A Comprehensive Review on Polymer Electrolytes for Advanced Electrochemical Energy Storage: Materials, Conductivity, and Emerging Applications

K. N. Sathish[1], J. Gurusiddappa[2], Vinay Prasad V[3], Madesh Kumar M.[4,5]**, Prashant Bhat[6], Pawan Kumar[7], Swathi Harithas[8]*

[1]*Govt. First Grade College, Doddaballapura 561203, Karnataka, India*
[2] *Srinivasa Ramanujan Institute of Technology, Anantapuramu, Andhra Pradesh 515701, India*
[3]*Department of Physics, Cambridge Institute of Technology (An Autonomous Institution), K. R. Puram, Bangalore-560036, India*
[4]*Department of Physics, School of Applied Sciences, REVA University, Rukmini Knowledge Park, Yelahanka, Kattigenahalli, Bengaluru, Sathanur, Karnataka, 560064, India*
[5]*Center for Advanced Materials and Research in Physics, REVA Research Center, REVA University, Bengaluru, Karnataka 560064, India*
[6]*Department of Physics, Ramaiah Institute of Science and Management Foundation 6th & 7th floor, Wing B, North Gate, Yelahanka Hobli, Bangalore North, Bangalore – 560064, Karnataka, India.*
[7]*Nitte (Deemed to be University), NMAM Institute of Technology (NMAMIT) Nitte, Department of Physics, Nanoscience Research Laboratory, Karkala - 574110, Karnataka, India*
[8]*Department of chemistry, AES National Degree College, Gauribidanur, Karnataka - 561208 INDIA*
Corresponding Authors: *harithasswathi@gmail.com, madeshkumar@reva.edu.in*

Abstract

The increasing demand for portable electronics, electric vehicles, and renewable energy systems has accelerated the development of advanced energy storage technologies with improved safety, efficiency, and sustainability. Among the key components of electrochemical energy storage devices, electrolytes play a crucial role in governing ionic transport, electrochemical stability, cycle life, and overall device performance. Conventional liquid electrolytes, although widely used in lithium-ion batteries, suffer from limitations such as leakage, flammability, and thermal instability. Polymer electrolytes have emerged as promising alternatives due to their enhanced safety, flexibility, and compatibility with solid-state energy storage systems. This review provides a comprehensive overview of solid polymer electrolytes (SPEs), gel polymer electrolytes (GPEs), and composite/hybrid polymer electrolytes (CPEs), highlighting their structures, ion transport mechanisms, advantages, and limitations. Various fabrication techniques, including solution casting, electrospinning, in-situ polymerization, and phase separation, are discussed. Particular emphasis is placed on conductivity enhancement strategies involving plasticizers, ionic liquids, nanofillers, and interfacial engineering. Key performance parameters such as ionic conductivity, cation transference number, electrochemical stability window, and mechanical and thermal stability are critically evaluated. Recent developments in biopolymer-based electrolytes and their applications in lithium-, sodium-, and zinc-ion batteries, as well as supercapacitors, are also examined. Finally, current challenges and future perspectives for developing high-performance, sustainable polymer electrolytes for next-generation energy storage devices are discussed.

Keywords: Polymer electrolytes; Ionic conductivity; Solid-state batteries; Energy storage devices; Composite electrolytes; Biopolymer electrolytes

1. Introduction

The growing global demand for portable electronics, electric vehicles, and large-scale renewable energy integration has intensified the need for efficient and reliable electrical energy storage systems. Among the various energy storage technologies, rechargeable batteries have emerged as indispensable components due to their high energy efficiency, long cycle life, and operational flexibility. To meet these requirements, a wide range of energy storage systems has been explored. Conventional organic liquid electrolytes remain

the most widely used due to their high ionic conductivity and good electrochemical performance. Alternative electrolyte systems, including aqueous, ionic liquid, solid-state, and gel polymer electrolytes, have therefore attracted significant attention. Solid-state electrolytes offer improved safety and thermal stability, whereas gel polymer electrolytes provide a balance between the high conductivity of liquid electrolytes and the mechanical strength of solid materials. The implementation renewable energy sources, such as solar and wind power, further highlights the importance of advanced battery technologies capable of storing and delivering energy on demand [1].

Higher cell voltages are generally associated with greater energy storage capabilities, making the selection of electrode materials with favorable redox potentials a critical aspect of battery design. In this context, lithium has attracted significant attention because of its extremely low standard reduction potential (−3.04 V versus the standard hydrogen electrode), low atomic weight, and high theoretical specific capacity. These unique properties enable lithium-based batteries to achieve higher energy densities than most conventional battery chemistries. Lithium-ion batteries (LIBs) have therefore become the dominant technology for modern energy storage applications [2]. Furthermore, understanding the distribution of electrochemical potential and minimizing interfacial resistance are essential for reducing voltage losses and improving power output. Consequently, continuous research efforts are focused on developing high-voltage electrode materials and advanced battery chemistries, including lithium redox-flow systems, to meet the growing demands of next-generation energy storage technologies [1,2]. Lithium-ion batteries (LIBs) have dominated the energy storage market for decades owing to their high energy density, long cycle life, and excellent electrochemical performance. Their widespread use in portable electronics, electric vehicles, and stationary energy storage systems has significantly contributed to modern technological advancements.

However, concerns regarding the limited availability of lithium resources, rising material costs, safety issues, and environmental sustainability have motivated the search for alternative battery chemistries capable of meeting future energy demands [4]. Among the emerging post-lithium technologies, sodium-ion and zinc-ion batteries have attracted considerable attention due to the natural abundance, low cost, and environmental friendliness of sodium and zinc. More recently, hybrid sodium zinc ion batteries have emerged as a promising energy storage platform that combines the advantages of both systems. By utilizing sodium-ion intercalation chemistry alongside zinc metal electrochemistry, these hybrid batteries offer improved cycling stability, enhanced safety, and reduced material costs compared with conventional lithium-ion batteries [3].

Advances in electrode materials, electrolyte design, and dual-ion transport mechanisms have further improved the energy density, rate capability, and long-term performance of sodium zinc systems. In particular, the use of Prussian blue analogue cathodes and sodium-rich electrolytes has shown potential in mitigating challenges such as zinc dendrite formation and active material degradation. Although issues related to interface stability and large-scale manufacturing remain, sodium zinc batteries are increasingly recognized as promising candidates for sustainable and cost-effective next-generation energy storage applications [3,4]. The increasing use of portable electronic devices, electric vehicles, and renewable energy technologies has significantly increased the demand for efficient, reliable, and sustainable energy storage systems. Among the various energy storage technologies available today, rechargeable batteries have emerged as one of the most practical and widely used options due to their high efficiency, long service life, and adaptability across diverse applications.

Most importantly, the electrolyte plays a pivotal role in determining its overall performance, safety, energy density, and operational lifespan. Acting as the medium for ion transport between the electrodes, the electrolyte directly influences charge-transfer kinetics, electrode stability, and the efficiency of energy storage processes. Consequently, the selection and design of suitable electrolyte systems have become central to the development of advanced batteries and supercapacitors. In lithium-based batteries, the electrolyte is particularly important as it governs the formation and stability of the electrode electrolyte interphase, which significantly affects cycling performance and safety. The demand for higher energy densities has driven the adoption of high-capacity anodes, such as lithium metal and silicon-based materials, as well as high-voltage cathodes. However, these advanced electrode materials often operate beyond the stability limits of conventional electrolytes, leading to undesirable side reactions, capacity fading, and reduced cycle life.

To address these challenges, extensive efforts have been devoted to developing advanced electrolyte formulations and multifunctional additives capable of stabilizing electrode interfaces, suppressing degradation reactions, and enhancing battery longevity. The importance of electrolytes is equally evident in supercapacitors, where electrolyte properties strongly influence the operating voltage window and, consequently, the achievable energy density. Optimized electrolyte electrode combinations can significantly improve ion transport, capacitance, and power delivery while maintaining excellent cycling

stability. Therefore, electrolyte engineering has emerged as a key strategy for enhancing the performance of next-generation energy storage devices. Continued advances in electrolyte chemistry are expected to play a crucial role in achieving safer, higher-energy, and longer-lasting electrochemical systems suitable for large-scale commercial applications [5,6].

Biopolymer electrolytes have emerged as a sustainable and environmentally benign alternative to conventional petroleum-derived polymer electrolytes for next-generation energy storage devices. Derived from abundant renewable resources, biopolymers such as cellulose, chitosan, pectin, alginate, starch, gelatin, carrageenan, agar, xanthan gum, guar gum, and sodium carboxymethyl cellulose (CMC) possess abundant polar functional groups, includinghydroxyl (OH), carboxyl (COOH), carbonyl (C=O), amino (NH□), ether (C O C), and sulfate(SO□H) moieties. These functional groups facilitate effective coordination with charge carriers, promote salt dissociation, and enable ion transport through the polymer matrix. Owing to their biodegradability, biocompatibility, low toxicity, abundance, and excellent film-forming ability, biopolymer-based solid polymer electrolytes (SPEs) are particularly attractive for developing safe and eco-friendly energy storage technologies. Furthermore, their intrinsic flexibility and non-toxic nature make them suitable for wearable, flexible, implantable, and disposable electronic devices. Nevertheless, their widespread application is hindered by relatively low ionic conductivity, moisture sensitivity, poor mechanical strength, limited electrochemical stability, and interfacial degradation during prolonged cycling. To overcome these challenges, current research focuses on polymer blending, plasticization, inorganic filler incorporation, crosslinking, and interface engineering to enhance electrochemical performance. Biocompatible SPEs have shown significant potential for lithium-ion, sodium-ion, zinc-ion, magnesium-ion, proton, and biodegradable batteries, where safety, environmental sustainability, and long-term electrochemical stability are equally important.

