c)，(e)]。该体系中，TPU承担增塑与抑制结晶功能，LMS承担阴离子锚定功能，多组分协同实现了“增塑-解离-传导”在体相的均匀分布。对称电池在60℃、0.1 mA/cm²下稳定循环超800 h，极化电压平稳[图7(d)]。Li/LFP电池在0.5 C下循环45圈后比容量仍高于对比体系，平均效率超99.8%，Li/NCM811电池在0.1 C下初始容量达155.4 mAh/g[图7(f)]。该工作展示了多组分协同设计在实现多功能一体化提升方面的显著优势。

2.3　Janus层构筑的LLZTO/PEO界面调控：化学相容性工程

石榴石型LLZTO等活性填料与PEO基体的界面不相容性常导致填料团聚和离子传输中断[82-83]。本团队设计了3-甲基丙烯酰氧丙基三甲氧基硅烷（MEMO）Janus层包覆的MEMO@LLZTO填料，系统揭示了界面层对离子输运的调控机制[84]。MEMO分子通过水解缩合与LLZTO表面形成Si—

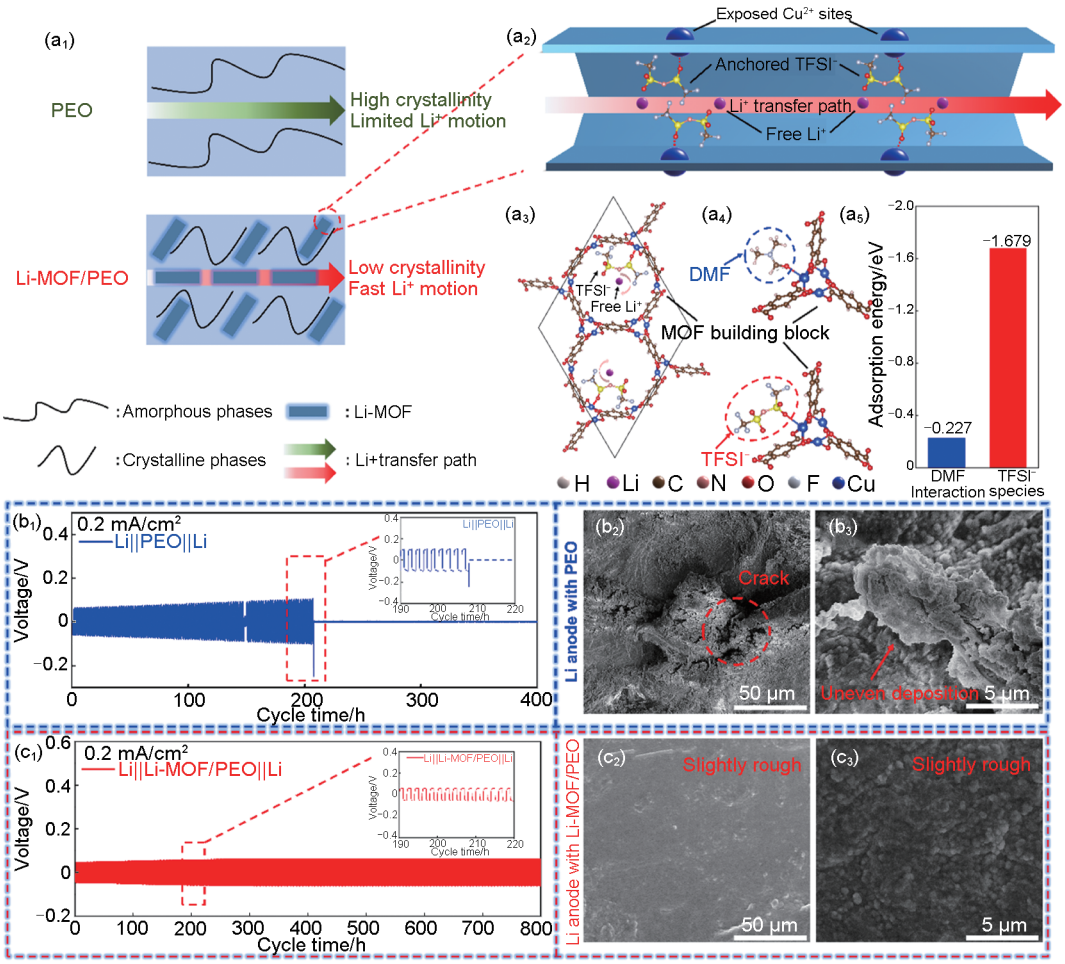

图 6 (a_1) Li-MOF 对 PEO 基体结晶度与 Li^+ 传导影响示意图；(a_2) 金属活性位点（OMSs）吸附 $TFSI^-$ 及 Li^+ 扩散路径；(a_3) Cu-MOF-74 锚定 $TFSI^-$ 释放自由 Li^+；(a_4) DMF 与 $TFSI^-$ 在单个活性位点上的优化构型；(a_5) DFT 吸附能计算结果；(b_1) Li||PEO||Li 对称电池循环测试；(b_2) (b_3) 循环后锂负极 SEM 图像；(c_1) Li||Li-MOF/PEO||Li 对称电池循环测试；(c_2) (c_3) 循环后锂负极 SEM 形貌

