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A Critical Review on Composite Polymer Electrolytes for High-Energy Solid-State Li–S Batteries

Omkar Sangabathula, Rushikesh Kale, and Chan-Jin Park†

Department of Materials Science and Engineering, Chonnam National University, 77, Yongbong-ro, Bukgu, Gwangju 61186, South Korea

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Composite polymer electrolytes (CPEs) are a promising class of solid-state electrolytes for high-performance lithium-sulfur (Li–S) batteries. Li–S chemistry offers a very high theoretical specific capacity (~1675 mAh g^{-1}) and energy density (~2600 Wh kg^{-1}) along with low material costs. However, its practical application is limited by the insulating nature of sulfur/Li_2S, the polysulfide shuttle effect, and the instability of lithium-metal anodes. Replacing the liquid electrolyte with a solid-state design can address many of these issues by eliminating flammable solvents and restraining polysulfide migration. However, traditional polymers often exhibit insufficient ionic conductivity, while ceramics tend to suffer from brittleness and poor electrode contact. CPEs overcome these trade-offs by integrating a flexible polymer matrix with functional inorganic fillers, creating synergistic ion-conduction networks that enhance Li^+ conductivity, immobilize polysulfides, and improve mechanical strength. This mini-review outlines the key challenges in Li–S batteries, highlights recent advances in polymer and polymer-ceramic hybrid electrolytes, and discusses strategies to improve Li^+ transport, interfacial stability, and sulfur utilization. It also examines remaining bottlenecks and future research directions to provide a focused perspective on the design principles needed for practical high-energy solid-state Li–S batteries (SSLSBs).

Keywords: *Composite polymer electrolytes, Solid-state Li–S batteries, Polysulfide suppression, Li-metal interface stability, Ion-transport networks*

1. Introduction

Lithium-sulfur (Li–S) batteries are regarded as a promising high-energy storage system, with a theoretical sulfur capacity of ~1675 mAh g^{-1} and an energy density up to ~2600 Wh kg^{-1}. Sulfur is inexpensive, abundant, and environmentally benign, making Li–S chemistry attractive for large-scale applications beyond conventional Li-ion batteries. However, Li–S cells face several persistent challenges that hinder their practical development [1]. In liquid-electrolyte Li–S cells, repeated cycling causes loss of active material, rapid capacity fade, and escalating safety risks (e.g., due to the polysulfide shuttle), ultimately preventing commercialization.

Replacing the LEs with a solid-state electrolyte (SSE) is a compelling approach to improve safety and eliminate the polysulfide shuttle in Li–S cells [3]. Among SSE options, composite polymer electrolytes (CPEs), which combine a polymer matrix with inorganic filler particles, have attracted significant attention. CPEs are designed to marry the high ionic conductivity and stability of inorganic solids with the flexibility and processability of polymers, thereby leveraging the strengths of each. Related electrolyte systems include solid polymer electrolytes (SPEs), gel polymer electrolytes (GPEs), and traditional liquid electrolytes (LEs), each with distinct trade-offs between conductivity, stability, and safety. In the following sections, this mini-review examines the fundamental challenges of Li–S batteries, explains how SSLSB configurations (particularly CPEs) address these issues, and summarizes recent progress in CPE development for SSLSBs [4,5].

2. Li–S Battery Challenges and Solid-State Rationale for SSLSBs

Li–S batteries face several intrinsic challenges that

†Corresponding author: parkcj@jnu.ac.kr
Omkar Sangabathula: Postdoctorial researcher, Rushkesh Kale: Doctorial student, Chan-Jin Park: Professor

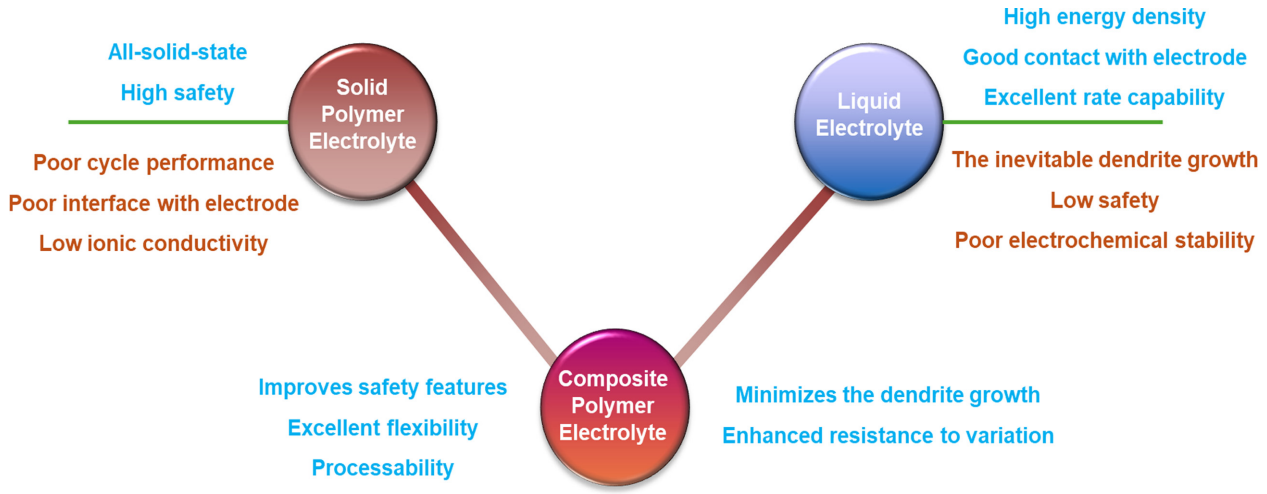

Fig. 1. Key electrolyte systems for SSLSBs: Solid Polymer Electrolytes (SPEs), Liquid Electrolytes (LEs), and Composite Polymer (CPEs), and Gel Polymer Electrolytes (GPEs) [2]Click or tap here to enter text.