Despite considerable progress, challenges related to interfacial resistance, ionic transport, and long-term stability remain. During battery operation, reactions between the lithium metal anode and the electrolyte lead to the formation of a thin passivation layer known as the solid electrolyte interphase (SEI). Ideally, the SEI should be chemically stable, electronically insulating, and ionically conductive, allowing lithium ions to pass while preventing further electrolyte decomposition. Consequently, the development of multifunctional and composite electrolytes with optimized conductivity, mechanical integrity, and interfacial compatibility is considered essential for realizing safer, longer-lasting, and high-performance next-generation energy-storage systems [7].

This review aims to provide a comprehensive overview of biopolymer-based electrolytes and their emerging role in advanced energy storage devices. Particular emphasis is placed on the use of naturally derived polymers as sustainable, environmentally friendly, and safer alternatives to conventional synthetic electrolyte materials. The review discusses the fundamental properties of biopolymers, their ion-transport mechanisms, and recent strategies

employed to enhance ionic conductivity, mechanical strength, thermal stability, and electrochemical performance. Furthermore, the challenges, opportunities, and future prospects of biopolymer electrolytes in lithium-ion batteries, solid-state batteries, supercapacitors, and other next-generation energy storage systems are critically examined.

2. Classification of Polymer Electrolytes

2.1 Solid Polymer Electrolytes (SPEs)

Solid polymer electrolytes (SPEs) are solvent-free electrolyte systems composed of a polymer host matrix and dissolved metal salts, where ion transport occurs through the solid phase without the presence of volatile liquid solvents. Common polymer hosts include poly(ethylene oxide) (PEO), polyacrylonitrile (PAN), poly(methyl methacrylate) (PMMA), and poly(vinyl chloride) (PVC), which possess functional groups capable of coordinating with metal ions. In these systems, the polymer matrix provides structural integrity and mechanical flexibility, while the incorporated salt supplies the mobile ionic species required for electrochemical conduction.

Unlike conventional liquid electrolytes, SPEs eliminate concerns related to solvent leakage, evaporation, and flammability, making them attractive candidates for next-generation energy-storage technologies. Ionic transport in SPEs generally occurs through segmental motion of polymer chains and ion hopping within amorphous regions of the polymer network. Consequently, the electrochemical performance of SPEs is strongly influenced by factors such as polymer crystallinity, chain mobility, salt dissociation, dielectric properties, and polymer ion interactions. Recent developments have focused on tailoring these parameters through polymer blending, nanofiller incorporation, plasticization, and structural modifications to enhance ionic conductivity and overall device performance.

Solid polymer electrolytes offer several advantages that have positioned them as promising alternatives to conventional liquid electrolytes in modern energy-storage systems. One of their most significant benefits is enhanced safety. Since SPEs are free from volatile organic solvents, they exhibit reduced flammability, negligible leakage, and improved thermal stability, thereby minimizing the risks of fire, explosion, and thermal runaway. These characteristics are particularly important for high-energy-density lithium-ion and lithium-metal batteries intended for electric vehicles and large-scale energy-storage applications.

Another important advantage is their excellent mechanical flexibility and processability. Polymer matrices can be fabricated into thin films, membranes, and flexible structures, making them suitable for wearable electronics, portable devices, and emerging flexible energy-storage systems. Furthermore, the solid-state nature of SPEs provides a physical barrier against lithium dendrite penetration, which helps improve battery safety and prolong cycle life.

The properties of SPEs can also be tailored through material engineering approaches such as polymer blending, salt optimization, and nanocomposite formation. The incorporation of ceramic nanoparticles, metal oxides, carbon nanomaterials, and quantum dots can enhance ionic conductivity, mechanical strength, thermal resistance, and electrochemical stability. Such modifications increase the amorphous content of the polymer matrix and create efficient pathways for ion migration. In addition, SPEs are compatible with a broad range of electrochemical devices, including lithium-ion batteries, sodium-ion batteries, fuel cells, supercapacitors, and flexible electronic systems. Their lightweight nature, environmental compatibility, and multifunctional characteristics make them attractive materials for future sustainable energy technologies.

Despite their numerous advantages, several challenges continue to hinder the large-scale commercialization of solid polymer electrolytes. The most significant limitation is their relatively low ionic conductivity at ambient temperature compared with conventional liquid electrolytes. In many polymer systems, ion transport is closely associated with polymer chain mobility, which decreases substantially in highly crystalline regions. As a result, achieving high ionic conductivity while maintaining mechanical integrity remains a major challenge.

Another critical issue involves the electrode electrolyte interface. Poor interfacial contact between the solid electrolyte and electrode surfaces can lead to increased interfacial resistance, reduced charge-transfer efficiency, and limited electrochemical performance. Furthermore, long-term electrochemical stability remains a concern, particularly when SPEs are paired with high-voltage cathodes or lithium-metal anodes. Interfacial degradation, side reactions, and insufficient compatibility may adversely affect cycle life and battery efficiency [11].

Although SPEs can suppress dendrite growth more effectively than liquid electrolytes, complete elimination of lithium dendrites remains difficult under high current densities and prolonged cycling conditions. Additional challenges include low lithium-ion transference numbers, restricted electrochemical stability windows, and difficulties associated with large-scale manufacturing. Therefore, further improvements in polymer design, interfacial engineering, and composite electrolyte development are required before SPEs can fully satisfy the performance demands of next-generation batteries [12].

The growing demand for safer, high-performance, and environmentally sustainable energy-storage systems has significantly expanded the scope of solid polymer electrolytes. Recent advances in polymer chemistry, nanocomposite engineering, and electrolyte optimization have led to substantial improvements in ionic conductivity, thermal stability, and electrochemical performance. As a result, SPEs are increasingly being explored for applications in solid-state lithium-ion batteries, lithium-metal batteries, sodium-ion batteries, supercapacitors, and flexible electronic devices.

Current research is particularly focused on incorporating ionic liquids, functional nanofillers, and advanced polymer architectures to overcome conductivity limitations while maintaining safety advantages. Among these approaches, ionic-liquid-based polymer electrolytes have emerged as promising candidates because they combine the non-flammable nature of ionic liquids with the mechanical stability of polymer matrices. Such systems offer enhanced ionic transport, wider electrochemical stability windows, and improved environmental sustainability [13].

Although SPE technology has not yet achieved widespread commercial deployment in high-energy-density battery systems, several prototype and pre-commercial solid-state battery platforms already utilize polymer-based electrolytes. Continued progress in interfacial stabilization, ion-conduction mechanisms, and scalable manufacturing processes is expected to accelerate their adoption. Given their unique combination of safety, flexibility, lightweight design, and environmental compatibility, SPEs are widely regarded as one of the most promising electrolyte technologies for next-generation energy-storage devices and sustainable energy applications [13].

2.2 Gel Polymer Electrolytes (GPEs) and Hydrogels

Gel polymer electrolytes (GPEs) represent an intermediate class of electrolyte materials that combine the advantages of liquid electrolytes and solid polymer electrolytes. In general, GPEs consist of a three-dimensional polymer network swollen with a liquid electrolyte, solvent, ionic liquid, or plasticizer. Unlike solid polymer electrolytes, where ion transport occurs solely through the polymer matrix, GPEs retain a significant amount of liquid phase within the polymer framework, resulting in enhanced ionic conductivity while maintaining the mechanical integrity of a solid material. Hydrogels, organogels, and ionogels are among the most widely investigated subclasses of gel electrolytes for electrochemical energy-storage applications [14].

The growing demand for flexible electronics, wearable devices, electric vehicles, and advanced energy-storage systems has stimulated extensive research into GPEs. Their unique structure enables the fabrication of deformable and mechanically robust batteries and supercapacitors capable of operating under bending, stretching, and compression conditions. Furthermore, the immobilization of liquid electrolytes within a polymer matrix minimizes electrolyte leakage while preserving high ionic conductivity. Compared with conventional liquid electrolytes, GPEs exhibit improved safety, enhanced dimensional stability, and reduced solvent evaporation. At the same time, they overcome many of the interfacial limitations commonly encountered in all-solid-state electrolyte systems [15]. The performance of GPEs depends strongly on the composition of the polymer network, the type of solvent or plasticizer employed, the concentration of lithium salts, and the nature of polymer electrolyte interactions. Recent studies have focused on tailoring these parameters to achieve high ionic conductivity, broad electrochemical stability windows, enhanced mechanical strength, and extended operating temperature ranges. Consequently, GPEs have emerged as highly promising electrolyte materials for next-generation rechargeable batteries and supercapacitors [14,15].

One of the most significant advantages of GPEs is their superior ionic conductivity. Because a liquid phase is retained within the polymer matrix, ion transport occurs more efficiently than in conventional solid polymer electrolytes, where ionic movement is often restricted by polymer chain mobility and crystallinity. As a result, GPEs typically exhibit ionic conductivities approaching those of liquid electrolytes while still retaining many of the safety benefits associated with solid-state systems [16]. Another important advantage is their excellent interfacial compatibility with electrode materials. In solid-state batteries, poor contact between rigid electrolytes and electrode surfaces often leads to increased interfacial resistance and sluggish charge-transfer kinetics. In contrast, the soft and flexible nature of GPEs allows intimate contact with electrode surfaces, thereby reducing interfacial impedance and facilitating efficient ion transport across the electrode electrolyte interface. This characteristic is particularly beneficial in lithium-metal batteries, where uniform lithium-ion flux is essential for stable electrochemical performance [16]. GPEs also play an important role in regulating the formation and stability of the solid electrolyte interphase (SEI). A stable SEI layer is crucial for preventing continuous electrolyte decomposition and ensuring long-term cycling stability. The composition of the polymer matrix, plasticizers, additives, and incorporated nanomaterials can significantly influence SEI formation and morphology. Properly engineered GPE systems promote the formation of uniform and robust SEI layers that help suppress lithium dendrite growth and improve battery safety [16].