Fig. 6 (a_1) Schematic of Li-MOF effect on PEO crystallinity and Li^+ conduction, (a_2) OMSs anchoring $TFSI^-$ and Li^+ diffusion path, (a_3) Schematic of $TFSI^-$ adsorption anchoring by EMSs and free Li^+ release, (a_4) Optimized configurations of DMF and $TFSI^-$ on a single, (a_5) DFT adsorption energies, (b_1) Cycling tests of Li||PEO||Li symmetric cell, (b_2) (b_3) SEM of cycled Li anode, (c_1) Cycling tests of Li||Li-MOF/PEO||Li cell, (c_2) (c_3) SEM of cycled Li anode

O 化学键，其外侧丙烯酰氧基团与 PEO 链段亲和，内侧化学屏蔽 Li_2CO_3 杂质，实现填料从不相容到相容的本质转变。10% MEMO@LLZTO（6 h）-PEO 在 30℃下离子电导率达 2.16×10^{-4} S/cm，较未修饰体系提升近一个数量级，活化能从 0.52 eV 降至 0.34 eV[图 8(d)]。DFT 计算揭示，MEMO 与 LLZTO 及 PEO 的界面结合能（−1.537 eV）显著优于 LLZTO/PEO 直接界面（−0.812 eV）；同时 MEMO 与 $TFSI^-$ 的吸附能（−1.5696 eV）远强于 Li^+-$TFSI^-$ 离子对自身结合（−0.5216 eV），高效解离离子对并释放自由 Li^+。t_{Li^+} 从 0.28 跃升至 0.53。引入无纺布骨架支撑后[图 8(c)]，$LiFePO_4$ 全电池在 0.5 C、60℃下循环 100 圈放电比容量仍高于 150 mAh/g[图 8(e)]。该工作通过 Janus 界面层同时解决了填料分散、阴离子迁移及界面副反应三大难题，充分验证了“阴离子锚定-界面钝化-体相连续导锂”的协同设计思路。