severely limit their long-term performance. First, sulfur and its fully reduced product Li_2S are both electronically and ionically insulating (for Li_2S, ~10^{-14} - 10^{-13} S cm^{-1} electronic and ~10^{-9} S cm^{-1} ionic conductivity), leading to sluggish redox kinetics, high polarization, and incomplete sulfur utilization in the absence of conductive additives [3,6]. Second, in a liquid-electrolyte Li–S cell, dissolved lithium polysulfide species (Li_2S_x, $4 \le x \le 8$) form during discharge and cause the well-known polysulfide shuttle. These mobile intermediates diffuse to the Li metal anode and are reduced parasitically, which consumes active material and electrolyte and yields a low Coulombic efficiency; the shuttle process also destabilizes the Li surface, creating heterogeneous Li^+ flux and promoting dendritic deposition [1]. Third, the lithium-metal anode is highly reactive and prone to dendrite formation, SEI rupture, and interfacial voids upon repeated plating/stripping, especially at high current densities [7]. These issues collectively undermine SSLSB safety, cycle life, and reversibility [5]. Moreover, the above challenges tend to exacerbate each other (e.g. polysulfide leakage accelerates Li corrosion), so an effective Li–S battery design must address all three simultaneously.

Adopting a solid-state electrolyte (SSE) in place of the liquid electrolyte is a direct route to improve safety and mitigate polysulfide degradation in SSLSBs. SSLSBs eliminate flammable solvents, avoid leakage, and physically confine lithium polysulfides, thereby greatly suppressing the shuttle effect [8]. Dense inorganic SSEs (e.g., NASICON-type phosphates or argyrodite sulfides) can effectively block polysulfide transport due to their negligible porosity and chemical stability. However, ceramic SSEs, despite their high ionic conductivities (10^{-4} - 10^{-2} S cm^{-1}), are brittle and form poor interfacial contacts with electrodes, leading to large interfacial impedance and incomplete sulfur utilization [8,9]. By contrast, solid polymer electrolytes (SPEs) are mechanically flexible, conformal to electrode surfaces, and easily fabricated into thin films (yielding excellent interface contact), but their room-temperature Li^+ conductivities (<10^{-4} S cm^{-1}) are low and their softness allows lithium dendrite penetration in high-current SSLSBs [10]. Thus, neither ceramics nor polymers alone are ideal; this trade-off motivates composite approaches that merge their advantages.

3. Composite Polymer Electrolytes: Design Principles and Advantages

Composite polymer electrolytes (CPEs, along with related gel polymer electrolytes, GPEs) are specifically designed to combine the strengths of polymers and ceramics by embedding inorganic filler particles into a polymer matrix, creating a hybrid Li^+ transport network. The ceramic fillers (e.g., Al_2O_3, SiO_2, ZrO_2, LAGP, LLZO) disrupt the polymer's crystallinity and provide

fast Li^+ conduction pathways, raising the electrolyte's ionic conductivity into the ~10^{-4} - 10^{-3} S cm^{-1} range as the filler content approaches a percolation threshold [7,9,11–13]. Notably, even a small loading of a superionic filler (for example, ~1 wt% of LSPS sulfide glass) can significantly enhance the conductivity - PEO-based composites with minimal LSPS have reached ~1.7 × 10^{-4} S cm^{-1} at 60 ºC. The polymer component of a CPE maintains intimate contact with electrode surfaces and provides mechanical flexibility, while the dispersed fillers reinforce the membrane's structural integrity and help stabilize the electrode interfaces [14]. In addition, many CPEs suppress polysulfide migration via Lewis acid-base interactions: for instance, polar oxide fillers like TiO_2, SiO_2 or ZrO_2 can chemically bind Li_2S_x species, and polar functional groups on the polymer can also immobilize polysulfides [8]. On the mechanical side, introducing a rigid inorganic phase into the polymer increases the modulus and improves resistance to dendrite penetration; for example, incorporating LLTO nanofibers into a PVDF-based electrolyte increased its tensile strength to ~9.5 MPa and enabled more stable Li-metal cycling [15]. Certain reactive fillers (e.g., sulfide additives) can even favorably interact with lithium metal to form a Li^+-conducting solid-electrolyte interphase (SEI), which promotes uniform Li deposition. By simultaneously enhancing ionic conductivity, stabilizing electrode interfaces, immobilizing polysulfides, and reinforcing mechanical strength, CPEs provide a uniquely balanced electrolyte architecture unattainable with either ceramics or polymers alone, making them a highly promising platform for long-life, high-energy, and safe SSLSBs [8,16,17].

4. Electrochemical Performance of CPEs

The rationale for using CPEs is best understood by contrasting the extremes of liquid vs. solid electrolytes. Liquid electrolytes provide excellent ionic transport and electrode wetting, leading to high sulfur utilization and fast kinetics, but they also pose safety hazards and allow polysulfide dissolution (the shuttle effect). Solid electrolytes (whether polymer or ceramic) avoid those liquid-related issues but often lack sufficient Li^+ conductivity or intimate contact with electrodes. This continuum of properties is illustrated in Fig. 1, which shows CPEs as a hybrid approach that combines polymeric flexibility with filler-conferred stability. By uniting these characteristics, a well-designed CPE aims to suppress dendrite growth, strengthen electrode interfaces, and sustain Li^+ transport, all while retaining the thin film processability needed for practical SSLSB architectures [2].