Recent developments have further enhanced GPE performance through the incorporation of advanced functional materials such as metal organic frameworks (MOFs), ceramic nanoparticles, carbon nanostructures, and ionic liquids. These additives improve ionic conductivity, mechanical strength, electrochemical stability, and thermal resistance. For example, MOF-based GPE systems provide interconnected ion-conduction pathways and facilitate rapid lithium-ion transport while simultaneously improving structural stability. Such innovations have enabled GPEs to support high-voltage cathode materials and achieve improved cycling performance in practical battery configurations [17]. Compared with SPEs, GPEs generally offer better room-temperature conductivity, lower interfacial resistance, wider applicability in flexible devices, and improved electrochemical performance. These advantages have made them one of the most actively investigated electrolyte classes for advanced lithium-ion and lithium-metal battery technologies. The schematic illustration of GPE and hydrogels is depicted in Fig.1.

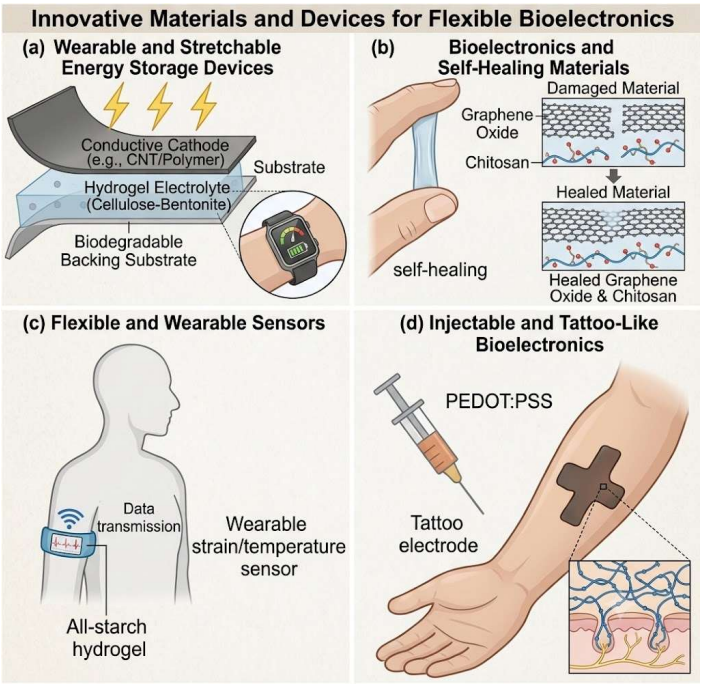

Figure 1. Gel Polymer Electrolytes (GPEs) and Hydrogels [Source information: (Generated By AI Gemini)

Despite their numerous advantages, several challenges continue to limit the widespread commercialization of gel polymer electrolytes. One of the primary concerns is the presence of liquid components within the gel structure. Although the liquid phase is immobilized by the polymer network, solvent evaporation may still occur during prolonged operation or under elevated temperatures. This can lead to reduced ionic conductivity, mechanical degradation, and diminished battery performance over time [18]. Safety concerns also remain an important consideration. Many GPEs still contain organic solvents that may exhibit flammability or chemical instability under extreme operating conditions. While the risk is generally lower than that associated with conventional liquid electrolytes, achieving completely non-flammable and environmentally benign GPE systems remains a major research objective [18].

Another challenge involves balancing mechanical strength and ionic conductivity. Increasing polymer content often improves structural integrity but can simultaneously reduce ion mobility. Conversely, increasing solvent content enhances conductivity but may compromise mechanical stability and dimensional robustness. Achieving an optimal balance between these competing factors is essential for practical applications. Long-term electrochemical stability is another critical issue. Continuous cycling can induce changes in polymer structure, electrolyte distribution, and interfacial chemistry, leading to capacity fading and reduced cycle life. Furthermore, although GPEs can suppress lithium dendrite formation more effectively than liquid electrolytes, complete dendrite elimination remains difficult under high current densities and prolonged cycling conditions [16,18].

Manufacturing complexity and production costs also pose challenges for large-scale commercialization. The preparation of advanced GPE systems often requires carefully controlled synthesis procedures, specialized materials, and precise optimization of polymer electrolyte compositions. Therefore, future research efforts should focus on developing low-cost, scalable, environmentally friendly, and high-performance GPE systems that combine excellent ionic conductivity, mechanical robustness, safety, and long-term stability. Addressing these challenges will be essential for realizing the full potential of gel polymer electrolytes in next-generation energy-storage technologies [18].

2.3 Composite and Hybrid Polymer Electrolytes (CPEs)

Composite polymer electrolytes (CPEs) are advanced electrolyte systems designed by integrating polymer matrices with inorganic fillers, ceramic particles, nanomaterials, ionic liquids, or other functional components to overcome the limitations of conventional polymer electrolytes. These materials combine the flexibility and processability of polymers with the superior ionic conductivity, mechanical strength, and thermal stability of inorganic materials. As a result, CPEs have emerged as promising candidates for next-generation solid-state batteries and other electrochemical energy-storage devices [19].

In a typical CPE, polymers such as poly(ethylene oxide) (PEO), polyacrylonitrile (PAN), poly(vinylidene fluoride) (PVDF), and their blends serve as the host matrix, while inorganic fillers including

metal oxides, ceramic nanoparticles, and lithium-ion-conducting compounds are incorporated to improve electrochemical performance. The addition of these fillers modifies the polymer microstructure, suppresses crystallinity, enhances amorphous regions, and creates additional pathways for ion migration. Consequently, CPEs often exhibit higher ionic conductivity and improved mechanical properties compared with conventional solid polymer electrolytes [21,22]. Closely related to CPEs are hybrid polymer electrolytes, which combine multiple electrolyte systems, such as aqueous, organic, ionic-liquid, gel, or polymer-based electrolytes, within a single framework. The primary objective of hybrid electrolyte design is to exploit the complementary advantages of different electrolyte classes while minimizing their individual limitations. By carefully balancing conductivity, safety, electrochemical stability, and thermal performance, hybrid electrolytes provide a versatile platform for advanced energy-storage technologies [20].

The performance of both composite and hybrid polymer electrolytes depends strongly on the interactions occurring at the electrode electrolyte interface. Stable interfacial contact, efficient lithium-ion transport, and minimized side reactions are essential for achieving long cycle life and high energy efficiency. Therefore, considerable research has focused on understanding and engineering these interfaces to enhance battery performance and reliability [19]. The usage of CPEs in energy storage applications and practical battery applications is shown in Fig.2.

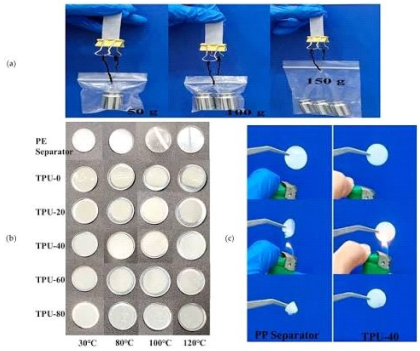

Figure 2. Composite and Hybrid Polymer Electrolytes (CPEs) [Souce Creative Commons Attribution (CC BY) license from MDPI https://doi.org/10.3390/polym16233280]

Compared with conventional solid polymer electrolytes (SPEs), composite and hybrid polymer electrolytes offer several important advantages. One of the most significant benefits is the enhancement of ionic conductivity. In SPEs, ion transport is often limited by polymer crystallinity and restricted chain mobility. The incorporation of inorganic fillers, ceramic particles, nanostructures, and ionic-liquid components disrupts polymer crystallization and facilitates the formation of continuous ion-conduction pathways, resulting in improved lithium-ion mobility [21].

Another major advantage is the reduction of interfacial resistance. Poor electrode electrolyte contact is one of the primary challenges associated with solid-state batteries. Composite electrolytes can form more stable and conductive interfaces through the presence of functional fillers that promote lithium-ion transport across the electrode electrolyte boundary. These fillers often assist in the formation of lithium-ion-conducting interphases, which significantly reduce charge-transfer resistance and improve electrochemical kinetics [21]. The incorporation of inorganic materials also improves the mechanical robustness and thermal stability of polymer electrolytes. Ceramic nanoparticles, metal oxides, and lithium-ion-conducting ceramics can reinforce the polymer matrix, increasing its resistance to deformation while maintaining flexibility. In addition, these materials often possess superior chemical and thermal stability, thereby improving the overall safety and durability of the electrolyte system [22].

Hybrid polymer electrolytes further expand the performance window by combining different electrolyte chemistries. For example, ionic liquid polymer hybrids can simultaneously provide high ionic conductivity, non-flammability, and wide electrochemical stability windows. Similarly, gel-based hybrid systems can offer enhanced interfacial contact and improved mechanical flexibility. Such multifunctional characteristics make CPEs particularly attractive for solid-state lithium-ion batteries, lithium-metal batteries, sodium-ion batteries, fuel cells, and supercapacitors [20].