3 总结与展望

本文从聚环氧乙烷基电解质离子传输机制出发，详细讨论了离子电导率、锂离子迁移数、机械强度与界面稳定性之间的内在制约，总结了近年来

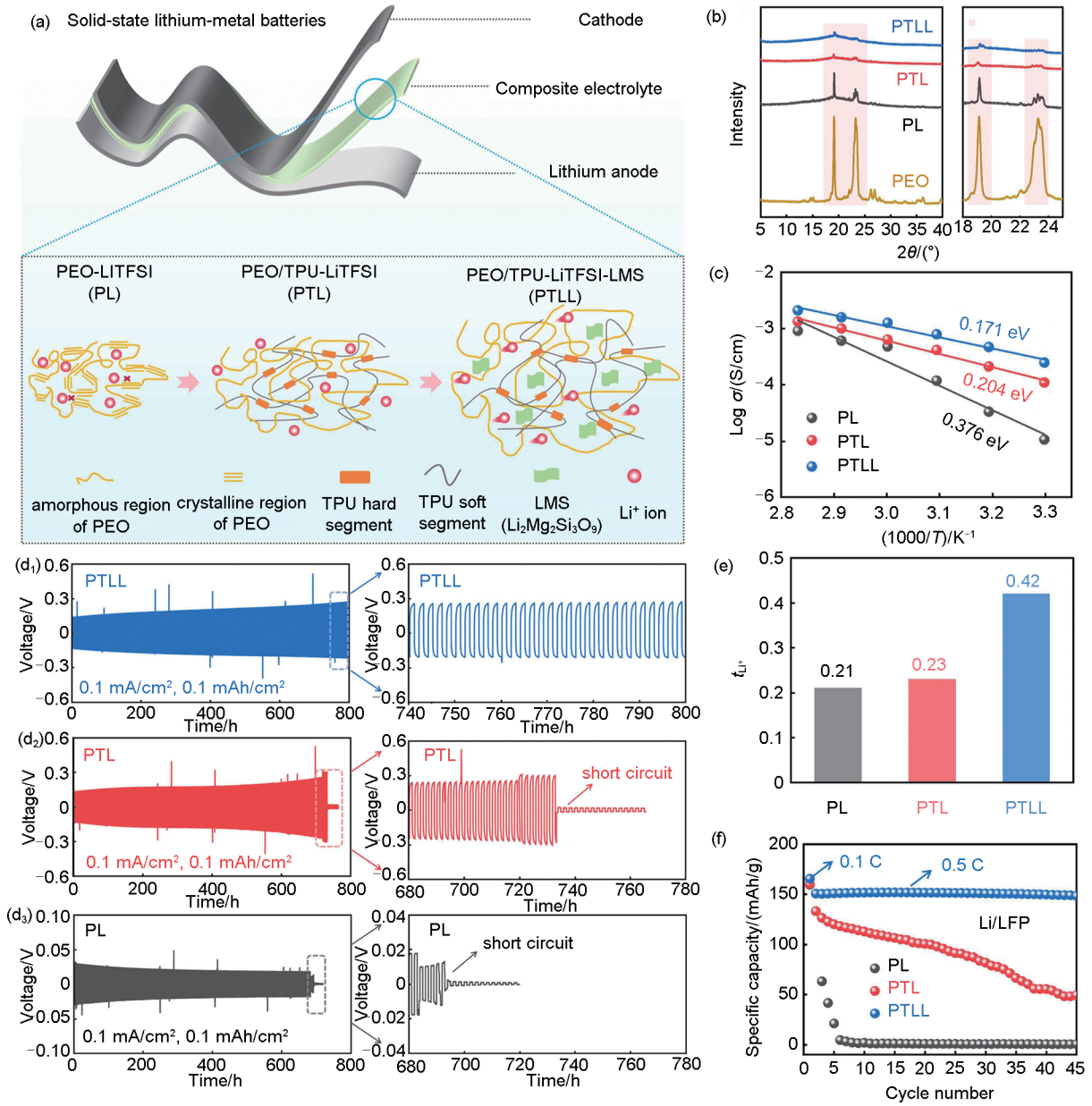

图 7　(a) PTLL 复合电解质结构设计示意图；(b) XRD 图谱；(c) 离子电导率对比；(d_1)~(d_3) 锂对称电池循环测试；(e) 锂离子迁移数对比；(f) Li/LFP 电池循环性能

Fig. 7　(a) Design of PTLL composite electrolyte, (b) XRD patterns. (c) Ionic conductivity, (d_1)—(d_3) Cycling tests of Li symmetric cells, (e) Li-ion transference numbers, (f) Cycling performance of Li/LFP cells

通过结构设计提升聚环氧乙烷电解质性能的研究成果。明确离子电导率的提升依赖于增加非晶区比例或引入快离子传导组分，但常以牺牲力学性能为代价；深共晶溶剂增塑与离子液体封装虽显著提升电导率，但半互穿网络与纳米纤维增强策略通过交联或骨架结构实现了更好的性能平衡；锂离子迁移数的提升依赖于对阴离子的有效固定，活性位点锚定能高效束缚阴离子，但需通过界面调控解决填料团聚与界面阻抗上升问题；界面稳定性的改善常伴随电导率或加工柔性的牺牲，双盐体系通过富氟化锂界面层提升负极稳定性，但需权衡对链段运动能力的影响。

当前匹配三元或富锂锰基正极时，聚合物链上醚氧基团易氧化分解，锂盐阴离子分解产生氢氟酸等腐蚀性物种，导致高阻抗正极电解质界面层生成和过渡金属溶出，界面阻抗持续增长。后续研究需