The contrast in sulfur redox behavior between liquid-electrolyte Li–S batteries and SSLSBs is illustrated in Fig. 2. In a liquid-electrolyte Li–S battery, sulfur undergoes a multistep reduction: S_8 rings open and form long-chain polysulfides in solution (Fig. 2a). This leads to the characteristic two-plateau discharge profile at ~2.3 V then ~2.1 V (Fig. 2c), corresponding to the stepwise conversion of S_8 into shorter polysulfides and ultimately Li_2S. Because these polysulfide intermediates are mobile, they diffuse to the lithium-metal anode and are reduced parasitically, yielding uneven SEI deposits and promoting dendritic Li growth (as seen in Fig. 2f). By contrast, an SSLSB prevents soluble intermediates from forming and forces the sulfur to convert via a direct solid-solid route (Fig. 2b, d). As a result, the discharge exhibits a single sloping plateau around ~2.0 V (Fig. 2e, g), which provides enhanced stability but typically limits the capacity to ~600 - 800 mAh g^{-1} unless additional ion-transport facilitators are introduced. The progression from Fig. 2a to 2g encapsulates the fundamental trade-off motivating CPE strategies: liquid electrolytes allow high active-material utilization but suffer poor stability, whereas SSLSBs give greater stability but at the cost of kinetic accessibility [19].

Recent progress with advanced polymer-based electrolytes illustrates how composite strategies overcome SSLSB limitations in practice. For instance, a molecularly engineered polymeric sulfur cathode (poly(S-PMEMA-DIB)) was shown to improve Li^+ coordination via its ethylene oxide-rich PMEMA segments. In an SSLSB, this sulfur host achieved ~981 - 1140 mAh g^{-1} discharge capacity with >98% Coulombic efficiency over extended cycling (Fig. 3a-d) [20]. Another example is a gel-polymer electrolyte formed by HFIP-induced in situ polymerization: this electrolyte attained a Li^+ transference number of ~0.60 and enabled Li‖Li symmetric-cell cycling for >800 hours at 0.5 mA cm^{-2} in an SSLSB configuration. When implemented in an SSLSB, the gel electrolyte delivered an initial ~1029 mAh g^{-1} and maintained ~698 mAh g^{-1} after 500 cycles (Fig. 3e-j) [21]. Ceramic-filled "quasi-