An additional advantage of CPEs is their ability to suppress lithium dendrite growth. The enhanced mechanical strength and uniform ion-distribution characteristics of composite electrolytes help regulate lithium deposition and reduce the formation of localized current hotspots. Consequently, batteries employing CPEs often exhibit improved cycling stability, enhanced safety, and prolonged operational lifetimes compared with conventional SPE-based systems [21]. Despite their significant advantages, composite and hybrid polymer electrolytes still face several challenges that limit their widespread

commercialization. One of the primary concerns is the complexity of material design and fabrication. Achieving a uniform dispersion of inorganic fillers throughout the polymer matrix is often difficult, particularly at high filler loadings. Poor dispersion can lead to particle agglomeration, non-uniform ion transport, and reduced electrochemical performance [23]. Another challenge involves balancing ionic conductivity and mechanical strength. Although fillers can improve both properties, excessive filler concentrations may increase electrolyte rigidity and hinder ion transport. Similarly, some hybrid electrolyte formulations achieve excellent conductivity but suffer from increased viscosity, higher cost, or reduced long-term stability [20].

Interfacial instability remains another critical issue. Although CPEs generally provide better electrode contact than traditional SPEs, continuous side reactions at the electrode electrolyte interface can still occur during prolonged cycling. Such reactions may lead to the formation of unstable interphases, increased resistance, and gradual capacity degradation. Furthermore, achieving stable compatibility with high-voltage cathodes and lithium-metal anodes remains a major research challenge [19]. Manufacturing scalability also presents obstacles. Many high-performance CPE systems rely on sophisticated synthesis methods, specialized nanomaterials, and precisely controlled processing conditions, which can increase production costs and complicate large-scale fabrication. In addition, ensuring reproducibility and maintaining consistent performance across large-area electrolyte membranes remain important considerations for industrial implementation.

Although composite electrolytes demonstrate improved resistance to dendrite growth compared with conventional polymer electrolytes, complete suppression of dendrites under high current densities and long-term cycling has not yet been fully achieved. Therefore, continued efforts are required to optimize filler selection, interfacial engineering strategies, electrolyte architectures, and fabrication techniques [23]. Overall, composite and hybrid polymer electrolytes represent one of the most promising directions in electrolyte research. By combining the favorable characteristics of polymers, inorganic materials, and advanced electrolyte chemistries, these systems offer a practical pathway toward safer, high-energy-density, and high-performance electrochemical energy-storage technologies. Continued advances in materials design and interface engineering are expected to accelerate their transition from laboratory research to commercial applications.

3. Fundamentals of Ion Transport in Polymer Matrices

The performance of polymer electrolytes in electrochemical energy-storage devices is fundamentally governed by the transport of ions through the polymer matrix. Unlike liquid electrolytes, where ions move freely through a solvent medium, ion transport in polymer electrolytes occurs within a complex and dynamically evolving polymer network. Understanding the structural and molecular factors that control ionic motion is therefore essential for designing high-performance polymer electrolytes with enhanced conductivity, stability, and safety.[24] A polymer electrolyte consists of long-chain macromolecules interconnected through physical entanglements, chemical crosslinks, or intermolecular interactions, forming a three-dimensional network capable of hosting ionic species. At the microscopic level, this network resembles a maze-like arrangement of polymer chains containing interconnected regions of free volume through which ions can migrate. The polymer matrix not only provides mechanical support but also serves as the medium that

coordinates and transports ionic charge carriers. Consequently, ion transport depends strongly on the architecture of the polymer network, chain flexibility, free-volume distribution, and the interactions between ions and polymer segments [25].

The internal structure of polymer electrolytes is generally composed of crystalline and amorphous domains. Crystalline regions possess highly ordered molecular arrangements that restrict polymer-chain mobility and impede ion migration. In contrast, amorphous regions are characterized by disordered chain conformations and larger free-volume elements, which facilitate ionic movement. Because ionic conduction predominantly occurs within these amorphous domains, achieving an appropriate balance between crystallinity and disorder is a crucial design strategy for polymer electrolytes. Excessive crystallinity limits ionic conductivity, whereas highly amorphous structures may compromise mechanical strength and dimensional stability. Therefore, modern electrolyte design often aims to suppress crystallinity while preserving sufficient structural integrity [26].

The mobility of polymer chains is closely related to the glass transition temperature (Tg), a fundamental parameter that governs the transition between rigid and flexible polymer states. Below Tg, polymer chains remain frozen in a glassy state, significantly restricting segmental motion and ionic transport. Above Tg, increased chain mobility creates transient free-volume pathways that enable ions to move more readily through the polymer matrix. As a result, polymer electrolytes with lower glass transition temperatures generally exhibit higher ionic conductivity. The relationship between Tg and ion transport has

become an important design principle for next-generation polymer electrolytes, as it provides a direct link between polymer dynamics and electrochemical performance [27].

Another critical aspect of ion transport involves the coordination of cations with electronegative heteroatoms present within the polymer structure. Atoms such as oxygen, nitrogen, sulfur, and fluorine possess lone-pair electrons that can interact strongly with metal cations, particularly lithium ions. In polymer systems such as poly(ethylene oxide) (PEO), ether oxygen atoms act as coordination sites that solvate lithium ions and facilitate their movement through the polymer matrix. Similar coordination interactions can occur through carbonyl, nitrile, sulfonyl, and other functional groups incorporated into polymer backbones or side chains. The strength and reversibility of these interactions significantly

influence salt dissociation, ion mobility, and overall ionic conductivity. Effective polymer electrolyte design therefore requires an optimal balance between strong ion coordination for salt dissolution and sufficiently weak interactions to allow rapid ion transport [28,29].

The movement of ions through polymer electrolytes is commonly described by ion-hopping mechanisms assisted by polymer segmental motion. In conventional polymer electrolytes, cations migrate by repeatedly coordinating and de-coordinating from neighboring functional groups along the polymer chains. This process may occur within a single polymer chain (intrachain hopping) or between adjacent chains (interchain hopping). Because these hopping events are strongly coupled to local polymer motion, ion transport is often directly linked to segmental relaxation processes. As polymer chains fluctuate and rearrange, temporary pathways are created that enable ions to jump between coordination sites. Consequently, ionic conductivity generally increases with increasing chain flexibility and temperature [30,31].

The temperature dependence of ionic transport in polymer electrolytes can be described using two principal models: the Arrhenius model and the Vogel Tammann Fulcher (VTF) model. The Arrhenius model is typically applied to systems where ion transport is largely decoupled from polymer-chain motion. In such systems, ions migrate through pre-existing pathways or hopping sites, and the conductivity follows a thermally activated process characterized by a single activation energy. The Arrhenius relationship is expressed as:

$$\square = \square_0 \exp\left(\frac{-\square_\square}{\square\square}\right) \qquad (1)$$

where σ is the ionic conductivity, σ□ is the pre-exponential factor, Ea is the activation energy for ion transport, R is the universal gas constant, and T is the absolute temperature. Arrhenius-type behavior is commonly observed in highly disordered systems, inorganic solid electrolytes, and polymer electrolytes exhibiting decoupled ion-hopping transport mechanisms [32,33].

In contrast, many polymer electrolytes exhibit ion transport that remains strongly coupled to segmental chain dynamics. In these cases, ionic conductivity deviates from simple Arrhenius behavior and is more accurately described by the Vogel Tammann Fulcher (VTF) equation:

$$\square = \square \exp\left[-\frac{\square}{\square-\square_0}\right] \qquad (2)$$

where A is a pre-exponential constant, B is associated with the activation energy for segmental motion, and T□ is the Vogel temperature, typically related to the glass transition temperature of the polymer. The VTF model reflects the strong coupling between ion transport and polymer-chain relaxation processes. As temperature increases, enhanced segmental mobility facilitates ion migration, leading to the characteristic non-Arrhenius conductivity behavior observed in many polymer electrolyte systems [34,35].

Overall, ion transport in polymer electrolytes is governed by a complex interplay of polymer structure, morphology, chain dynamics, ion coordination chemistry, and temperature-dependent relaxation processes. A comprehensive understanding of these fundamental mechanisms is essential for the rational design of advanced polymer electrolytes capable of delivering high ionic conductivity, improved electrochemical stability, and long-term operational reliability in next-generation energy-storage devices.

4. Fabrication techniques for SPE

4.1 Solution casting technique

The solution casting technique is one of the most widely used and effective methods for the preparation of polymer electrolyte films for electrochemical energy storage applications due to its simplicity, low cost, uniform film formation, and ease of compositional control. This technique is particularly suitable for the fabrication of solid polymer electrolytes (SPEs), gel polymer electrolytes (GPEs), and polymer nanocomposite electrolytes used in batteries, supercapacitors, and electrochemical sensors. In this method, the required polymer host material is first dissolved in a suitable solvent such as distilled water,

tetrahydrofuran (THF), dimethyl sulfoxide (DMSO), N-methyl-2-pyrrolidone (NMP), acetonitrile, or ethanol under continuous stirring to obtain a homogeneous polymer solution. Subsequently, ionic salts, plasticizers, nanofillers, ionic liquids, or other functional additives are incorporated into the

polymer solution in desired proportions (usually in wt. %) to improve ionic conductivity, thermal stability, and electrochemical performance. The resulting mixture is stirred continuously using a magnetic stirrer or overhead stirrer for several hours to ensure complete dissolution and uniform dispersion of all components within the polymer matrix.