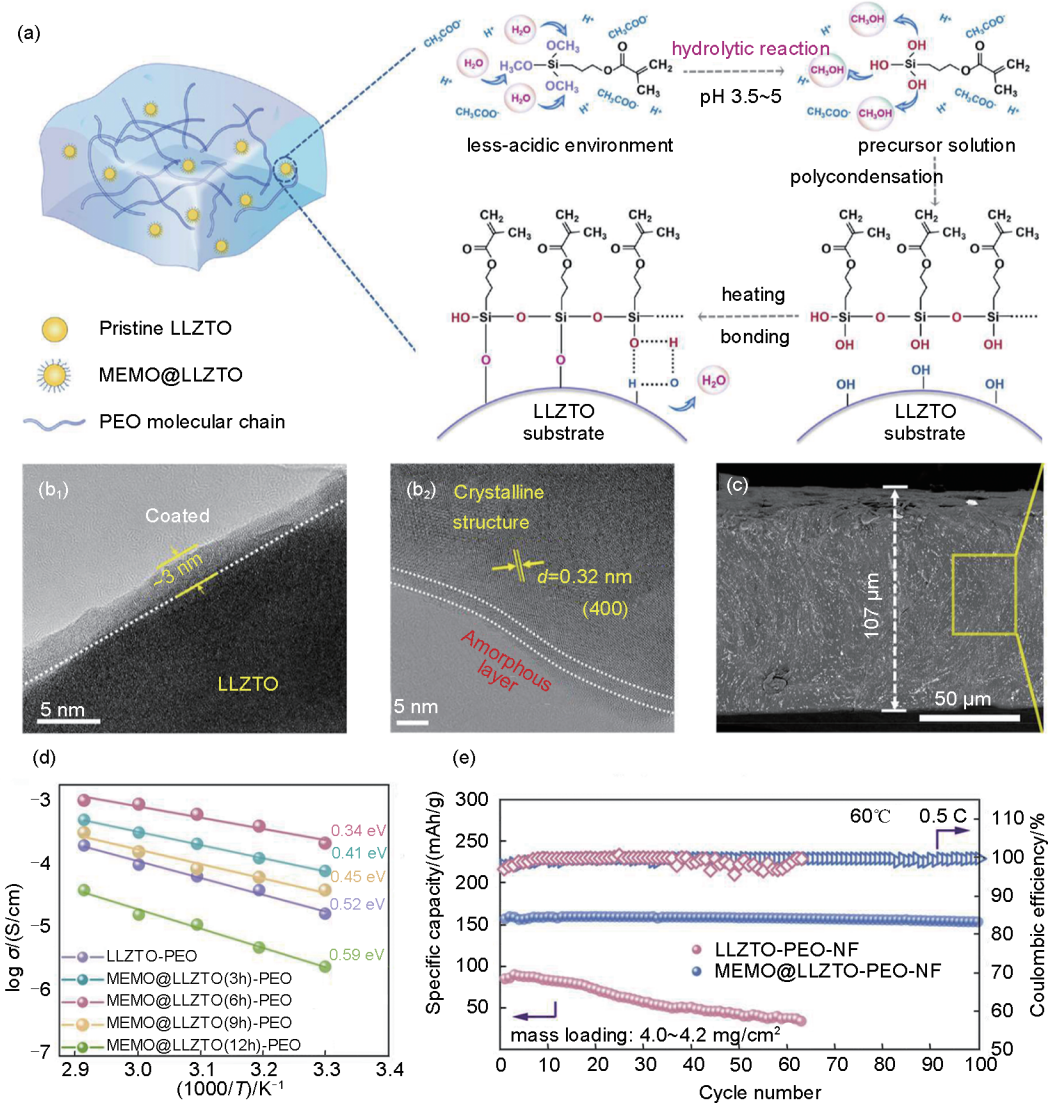

图 8 (a) 基于 MEMO@LLZTO-PEO 的全固态锂电池结构示意图；(b_1) (b_2) MEMO@LLZTO（6 h）颗粒的 TEM 形貌；(c) MEMO@LLZTO（6 h）-PEO-NF 复合电解质膜截面 SEM 形貌；(d) 不同 MEMO 包覆时间的 Arrhenius 曲线；(e) Li||MEMO@LLZTO-PEO-NF||$LiFePO_4$ 全电池在 60℃、0.5 C 下的循环性能

Fig. 8 (a) Schematic of all-solid-state battery using MEMO@LLZTO-PEO electrolyte, (b_1) (b_2) TEM of MEMO@LLZTO(6 h), (c) Cross-sectional SEM of MEMO@LLZTO-PEO-NF. (d) Arrhenius plots, (e) Cycling performance at 60℃, 0.5 C

结合原位表征深入解析界面失效机制，发展抗氧化聚合物基体或正极侧界面保护层。着力发展室温链段运动能力强的低结晶聚合物基质，结合界面工程拓宽电化学窗口，探索可规模化的制膜工艺。基于此，面向实际应用的超薄化、梯度化复合电解质结构及其连续制造技术，将成为未来重点发展方向。

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