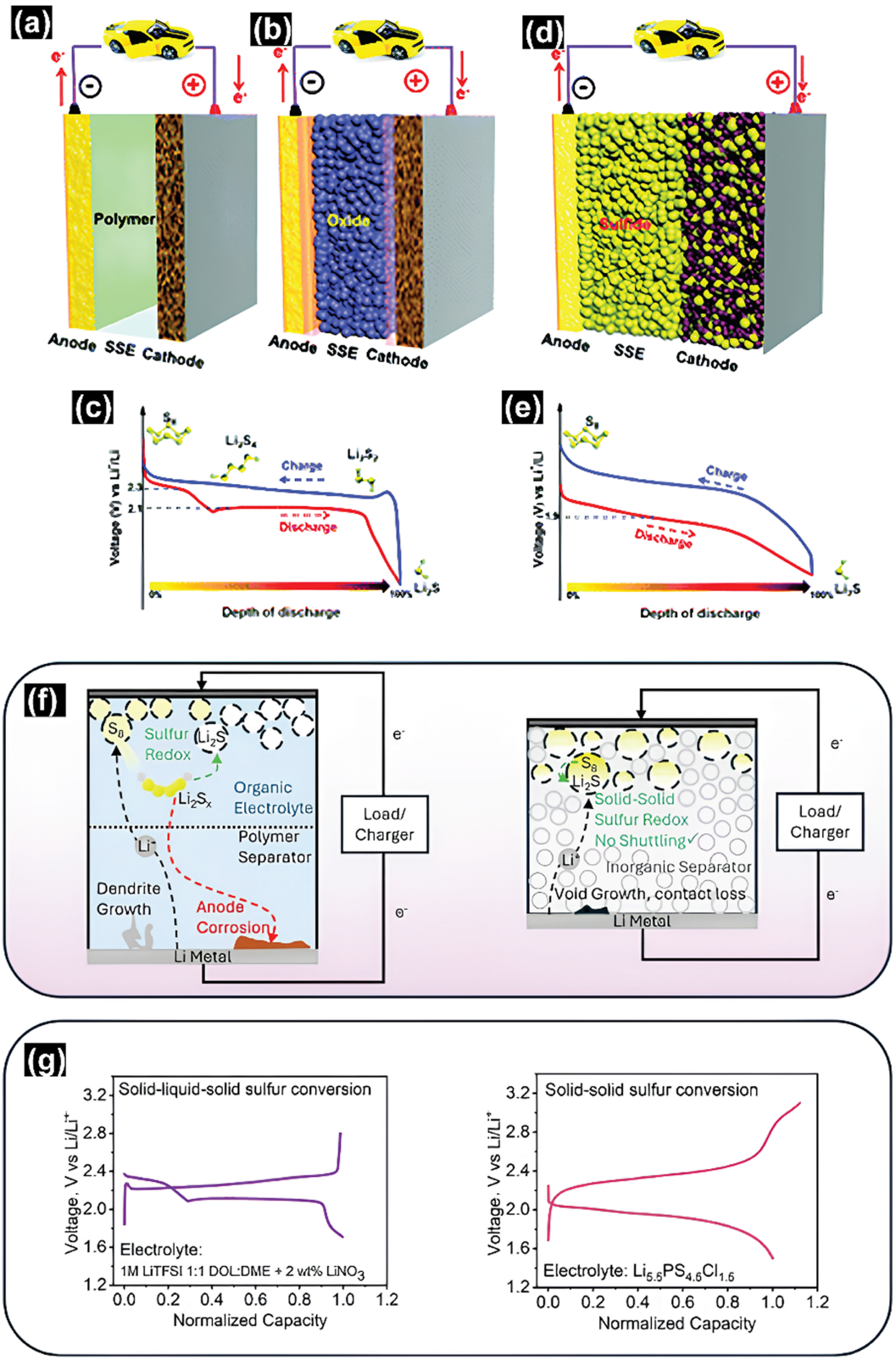

Fig. 2. Schematic comparison of electrolyte configurations and voltage profiles in LE vs.S SLSBs, (a-e) showing configurations for polymer, oxide and sulfide SSEs, and corresponding discharge profiles. Reprinted with permission from ref. [18]Click or tap here to enter text. Copyright 2020, Royal Society of Chemistry. Illustrating the transition from dual-plateau (LE) to single-plateau (SSE) behavior (f-g) [19]

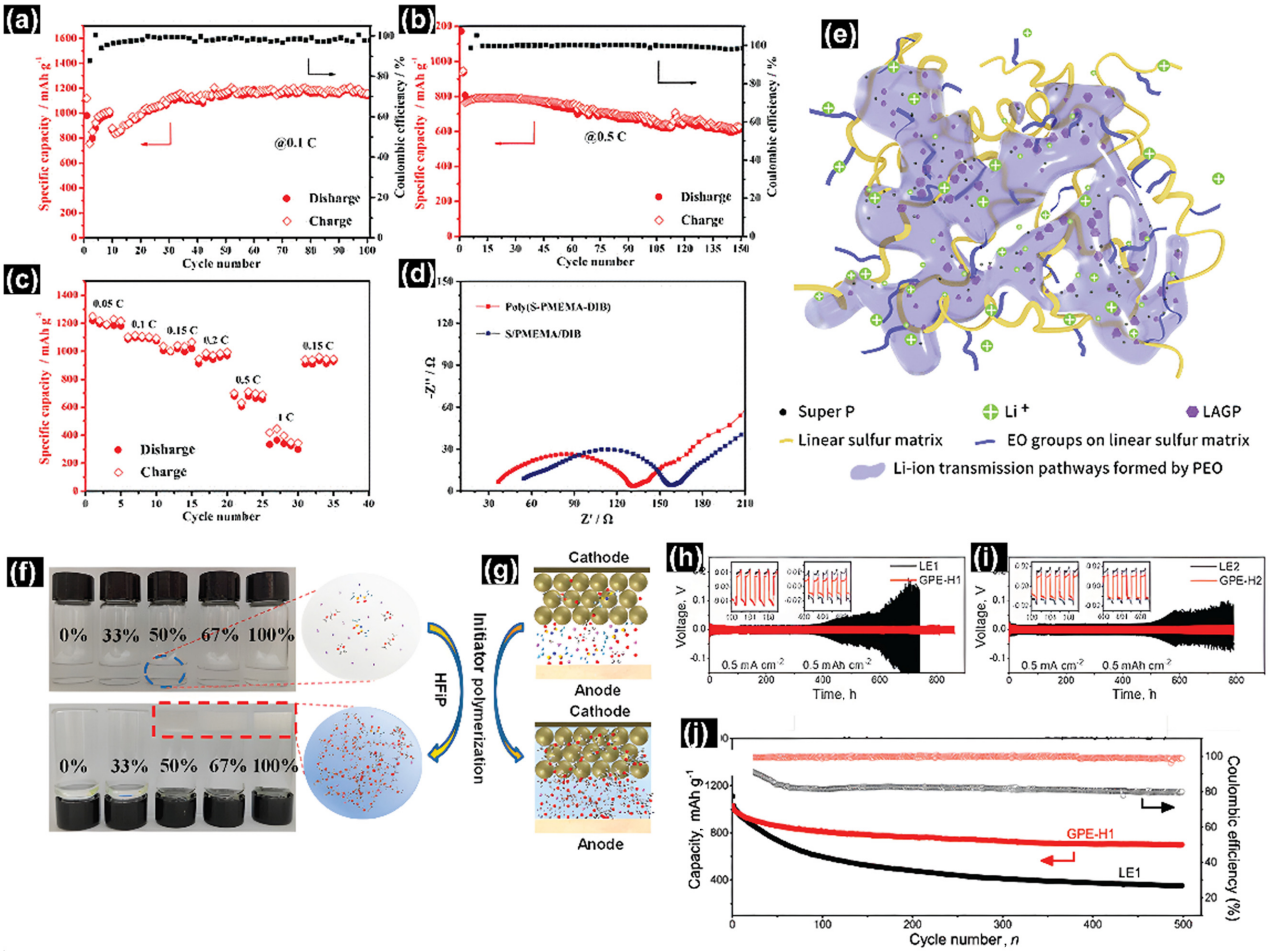

Fig. 3. Electrochemical performance of advanced polymer-based electrolytes for SSLSBs. (a-e) Cycling, rate capability, EIS, and electrode schematic for poly (S-PMEMA-DIB). Reprinted with permission from ref. [20]Click or tap here to enter text. Copyright 2020, American Chemical Society. (f–j) In situ gel electrolyte preparation, symmetric cell cycling, and full-cell performance [21]Click or tap here to enter text.

solid" polymer electrolytes have also demonstrated remarkable performance.