In many cases, ultrasonication is also employed to avoid agglomeration of nanoparticles and to improve homogeneity of the solution. Most of the studies employs the ultrasonication of nanoparticles in a desired solvent before adding it to the solution to have a better dispersion in unform mixture. Conversely, in some studies the ultrasonication is done after adding all the components. However, the former method of sonicating the nanoparticles is found to be have a uniform dispersion compared to the later. After achieving a clear and viscous homogeneous solution, the prepared mixture is cast onto a clean and inert substrate such as a glass plate, petri dish, Teflon mold, or polypropylene surface. The solvent present in the cast solution is then allowed to evaporate slowly under controlled conditions, either at room temperature or in a hot air oven maintained at moderate temperatures. Slow solvent evaporation is important for obtaining flexible, crack-free, and uniform polymer electrolyte films with good mechanical integrity. Once the solvent is completely removed, the dried polymer electrolyte film is carefully peeled off and stored in a moisture-free environment or vacuum desiccator prior to characterization and electrochemical testing.

The solution casting technique offers several advantages including ease of fabrication, excellent control over film thickness and composition, uniform distribution of salts and fillers, low processing temperature, and compatibility with a wide range of polymer systems. The method is highly adaptable for incorporating nanofillers, ceramic particles, ionic liquids, and conductive additives into the polymer matrix to tailor the electrochemical and mechanical properties of the electrolyte. However, the quality of the prepared films strongly depends on parameters such as solvent selection, stirring time, evaporation rate, casting conditions, and concentration of additives. Improper solvent removal or poor dispersion of fillers may lead to phase separation, pore formation, or reduced electrochemical performance. Despite these limitations, solution casting remains one of the most preferred fabrication techniques for polymer electrolytes because of its versatility and effectiveness in producing high-quality films for advanced energy storage applications. The schematic illustration of solution casting technique is as shown in Fig.3.

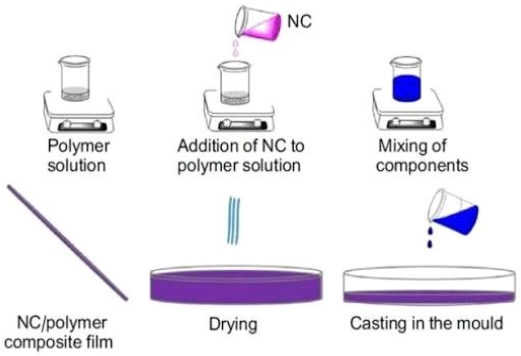

Figure 3. Solution casting technique [Source Information: Creative Common License from Advanced Research in Conservation Science 10.21608/arcs.2024.271433.1047]

4.2 Electrospinning

Electrospinning is an advanced and highly versatile fabrication technique widely employed for the preparation of solid polymer electrolytes (SPEs) with nanofibrous architectures for next-generation energy storage applications. The technique has attracted significant attention in lithium-ion, sodium-ion, zinc-ion, and solid-state battery research because it enables the production of ultrafine polymer fibers with high surface area, interconnected porous structures, and enhanced ion transport pathways. In the electrospinning process, a polymer solution or polymer salt composite solution is first prepared by dissolving the required polymer host, ionic salt, plasticizer, and functional additives in a suitable solvent system under continuous stirring until a homogeneous viscous solution is obtained. The prepared solution is then loaded into a syringe equipped with a metallic needle connected to a high-voltage power supply. During operation, a strong electric field is applied between the needle tip and a grounded collector. When

the applied electrostatic force overcomes the surface tension of the polymer solution, a charged jet is ejected from the needle tip, forming a characteristic Taylor cone. As the jet travels toward the collector, rapid solvent evaporation occurs, resulting in the deposition of continuous nanofibers that form a porous nonwoven membrane.

Electrospun polymer electrolyte membranes possess several advantageous properties for electrochemical applications. The interconnected porous structure and high surface-to-volume

ratio facilitate rapid ion migration and efficient electrolyte uptake, leading to enhanced ionic conductivity compared to conventionally cast polymer films. The nanofibrous morphology also improves electrode electrolyte interfacial contact, thereby reducing interfacial resistance and improving charge-transfer kinetics. In addition, electrospinning allows precise control over fiber diameter, porosity, membrane thickness, and structural morphology by adjusting parameters such as solution viscosity, applied voltage, feed rate, and tip-to-collector distance. The technique is also highly adaptable for incorporating ceramic nanofillers, ionic liquids, carbon materials, and active inorganic particles into the polymer matrix to further improve thermal stability, mechanical strength, and electrochemical performance.

Despite these advantages, electrospinning also has certain limitations. The process requires careful optimization of multiple operating parameters, and improper conditions may lead to bead formation, fiber breakage, or nonuniform membranes. The use of toxic organic solvents in some polymer systems can create environmental and safety concerns. In addition, large-scale production and thickness uniformity remain challenging for industrial applications. Nevertheless, electrospinning offers significant novelty in SPE fabrication because it enables the design of defect-engineered, highly porous, and mechanically flexible electrolyte membranes with superior ion-conduction characteristics. Recent advancements involving coaxial electrospinning, multilayer nanofiber architectures, aligned fiber structures, and hybrid organic inorganic electrospun electrolytes have further expanded the potential of this technique for developing safer, high-energy-density, and high-performance solid-state energy storage devices. The schematic illustration of Electrospinning technique is as depicted in Fig.4.

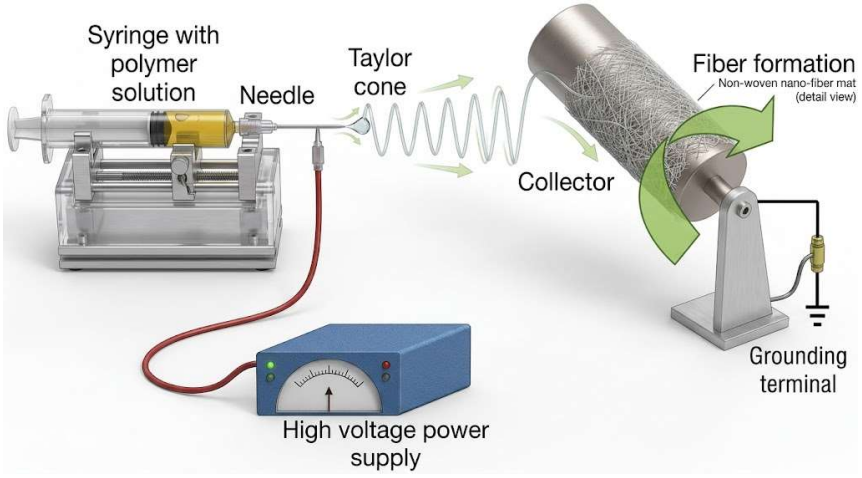

Figure 4. Electro Spinning Technique [Source Information: Generated from AI Gemini]

4.3 In-situ Polymerization

In-situ polymerization is an advanced fabrication technique widely used for the preparation of solid polymer electrolytes (SPEs) and gel polymer electrolytes (GPEs) for high-performance energy storage applications. In this method, polymerization occurs directly within the electrolyte system or inside the assembled battery configuration, leading to the formation of a highly integrated and uniform polymer network. The technique generally involves dissolving monomers, ionic salts, initiators, plasticizers, and functional additives in a suitable solvent to form a precursor solution. This precursor is then subjected to thermal, chemical, or photo-induced polymerization to convert the monomers into a cross-linked polymer matrix containing uniformly distributed ionic species. The polymerization process may occur either before electrolyte film formation or directly inside the battery cell after electrode assembly, resulting in intimate electrode electrolyte contact and improved interfacial compatibility.

One of the major advantages of in-situ polymerization is its ability to produce highly homogeneous electrolyte systems with enhanced interfacial adhesion between the electrolyte and electrode materials. Unlike conventional solution-cast electrolytes, in-situ polymerized electrolytes can effectively penetrate porous electrode structures, thereby minimizing void formation and reducing electrode electrolyte interfacial resistance. This improved interfacial contact facilitates efficient ion transport and enhances

electrochemical performance during repeated charge discharge cycles. The method also allows precise control over polymer network structure, cross-linking density, porosity, and mechanical properties by adjusting monomer composition, initiator concentration, and polymerization conditions. Furthermore, the incorporation of ionic liquids, ceramic nanofillers, plasticizers, and functional additives during polymerization enables the development of mechanically robust and thermally stable electrolyte systems with improved ionic conductivity and electrochemical stability.

In-situ polymerization offers several advantages for advanced battery systems, particularly lithium-metal and zinc-based batteries, where interfacial stability plays a critical role in long-term cycling performance. The formation of conformal polymer electrolyte layers helps suppress dendrite growth, reduce electrolyte leakage, and minimize undesirable side reactions. Additionally, cross-linked polymer networks formed during polymerization often exhibit superior dimensional stability, flexibility, and safety characteristics compared to conventional liquid electrolytes. The technique is also suitable for fabricating thin-film and flexible electrolytes for wearable and miniaturized energy storage devices.

However, the method also presents certain challenges. Precise control of polymerization kinetics is necessary to avoid incomplete polymerization, phase separation, or excessive cross-linking that may hinder ion mobility. Some polymerization initiators and solvents may also affect electrode stability or introduce unwanted impurities into the system. Despite these limitations, in-situ polymerization remains a highly promising and innovative approach for designing next-generation polymer electrolytes because of its capability to enhance interfacial engineering, improve mechanical integrity, and achieve high electrochemical performance in solid-state energy storage technologies.

4.4 Phase separation technique

Phase separation is an important fabrication technique widely employed for the preparation of porous polymer membranes and solid polymer electrolytes (SPEs) for advanced energy storage applications. The method is particularly attractive for developing polymer electrolytes with interconnected porous architectures, enhanced electrolyte uptake, and improved ion transport properties. In this technique, a homogeneous polymer solution containing polymer host materials, salts, plasticizers, or nanofillers is initially prepared using a suitable solvent system. Phase separation is then induced through solvent evaporation, temperature variation, nonsolvent immersion, or vapor-induced processes, resulting in the separation of polymer-rich and polymer-poor phases. Subsequent removal of the solvent or nonsolvent produces a porous polymer matrix with controlled microstructure and interconnected ion-conduction pathways.

The major advantage of the phase separation technique lies in its ability to fabricate highly porous and mechanically flexible electrolyte membranes with tunable pore size, morphology, and surface area. The interconnected porous structure facilitates enhanced electrolyte absorption and ionic mobility, thereby improving ionic conductivity and electrode electrolyte interfacial contact. The technique also enables incorporation of ceramic nanofillers, ionic liquids, and functional additives to tailor the mechanical, thermal, and electrochemical properties of the SPE. Furthermore, phase-separated membranes often exhibit lower crystallinity and enhanced amorphous regions, which are beneficial for ion transport in

polymer electrolyte systems. The process is comparatively simple, cost-effective, and adaptable for large-area membrane fabrication.

Despite these advantages, several processing challenges are associated with the phase separation method. Precise control over solvent evaporation rate, polymer concentration, temperature, humidity, and nonsolvent diffusion is critical for obtaining uniform pore distribution and defect-free membranes. Improper processing conditions may lead to pore collapse, irregular morphology, phase inhomogeneity, or weak mechanical strength. Excessive porosity can also reduce dimensional stability and mechanical robustness of the electrolyte membrane. Additionally, residual solvents trapped within the polymer matrix may adversely affect electrochemical stability and long-term device performance. The reproducibility of membrane morphology is another challenge, particularly when scaling the process for industrial applications.

From a critical perspective, the effectiveness of phase separation strongly depends on balancing porosity and mechanical integrity. While increased porosity enhances ion transport and electrolyte uptake, excessive pore formation may compromise structural stability and cycling durability. Therefore, optimization of processing parameters and composition is essential to achieve high-performance SPE systems. The technique is especially suitable for quasi-solid-state and gel polymer electrolytes, where porous structures can effectively retain liquid electrolytes while maintaining mechanical flexibility. In solid-state battery applications, phase-separated SPEs offer promising opportunities for improving ionic conductivity, interfacial contact, and electrochemical performance, making the method highly relevant for next-generation lithium-, sodium-, and zinc-ion energy storage technologies.

5 Performance Metrics & Characterization.

1.1 Ionic conductivity optimization

Ionic conductivity optimization is one of the most critical aspects in the development of high-performance solid polymer electrolytes (SPEs) for advanced energy storage applications. Ionic transport in polymer electrolytes is strongly governed by the amorphous nature of the polymer matrix, polymer chain flexibility, salt dissociation, and the availability of active coordination sites [36]. In general, higher amorphous content enhances ionic conductivity because ion migration predominantly occurs through the segmental motion of flexible polymer chains within disordered regions rather than crystalline domains. Crystalline regions restrict chain mobility due to tight polymer packing, whereas amorphous regions provide higher free volume and dynamic ion-conduction pathways [37]. Therefore, reducing crystallinity through polymer blending, plasticization, nanofiller incorporation, and defect engineering has become an effective strategy for improving ionic conductivity in SPE systems [38]. The presence of polar functional groups and active coordination sites within the polymer matrix also plays a crucial role in ion transport. Functional groups such as ether (O), hydroxyl (OH), carboxyl (COOH), carbonyl (C=O), and amine groups facilitate ion dissociation and polymer salt complexation through Lewis acid base interactions [39]. These groups provide transient hopping sites for cation migration and significantly influence the concentration of free mobile ions. Polymers such as Poly(ethylene oxide), Poly(vinyl alcohol), chitosan, pectin, and Na-CMC exhibit enhanced ionic conductivity due to the presence of oxygen-rich or polar functional groups capable of coordinating with alkali metal ions [39, 40]. The optimization of active site density and polymer flexibility directly affects dielectric properties, ion mobility, and electrochemical stability.

Several conductivity enhancement strategies have been extensively explored in SPE systems. Plasticizers such as glycerol, ethylene carbonate, propylene carbonate, and PEG are incorporated to reduce glass transition temperature (Tg), increase polymer chain flexibility, and enhance amorphousness, thereby improving ionic conductivity [41]. Nanofillers such as Zr0□, Ti0□, Si0□, Al□0□, LLZO, LATP, graphene oxide, CNTs, and MXene-based materials play a multifaceted role in enhancing ionic conductivity and electrochemical stability in solid polymer electrolytes. These fillers disrupt the ordered packing of polymer chains and suppress crystalline domain formation, thereby increasing the amorphous phase and promoting segmental chain mobility essential for ion transport [42]. Beyond simple crystallinity reduction, nanofillers create extensive polymer filler interfacial regions that act as preferential ion-conduction pathways due to localized space-charge effects and Lewis acid base interactions. Surface functional groups present on ceramic fillers interact with ionic salts, promoting salt dissociation and

increasing the concentration of free mobile ions while suppressing ion-pair formation and recombination [43].

Fillers possessing oxygen vacancies or high dielectric constants, particularly Ti0□, LLZO, and Al□0□, enhance local dielectric environments and reduce Coulombic attraction between cations and anions, thereby facilitating faster ionic migration. The conductivity enhancement strongly depends on filler size, surface chemistry, morphology, aspect ratio, and dispersion quality [44]. Nanosized fillers with high surface area generate larger interfacial regions and stronger polymer filler interactions compared to microsized particles. Surface-modified fillers and functionalized nanomaterials further improve compatibility with the polymer matrix, preventing agglomeration and ensuring homogeneous ion-transport networks. Strategies such as hybrid nanofiller incorporation, core shell fillers, defect-engineered ceramics, and aligned nanostructured fillers are increasingly employed to optimize continuous ion-conduction pathways and interfacial charge transport [45,46]. Similarly, incorporation of ionic liquids and plastic crystal additives enhances conductivity through dual mechanisms involving plasticization and additional ionic charge carriers. Ionic liquids reduce glass transition temperature, improve polymer flexibility, increase dielectric constant, and broaden electrochemical stability windows while simultaneously suppressing volatility and flammability.

However, conductivity optimization requires careful compositional balancing. Excessive filler loading leads to nanoparticle agglomeration, blockage of ion-conduction pathways, increased tortuosity, and deterioration of mechanical integrity. Similarly, excessive salt concentration may cause ion clustering, ion-pair formation, reduced segmental mobility, and phase separation, ultimately lowering ionic conductivity. Therefore, advanced optimization strategies including controlled filler loading, surface functionalization, cross-linking regulation, multilayer architectures, interfacial engineering, and software-assisted compositional optimization are essential for achieving synergistic enhancement of ionic conductivity, mechanical robustness, and long-term electrochemical stability in advanced SPE systems [47,48].

Another important parameter influencing ionic conductivity is the salt-to-functional group ratio. At low salt concentrations, the number of charge carriers remains insufficient for efficient ion transport. Increasing

salt concentration initially enhances ionic conductivity by increasing free ion concentration and promoting polymer salt interactions. However, beyond an optimum concentration, conductivity decreases due to ion-pair formation, reduced polymer chain mobility, and salt aggregation effects [49]. Therefore, optimizing the ratio between coordinating functional groups and salt concentration is essential to maximize free ion availability while minimizing ion association phenomena. This optimization strongly depends on polymer chemistry, dielectric constant, molecular architecture, and additive interactions. Significant progress has been achieved using various synthetic and biopolymer matrices for SPE fabrication. Recent developments involving ionic liquid-assisted systems, defect-engineered membranes, and interfacial modification strategies have further improved ionic conductivity, dielectric properties, mechanical strength, and electrochemical stability for metal-ion battery applications [50]. Recently Wang et al. [51] embed photoexcitation□modulated heterojunction fillers and bacterial cellulose into a PEO solid polymer electrolyte, showing that the photogenerated electric field boosts salt dissociation and raises room- temperature conductivity to 0.135 mS cm^{-1} with a Li^{+} transference number of 0.46, enabling flexible Li metal pouch cells retaining 86.7% capacity after 250 cycles at 25 °C. While Liu et al. [52] report a bioinspired gel polymer electrolyte with double dipole coupled side chains that weaken Li^{+} solvation, giving fast ion transport from −40 to 80 °C, including 1.03×10^{-4} S cm^{-1} at −40 °C, a Li^{+} transference number of 0.83, and LiNiCoMnO□||Li cells delivering 121.4 mAh g^{-1} at −30 °C and 172.2 mAh g^{-1} at 80 °C.

1.2 Cation Transference Number

The cationic transference number (t^{+}) is one of the most important electrochemical parameters governing the efficiency and stability of solid polymer electrolytes (SPEs) in rechargeable battery systems. It represents the fraction of total ionic current carried by cations within the electrolyte and directly influences ion transport kinetics, concentration polarization, charge-transfer behavior, and cycling stability. In conventional polymer electrolytes, both cations and anions contribute to ionic conduction, however, anions often possess higher mobility than cations because of their weaker coordination with the polymer matrix. As a result, the cationic transference number in many conventional SPEs remains relatively low, typically in the range of 0.1 0.5. A low cation transference number is undesirable in practical battery systems because only cations actively participate in electrochemical charge storage reactions, whereas mobile anions contribute primarily to parasitic ion redistribution and concentration gradients within the electrolyte [53]. One of the major consequences of low cationic transference number is concentration polarization arising from nonuniform anion accumulation near the electrode electrolyte interface during battery operation. Under prolonged current flow, rapidly moving anions migrate in the opposite direction of cations and accumulate locally, producing concentration gradients and ionic depletion regions. This phenomenon increases internal resistance, reduces ionic flux uniformity, and causes polarization losses, ultimately limiting rate capability, energy efficiency, and cycling stability. In lithium- and sodium-metal batteries, severe concentration polarization can also promote uneven metal deposition, dendrite formation, localized current hotspots, and interfacial instability. Therefore, suppression of anion mobility and enhancement of selective cation transport have become major research objectives in next-generation SPE development[54].