For example, a PPG-co-PETA polymer matrix loaded with ~66.8 wt% LAGP ceramic filler achieved ≈5.95×10^{-3} S cm^{-1} ionic conductivity with a Li^+ transference number of 0.83 (Fig. 4c, d). In SSLSBs this composite delivered an initial 1508 mAh g^{-1} and retained ~1109 mAh g^{-1} after 200 cycles at 0.25 C (Fig. 4f - h) [22]. Similarly, a PVDF-based CPE reinforced with LLTO nanofibers exhibited ~10^{-4} - 10^{-3} S cm^{-1} conductivity along with greatly improved mechanical properties (tensile strength 8-10 MPa, elongation >300%; Fig. 5a - e). Thanks to its broad electrochemical stability window (~5.1 V, Fig. 5f), this LLTO-PVDF electrolyte enabled a $LiFePO_4$‖Li SSLSB to cycle stably for over 200 cycles (Fig. 5g) [23]. Together, the results in Figs. 2 - 4 show that combining polymer coordination chemistry with ceramic percolation networks can synergistically enhance ionic conductivity, mechanical integrity, and interfacial stability in SSLSBs. The most advanced electrolyte architectures integrate selective ion transport, chemical confinement, and mechanical adaptability within a single composite framework. Asymmetric interpenetrating polymer networks coordinate polysulfides on the cathode side through carbonyl-rich PVA-CN domains while promoting uniform Li deposition via softer PDXL segments adjacent to the anode. These features produce ionic conductivities of 5.25×10^{-3} S cm^{-1} and Li^+ transference numbers approaching 0.81 (Fig. 6f-g), along with strong suppression of polysulfide absorbance (Fig. 6a) and smooth lithium

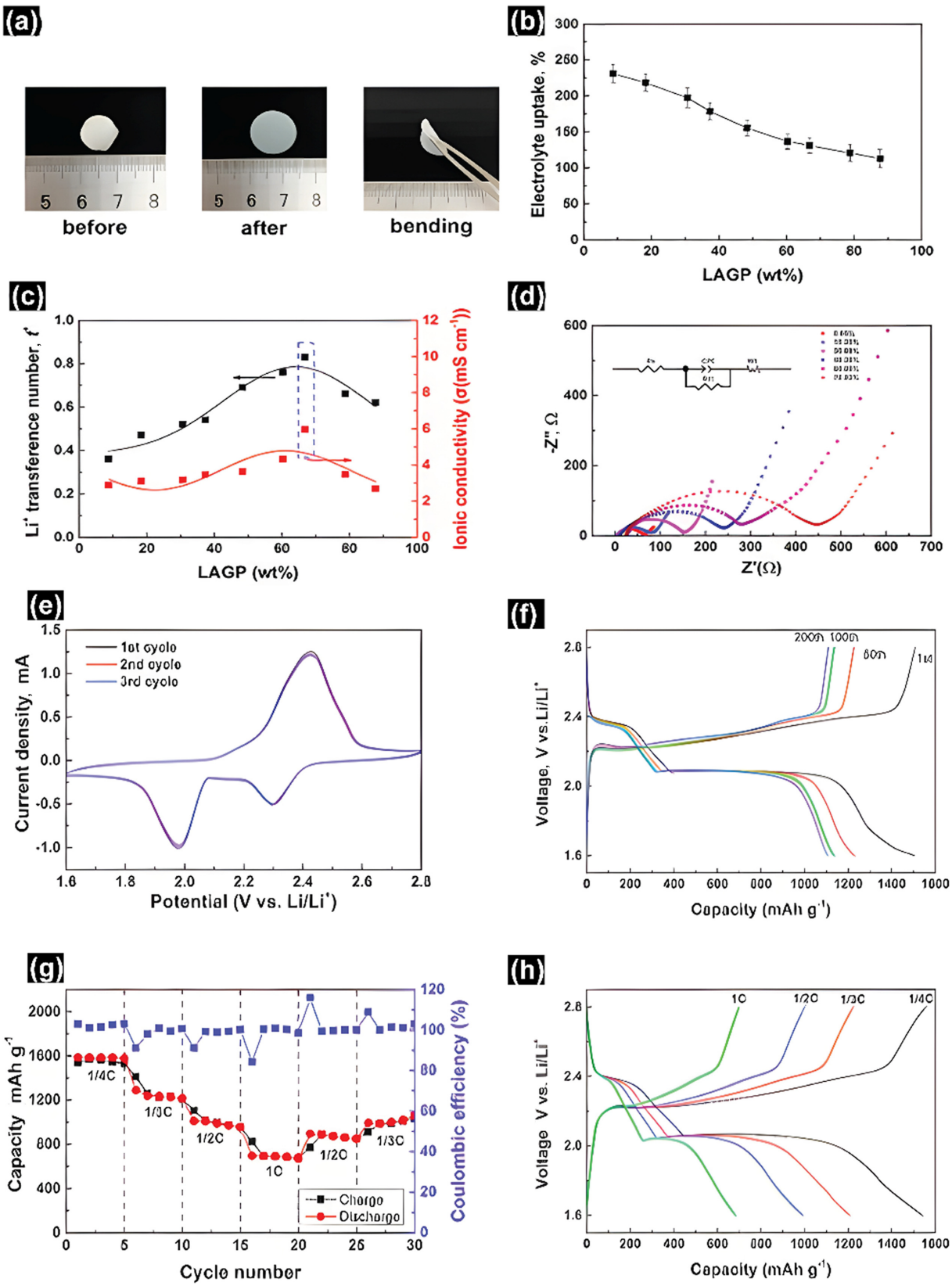

Fig. 4. Physicochemical and electrochemical properties of a quasi-solid composite electrolyte (QSSCE) with LAGP filler. (a, b) Flexibility and liquid uptake. (c, d) Ionic conductivity, transference number, and EIS. (e-h) CV, galvanostatic cycling, and rate performance data [22]Click or tap here to enter text.