To overcome these limitations, considerable attention has been devoted toward the development of single-ion conducting polymer electrolytes (SIPEs), in which anionic groups are covalently immobilized onto the polymer backbone while only cations remain mobile. In SIPE systems, fixed anionic moieties such as sulfonates, carboxylates, borates, phosphates, or imides are chemically tethered to the polymer structure, thereby eliminating anion migration and significantly increasing cation transference number, often approaching unity ($t^{+} \approx 1$). This immobilization minimizes concentration polarization, reduces internal resistance, enhances ion-utilization efficiency, and improves electrochemical stability during long-term cycling [55]. Additionally, SIPEs promote uniform metal-ion flux at electrode interfaces, which is highly beneficial for suppressing dendrite growth and improving safety in metal-anode battery systems. Several optimization strategies have been developed to enhance cationic transference number in polymer electrolytes. Functionalization of polymer matrices with negatively charged groups enables partial anion immobilization and increases cation selectivity [56]. Cross-linking strategies, block copolymer architectures, ionic liquid incorporation, and polymer grafting techniques are also employed to regulate ion dissociation and ion mobility. Optimization of salt concentration, polymer flexibility, and interfacial compatibility remains essential to achieving high ionic conductivity simultaneously with high cation transference number, since these two parameters are often interdependent. Recent advances in SIPEs, hybrid nanocomposite electrolytes, and defect-engineered polymer systems continue to provide promising pathways toward high-energy-density, dendrite-free, and electrochemically stable solid-state battery technologies [57].

5.3. Electrochemical Stability Window (ESW)

The electrochemical stability window (ESW) is one of the most critical parameters governing the applicability of solid polymer electrolytes (SPEs) in advanced electrochemical energy storage systems. ESW represents the potential range within which the electrolyte remains electrochemically inert without undergoing oxidative or reductive decomposition at the electrode electrolyte interface [58]. In practical battery systems, the electrolyte must remain stable under the operating voltage of the electrode materials to prevent parasitic side reactions, gas evolution, interface degradation, capacity fading, and thermal instability. Therefore, a wider electrochemical stability window is essential for enabling high-voltage battery chemistries, improving energy density, and ensuring long-term cycling durability. Conventional polymer electrolytes often suffer from limited oxidative stability due to the presence of weak chemical bonds, unstable functional groups, residual solvents, or poor interfacial compatibility with highly reactive electrode materials [59].

The electrochemical stability of SPEs is strongly influenced by the chemical structure of the polymer matrix, the nature of functional groups, salt chemistry, dielectric properties, molecular orbital energies, and electrode electrolyte interfacial interactions. Functional groups present within the polymer backbone play a particularly important role in determining oxidation and reduction resistance. Electron-rich functional groups such as ether (O), hydroxyl (OH), carbonyl (C=O), carboxylate (COO^-), nitrile (C≡N), and fluorinated groups influence both ion coordination and electrochemical stability through their electron-donating or electron-withdrawing characteristics. Polymers containing highly electronegative groups, especially fluorinated moieties such as PVDF-based systems, generally exhibit superior oxidative stability because strong C F bonds possess high bond dissociation energies and lower highest occupied molecular orbital (HOMO) levels, making them less susceptible to oxidation at high voltages. In contrast, polymers containing highly reactive hydroxyl-rich structures may exhibit limited electrochemical stability due to easier oxidation or side reactions at elevated potentials [60].

The relationship between functional groups and ESW is closely associated with molecular orbital theory. Electrolytes with lower HOMO energy levels exhibit greater resistance toward oxidation, while higher lowest unoccupied molecular orbital (LUMO) levels improve reduction stability. Functional groups capable of delocalizing electron density or stabilizing coordinated ions contribute to improved electrochemical robustness. Strong polymer salt interactions also influence stability by modifying ion dissociation behavior and suppressing decomposition pathways. For instance, carboxylate and ether-rich polymers effectively coordinate alkali metal ions and reduce localized charge accumulation, thereby minimizing interfacial instability. Additionally, incorporation of cross-linkable groups, aromatic structures, and fluorinated segments enhances mechanical rigidity and suppresses chain degradation during electrochemical cycling [61].

Several advanced strategies have been developed to optimize the electrochemical stability window of SPE systems. Nanofillers such as Al□O□, TiO□, SiO□, LLZO, LATP, and ZrO□ improve ESW by reducing localized current density, stabilizing electrode electrolyte interfaces, scavenging impurities, and suppressing side reactions through Lewis acid base interactions [62]. Ceramic fillers also form mechanically robust interfacial regions that inhibit dendrite penetration and reduce electrolyte decomposition under high-voltage operation. Ionic liquids are widely employed to enhance ESW due to their intrinsically high oxidative stability, negligible vapor pressure, and nonflammable nature. Incorporation of ionic liquids lowers volatility while broadening operational voltage windows beyond conventional carbonate-based systems. Similarly, cross-linking strategies improve electrochemical stability by restricting excessive polymer chain mobility and enhancing dimensional integrity under electrochemical stress [63]. Surface coatings, multilayer electrolyte architectures, artificial interphase engineering, and polymer grafting techniques are increasingly utilized to stabilize electrode interfaces and suppress parasitic reactions [64].

One of the major challenges associated with limited ESW is interfacial degradation caused by electrolyte decomposition at high operating voltages. During electrochemical cycling, unstable electrolytes undergo oxidation at cathode surfaces or reduction near metal anodes, producing resistive interphase layers, gas generation, and irreversible side products. These processes increase interfacial impedance, reduce ion transport efficiency, and accelerate capacity fading. In metal-anode batteries, unstable electrolytes further promote dendritic deposition and uneven current distribution. Therefore, achieving high ionic conductivity alone is insufficient unless accompanied by wide electrochemical stability and interfacial compatibility. Optimization of ESW requires balancing polymer flexibility, ion dissociation capability, dielectric behavior, and chemical robustness simultaneously [65].

Recent developments in high-voltage SPE systems increasingly focus on molecular-level electrolyte engineering involving functional group modification, fluorination strategies, single-ion conducting architectures, nanostructured ceramic interfaces, and hybrid organic inorganic frameworks. Functionalized polymers with tailored electron density distribution and controlled interfacial chemistry have shown

significant improvements in oxidative stability beyond 4 5 V versus Li/Li$^+$. Furthermore, defect-engineered fillers, self-healing polymers, and artificial solid electrolyte interphase (SEI) forming additives are emerging as promising approaches for enhancing long-term electrochemical durability. Consequently, understanding the relationship between polymer chemistry, functional group characteristics, and electrochemical stability remains essential for designing next-generation SPEs capable of operating safely under high-voltage and high-energy-density battery conditions [66].

5.4. Mechanical and Thermal Integrity

Mechanical and thermal integrity are among the most critical requirements for solid polymer electrolytes (SPEs) intended for advanced lithium-, sodium-, and zinc-ion battery systems. In addition to high ionic conductivity, SPEs must possess sufficient mechanical robustness and thermal stability to maintain structural integrity, suppress dendrite penetration, tolerate electrochemical stress, and ensure long-term operational safety under varying thermal and electrochemical conditions. Mechanical and thermal properties are strongly interrelated with polymer chain dynamics, glass transition behavior, ion transport mechanisms, interfacial stability, and electrolyte morphology. Consequently, optimization of these parameters is essential for developing reliable solid-state battery technologies with improved cycle life, safety, and energy density [67].

Mechanical stability plays a decisive role in maintaining continuous electrode electrolyte contact during repeated charge discharge cycling. During battery operation, volume fluctuations, electrode expansion/contraction, interfacial stress accumulation, and localized current density variations can induce cracks, delamination, or structural failure within mechanically weak polymer electrolytes. Such degradation leads to increased interfacial resistance, poor ion transport continuity, and rapid capacity fading. Therefore, SPEs require adequate tensile strength, elastic modulus, flexibility, and dimensional stability to withstand prolonged electrochemical cycling without mechanical deterioration. One of the most important functions of mechanically robust SPEs is the suppression of dendritic metal growth in alkali metal batteries. During repeated metal stripping/plating processes, nonuniform ion flux and localized electric field enhancement promote dendrite nucleation and propagation through the electrolyte. If the electrolyte possesses insufficient mechanical modulus, dendrites can penetrate the polymer matrix, causing internal short circuits, thermal runaway, and catastrophic battery failure. According to the Monroe Newman criterion, an electrolyte with sufficiently high shear modulus can mechanically suppress dendrite penetration by resisting localized deformation at the electrode interface. Consequently, strategies such as ceramic nanofiller incorporation, cross-linking, polymer blending, reinforcement networks, and multilayer architectures are widely employed to improve the mechanical strength and dendrite resistance of SPE systems.