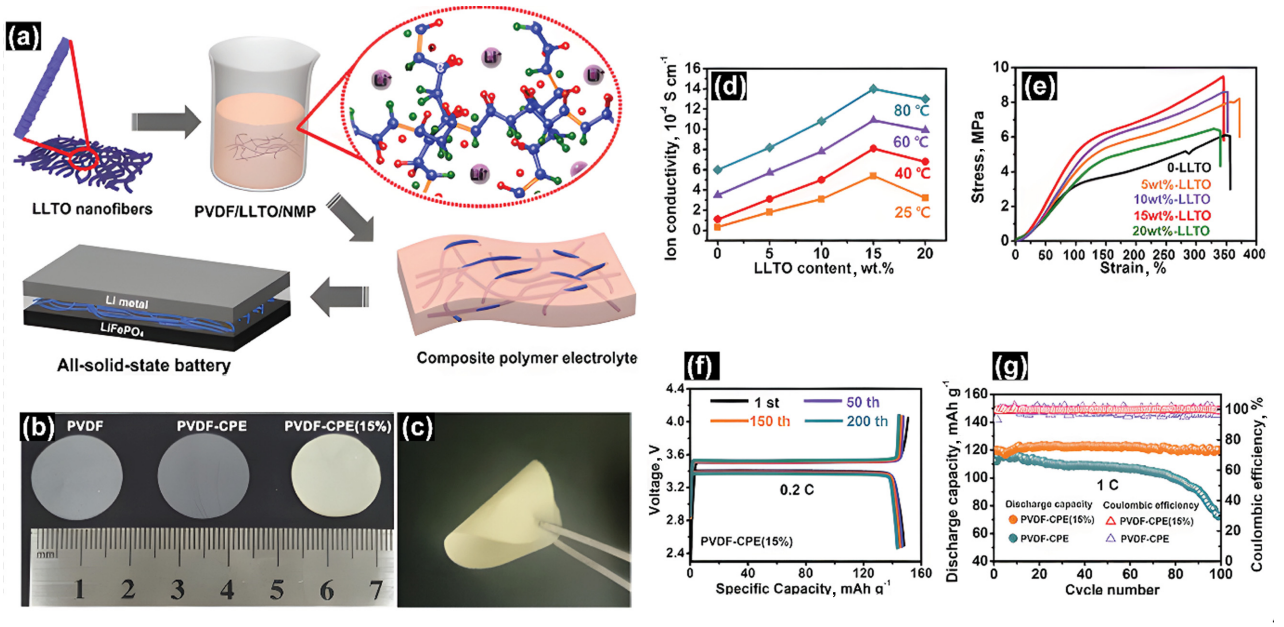

Fig. 5. LLTO-nanofiber-reinforced PVDF-based CPE: (a-c) Synthesis and flexibility, (d, e) Conductivity and mechanical behavior, (f, g) Electrochemical stability and cycling in $LiFePO_4$||Li cells. Reprinted with permission from ref. [23]Click or tap here to enter text. Copyright 2019, American Chemical Society

morphologies after cycling (Fig. 6b-e). Resulting SSLSBs achieve ~807 mAh g^{-1} after 500 cycles at 0.5 C and excellent rate performance, as shown in Fig. 6h-j [24]. Polymer-in-salt matrices further push the limits by delivering 1.67×10^{-3} S cm^{-1} conductivity at room temperature (Fig. 7a) and exceptional electrochemical stability up to 5.5 V (Fig. 7b). They maintain ~1050-1100 mAh g^{-1} in early cycling and ~720 mAh g^{-1} after 300 cycles (Fig. 7e - h), while exhibiting minimal self-discharge (~0.2% over 14 days; Fig. 7h). The two-peak CV signature near ~2.3 and ~2.0 V persists in these systems (Fig. 7q), confirming that multi-step sulfur conversion can be preserved under quasi-solid confinement with significantly reduced shuttle activity [25].

Despite differences in design, these advanced architectures all achieve higher ionic conductivity, better interfacial stability, effective polysulfide regulation, and improved long-term cycling. This clearly demonstrates that well-engineered CPEs can bridge the gap between the high utilization offered by liquid electrolytes and the superior stability of true solid electrolytes. Accordingly, CPEs have emerged as one of the most versatile and scalable electrolyte platforms for enabling durable, high-energy SSLSBs. To further put the above results in context, Table 1 summarizes various CPE formulations, highlighting how different active vs. passive fillers impact ionic conductivity, cell polarization (overpotential), and cycling performance in SSLSBs.

5. Future Directions

Composite polymer electrolytes have made SSLSBs increasingly viable, but several key limitations still prevent practical operation. Achieving high room-temperature ionic conductivity while preserving mechanical strength remains difficult, since methods such as increasing polymer amorphicity, adding plasticizers, or using high filler loadings often introduce brittleness, phase discontinuities, or unstable polymer-filler interfaces. These defects reduce dendrite suppression and can lead to non-uniform Li plating during long-term cycling. Ensuring continuous electrolyte contact with the sulfur cathode also remains challenging due to poor wetting, mechanical mismatch, and the large volume changes associated with the S to Li_2S conversion, which can cause delamination and increasing interfacial resistance. Scalability is another major concern. Many CPEs rely on fragile, moisture-sensitive membranes or synthesis schemes incompatible with roll-to-roll manufacturing [8]. Meanwhile, the thickness and stack-pressure requirements of solid

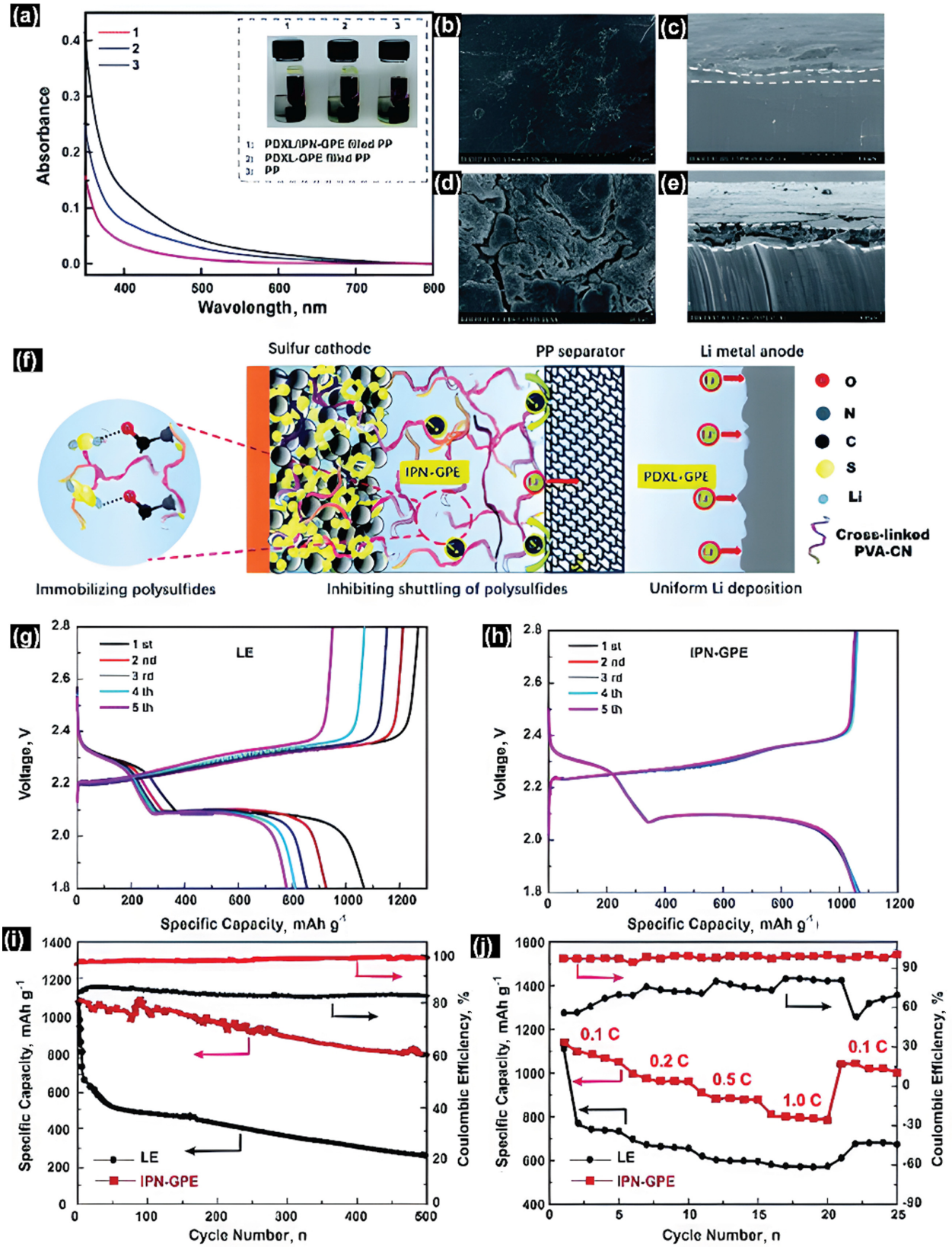

Fig. 6. Asymmetric interpenetrating network gel polymer electrolyte (IPN-GPE): (a) Polysulfide diffusion suppression, (b-e) Li morphology after cycling, (f) Schematic structure, (g-j) Electrochemical performance in Li–S cells. Reprinted with permission from ref. [24]Click or tap here to enter text. Copyright 2021, Royal Society of Chemistry

Table 1. Active and Passive fillers in CPEs for SSLSBs

Fillers	Polymer matrix	Conductivity σ (S cm^{-1})	Overpotential (V)	Initial capacity (mAh g^{-1})	Capacity retention (%)/ cycle numbers	C rate	Ref
Al_2O_3	PEO/LiTFSI	1.5×10^{-5} (30 °C)	1.4–2.8	1050	63/4 cycles (25 °C)	0.05	[27]
SiO_2	PEO/LiTFSI	7.15×10^{-5} (37 °C)	1.8–2.6	1316	45.5/40 cycles (50 °C)	0.1	[28]
TiO_2	PEO/LiTFSI	1.3×10^{-4} (37 °C)	1.8–2.6	1582	61.9/40 cycles (50 °C)	0.1	[28]
LGPS	PEO/LiTFSI	0.42×10^{-3} (20 °C)	1.0–2.5	1772	33.1/50 cycles (60 °C)	0.1	[29]
LLZTO	PEO/LiTFSI	1.58×10^{-4} (50 °C)	1.7–2.8	351.7	94.1/300 cycles (50 °C)	0.5	[30]
LLTO	PEO/LiTFSI	1.49×10^{-4} (30 °C)	1.7–2.8	1234.6	73.5/100 cycles (20 °C)	0.05	[31]

*Ionic conductivities are from bulk electrolyte measurements except Al_2O_3/PEO (in assembled Li–S cells)