Nanofillers including Al□O□, SiO□, TiO□, ZrO□, LLZO, and graphene derivatives significantly enhance mechanical integrity through strong polymer filler interactions and stress-transfer mechanisms. Cross-linked polymer networks restrict excessive chain mobility and improve dimensional stability under electrochemical stress, while fibrous and porous reinforcement architectures provide mechanically interconnected ion-conduction pathways. However, excessive cross-linking or rigid filler loading may adversely reduce segmental chain mobility and ionic conductivity, highlighting the need for balanced optimization between mechanical rigidity and ion transport [68].

Thermal transition temperatures, particularly glass transition temperature (Tg) and melting temperature (Tm), are fundamentally linked to ionic transport behavior and polymer chain dynamics in SPEs. The glass transition temperature represents the temperature at which

polymer chains transition from a rigid glassy state to a flexible rubbery state. Ionic conduction in polymer electrolytes predominantly occurs through coordinated ion hopping assisted by segmental motion of polymer chains; therefore, lower Tg values generally correspond to enhanced chain flexibility, increased free volume, and improved ionic conductivity.

The relationship between ionic conductivity and polymer dynamics is commonly described using transport models such as the Vogel Tamman Fulcher (VTF) model and Arrhenius behavior. The VTF model explains temperature-dependent ionic conductivity in amorphous polymer systems where ion transport is strongly coupled with polymer segmental motion [69].

Here, ionic conductivity depends on polymer free volume and chain relaxation dynamics rather than simple thermally activated hopping alone. Lower Tg values reduce the activation barrier for segmental motion and facilitate rapid ion migration. Plasticizers, ionic liquids, and nanofillers are therefore incorporated to lower Tg and enhance amorphousness. However, excessively low Tg may compromise dimensional stability and mechanical integrity under elevated temperatures [69].

Relaxation dynamics are also associated with dielectric relaxation time, polymer chain mobility, and ion-coordination kinetics. Reduced relaxation times indicate faster dipolar orientation and enhanced ionic mobility. The coupling between dielectric relaxation and ionic transport is particularly important in highly amorphous SPE systems, where ion conduction is strongly dependent on dynamic polymer rearrangement. Consequently, optimization of Tg, chain flexibility, and dielectric behavior remains essential for balancing conductivity and mechanical stability [70].

Table 1. Comparison of recent studies on bio polymer electrolytes

Polymer matrix	nanofiller	Salt/ Ionic liquid (IL)	Conductivity	Electrochemical stability window	Cationic Transference number	Reference
PEO-PMMA	SiO2/Al2O3/ZnO / SnO2	LiClO4	10-5 S/cm	3 V	0.98	[71]
PVA:CA	ZnO nanoparticles	K2CO3	3.70 mS/cm	3.24 V	--	[72]
Chitosan/ Methylcellulose	--	NH4SCN	0.229 mS/cm	2.11 V	0.976	[73]
PVA:MC (60:40)	--	NaI	15.3 μS/cm, ambient Temperature	--	--	[74]
PVDF-HFP	g-C3N4 nanosheets	NaClO4	0.167 mS/cm	4.8 V	0.78	[75]
PEO/PVDF-HFP	Li1.5Al0.5Ge1.5 (PO4)3 + solvate ionic liquid	LiFSI	3.27 mS/cm, RT	4.9 V	--	[76]
Guar gum gel	Al2O3 nanofibers	LiClO4	2.37 mS/cm	4.6 V	0.59	[77]
PNPU-PVDF-HFP	Hydrogen-bond-rich PNPU network	--	0.413 mS/cm	--	--	[78]
Chitosan:HPMC:PEG 400	TiO2 nanofiller	LiClO4	0.129 mScm	3.35 V	0.70	[79]
Xanthan gum/ PVA blend	Carbon quantum dots	MgCl2 + ionic liquid	4.0 mS/cm at 60 °C	--	--	[80]
Cellulose acetate	BaTiO3	Mg(CF3SO3)2	2.4 mS/cm	3.51 V	0.41	[81]
PEO	ZIF-90-g-IL MOF nanofiller	--	0.117 mS/cm at 30 °C	4.8 V	0.44	[82]

6. Emerging Applications in Energy Storage Applications

The advanced solid state energy storage devices with Polymer electrolyte benefits are strongly constrained by conductivity, mechanics and interface trade□offs. For Li metal systems, dielectric□engineered and polymer□in□salt SPEs push room□temperature conductivities to 7.7×10^{-4} to 1.67×10^{-3} S cm^{-1} in PVDF LiFSI and PVDF□HFP/LiFSI/LLZTO, enabling Li||LiFePO□ cycling over 350 300 cycles and Li symmetric operation to 1500 h, but only at modest current densities (0.1 0.3 mA cm^{-2}) and often with relatively low cathode loadings, indicating that "high performance" is still defined under soft conditions (Kang et al. 2022) [83]. Recent study on single□phase local high□concentration blends and single□ion composite SPEs partially alleviate transference and dendrite issues, (reaching Li+ transference number up to 0.96 at an critical current density (CCD) approximately 3.2 3.7 mA cm^{-2}; 450 cycles with NMC811 at 4.5 V), yet they remain limited by processing complexity and unproven manufacturability at meter scale[84, 85] (Zhang et al. 2024; Wen et al. 2024). Low□temperature quasi□solid electrolytes based on 1,3,5- trioxane show 2.2×10^{-4} S cm^{-1} at −20 °C and maintain ~75% capacity at −20 °C vs 30 °C, but such temperature□robust behavior is validated only in coin and small pouch cells [86] (Z. Li, Yu, et al.

2023). Recent trend on low cost and inexpensive Zn and Mg ion batteries leads to major breakthroughs in divalent metal ion batteries. Interesting study by D wang et al on Zn□polymer electrolytes, clearly lag Li: PEO membranes softened with [Emim]OTF and ZnO achieve only 2.3×10^{-5} S cm^{-1}, and Zn||MnO□ cells reach 137 mAh g^{-1}, with anti□drying and flexibility as main strengths rather than rate or areal capacity (D. Wang et al. 2022) [87]. More conceptually advanced single□Zn^{2+} conductors and nanoconfined SPEs (PEGDA/PAM MOF and SPAF□PEtOx in PVDF) raise σ to 1.52 mS cm^{-1} and 5.04×10^{-4} S cm^{-1}, tZn^{2+} to 0.83, and stabilize Zn plating over 1000 3000 h and Zn||VO_2 full cells over 1000 2000 cycles, even at −40 °C, yet again under carefully controlled, relatively low□current conditions and without clear metrics on stack energy density (Hui et al. 2024; Yu et al. 2025) [88]. For supercapacitors, the contrast between “dry” SPE composites and GPEs is sharp: a NASICON polymer SPE requires an added acetonitrile “solvent layer” to recover interfacial utilization, boosting carbon□based device capacitance from ~175 to 263 F g^{-1} and retention to ~90% after 10,000 cycles, effectively admitting that solid solid contacts alone are inadequate. In comparison, succinonitrile/ionic□liquid GPEs with hydroquinone or $LiPF_6$ GPEs with MnCo- LDH routinely deliver 198 289 F g^{-1}, 40 Wh kg^{-1}, and ~10,000 cycles with ~85 100% Coulombic efficiency and inherent flexibility [89-95] (Yadav, Yadav, and Hashmi 2021; Fakhraei, Mirzaei-Saatlo, and Asghari 2024). Flexible□device reviews for Zn batteries and general polymer□based storage stress that many “wearable” demonstrations rely on thin, small□area cells, artistic form factors (fibers, papers, cables), and qualitative deformation tests rather than standardized bending/strain protocols or long□term cycling under simultaneous mechanical load, exposing a pronounced reality gap between laboratory prototypes and industrially relevant modules. Finally, explicit use of bio□derived matrices is sparse in this corpus; a 3D cellulose/ZIF□reinforced PEO succinonitrile electrolyte illustrates mechanical and electrochemical gains (σ ≈1.17×10^{-4} S cm^{-1}, tLi^{+} =0.40, 5 V stability, 18.7 MPa, >150 cycles with 99% retention) but is still a niche example rather than evidence of a mature “green SPE” paradigm, underscoring that biodegradable and bio□derived polymers for high□performance storage remain largely aspirational [95-104].

7. Conclusion

Polymer electrolytes have emerged as one of the most promising alternatives to conventional liquid electrolytes for next-generation energy storage systems owing to their enhanced safety, flexibility, and design versatility. Solid polymer electrolytes, gel polymer electrolytes, and composite/hybrid polymer electrolytes each offer unique advantages in terms of ionic transport, electrochemical stability, mechanical integrity, and interfacial compatibility. Significant progress has been achieved through the incorporation of plasticizers, ionic liquids, nanofillers, ceramic additives, and advanced interfacial engineering strategies, leading to substantial improvements in ionic conductivity, cation transference number, thermal stability, and cycling performance.

Among the various approaches, biopolymer-based electrolytes have gained increasing attention due to their renewable nature, biodegradability, low toxicity, and environmental sustainability. Recent advances demonstrate that natural polymers can serve as effective electrolyte hosts when combined with suitable salts, fillers, and functional additives. Despite these developments, challenges such as limited room-temperature ionic conductivity, interfacial instability, dendrite suppression, and large-scale manufacturing remain obstacles to commercialization. Future research should focus on multifunctional composite architectures, defect engineering, single-ion conducting systems, and scalable fabrication technologies to simultaneously optimize conductivity, mechanical strength, and electrochemical stability. Overall, polymer and biopolymer electrolytes are expected to play a pivotal role in the development of safer, sustainable, flexible, and high-energy-density batteries and supercapacitors, supporting the growing demands of modern energy storage applications.

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