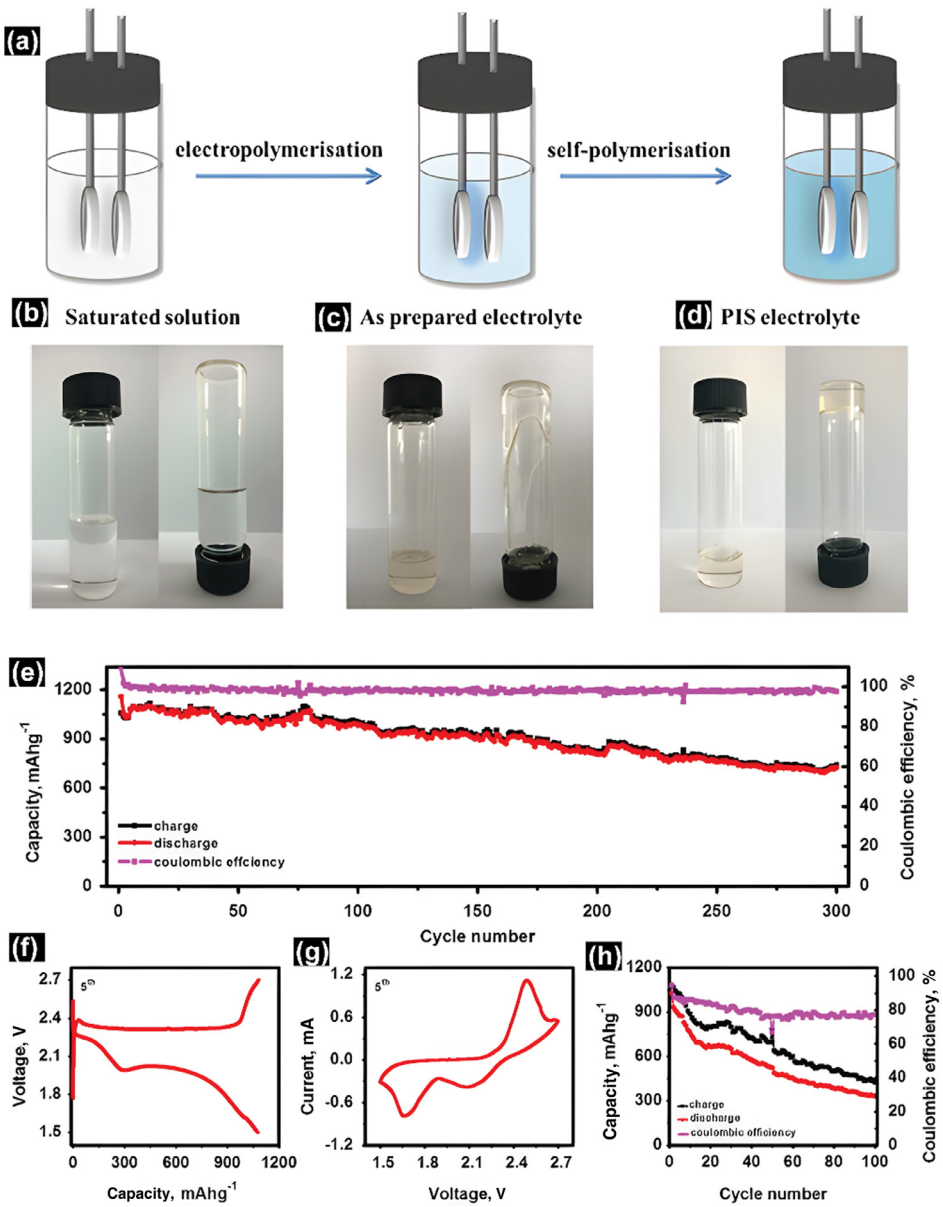

Fig. 7. Fabrication and electrochemical performance of a polymer-in-salt (PIS) electrolyte (a-d) Synthesis steps. (e-h) Cycling stability, voltage profiles, CV, and long-term behavior in Li–S cells [25]

electrolytes reduce practical energy density, highlighting the need for thin (<50 μm), robust membranes paired with high-loading sulfur cathodes. Future progress will rely on several emerging directions: single-ion conducting polymer matrices to raise t_{Li^+}, advanced fillers (sulfides, halides, MOFs, COFs) to introduce fast Li^+ pathways and strong polysulfide affinity, and multilayer or graded architectures that place ceramic-rich layers near the Li anode and polymer-rich, polysulfidephilic regions near the sulfur cathode [26]. Integrated electrolyte-electrode concepts, such as ion-conducting binders or polymeric sulfur hosts, can further reduce interfacial resistance by creating continuous ion-transport networks. At the same time, scalable fabrication strategies including UV-curing, hot-pressing, co-extrusion, and 3D printing will be essential for producing thin, defect-free membranes [13]. Combined with computational tools for rational materials design, these approaches outline a realistic pathway toward CPEs that deliver high conductivity, stable Li-metal cycling, strong polysulfide confinement, and manufacturability suitable for next-generation SSLSBs.

6. Conclusions

Composite polymer electrolytes offer a promising pathway toward practical SSLSBs by combining the safety and stability of inorganic solids with the flexibility and interface compatibility of polymers. Through tailored polymer-filler interactions, CPEs effectively suppress polysulfide migration, enhance Li^+ transport, and improve interfacial stability at both the sulfur cathode and lithium-metal anode. Recent developments have demonstrated markedly improved cycle life, high Coulombic efficiency, and reduced dendrite formation, highlighting the strong potential of these hybrid electrolytes. Despite this progress, challenges remain in achieving high room-temperature conductivity, maintaining stable Li-metal interfaces, and scaling up membrane fabrication. However, emerging solutions, including single-ion conducting polymers, superionic fillers, multilayer composite structures, and integrated electrode-electrolyte designs provide clear pathways for further improvement. Overall, continued advancements in materials design and scalable processing are expected to bring CPE-enabled SSLSBs closer to practical deployment, positioning them as strong candidates for high-energy, safe, and sustainable energy storage.

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