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Review Article

Hybrid Pathways to Progress: Merging Solid-State Innovations with Sustainable Energy Design-

A comprehensive review on hybrid and solid-state material integration for next-generation sustainable energy storage

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ABSTRACT

The growing demand for safer and more sustainable rechargeable batteries has made the drawbacks of conventional liquid electrolytes increasingly clear, particularly their tendency to leak, their flammability and their unstable electrode–electrolyte interfaces. Hybrid and composite gel polymer electrolytes (GPEs) have emerged as promising alternatives because they combine the flexibility of polymers with the ionic conductivity and stability provided by liquid and solid components. However, most studies still examine individual hybrid GPE systems on their own and report performance gains without offering broader comparisons, so there is limited understanding of how specific material choices and interfacial engineering strategies control ion transport, interfacial stability and full-cell performance. This review addresses that gap by systematically compiling and critically analysing recent work on hybrid and composite GPEs for lithium-ion and sodium-ion batteries. It discusses different polymer matrices, fillers, ionic liquids, plasticizers and crosslinking approaches in terms of their effects on ionic conductivity, electrochemical stability, mechanical properties and electrode compatibility. Based on the reported results, key structure property performance relationships are highlighted and translated into practical design guidelines for next-generation hybrid GPEs. Remaining challenges such as limited room-temperature conductivity, interfacial resistance, dendrite growth and scalable processing are outlined, and future research directions are suggested for the development of robust, high-safety gel polymer electrolyte systems for advanced energy storage.

Keywords: Silicon-graphite hybrid anode, gel polymer electrolytes(GPE), solid state batteries(SSB), Nano structured functional composite, electrochemical impedance spectroscopy, sodium-ion batteries, interface engineering, ionic conductivity, battery characterization

1. INTRODUCTION

The twenty-first century is fundamentally powered by electrochemical energy storage systems. From electric vehicles and renewable energy grids to mobile electronics and defence technologies, batteries have become the central enabler of global technological advancement. As society accelerates toward an electrified and carbon-neutral future, the performance, safety, and scalability of batteries are no longer secondary considerations—they define the pace and sustainability of this transition [9,13].

Contemporary energy storage technologies must simultaneously satisfy three essential criteria: high energy and power density, robust safety and thermal stability, and scalable cost-effective manufacturing. Meeting these interconnected objectives

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ensures that the global shift toward sustainable energy remains technologically and economically viable [9,11].

Over the past three decades, lithium-ion batteries (LIBs) have dominated the energy storage domain due to their high specific energy, lightweight architecture, and stable cycling performance [6,9]. However, as demand surges across sectors such as electric mobility and grid-scale storage, the inherent limitations of conventional LIB configurations have become increasingly evident. Lithium resources remain geographically concentrated and expensive to extract, introducing supply chain vulnerabilities [11]. Moreover, graphite anodes—though mature and reliable—are limited by their theoretical capacity of only 372 mAh g^{-1}, restricting further energy density improvements [6]. Additionally, liquid electrolytes used in traditional LIBs are flammable and prone to dendrite formation, which raises significant safety and performance concerns [9].

To address these limitations, current research emphasizes synergistic material integration rather than isolated innovation. Instead of focusing solely on individual components, next-generation battery development now centres on multi-material systems capable of working in harmony to deliver system-level improvements [1–3,9]. Among these, silicon–graphite hybrid anodes, gel polymer electrolytes (GPEs), nanostructured functional composites, and solid-state electrolytes have emerged as the most promising technologies for achieving both high performance and safety [1–4,9]. Silicon–graphite hybrids enhance energy storage capacity by leveraging silicon's ultrahigh theoretical capacity while mitigating its volume-expansion challenges through graphite's mechanical buffering [1,4,6,7]. Gel polymer electrolytes bridge the performance gap between liquid and solid systems, providing superior ionic conductivity, thermal stability, and interfacial compatibility [2,5,8]. Nanostructured composites further stabilize electrochemical interfaces and accelerate charge transport, while solid-state electrolytes offer higher safety margins, increased energy density, and improved cycle life [9,11]. In summary, the evolution of battery technology is no longer defined by incremental progress but by the integration of complementary materials and architectures. This review investigates how the synergistic combination of silicon–graphite hybrid anodes, GPEs, nanostructured composites, and solid-state systems can collectively redefine the future of sustainable, high-performance energy storage.

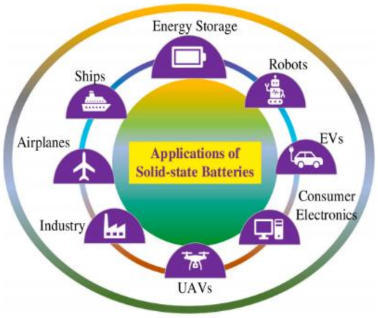

Figure1.:
Overview of the key application domains for next-generation battery technologies, including electric mobility, grid storage, consumer electronics, and defence systems. Adapted from [9].

1.1 Silicon–Graphite Hybrid Anodes

The anode remains a central performance-defining component in both lithium-ion and sodium-ion batteries. Silicon has emerged as one of the most promising anode materials because of its exceptionally high theoretical capacity (~4200 mAh g^{-1}), which is nearly ten times higher than that of commercial graphite (372 mAh g^{-1}) [1,4,6]. Furthermore, silicon's natural abundance, low environmental impact, and cost-effectiveness make it an attractive candidate for next-generation energy storage systems [1,4]. Despite these advantages, silicon anodes face significant technical challenges. During lithiation and delithiation, silicon undergoes volumetric expansion of nearly 300%, leading to pulverization, electrical

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disconnection, and unstable solid–electrolyte interphase (SEI) formation. These structural instabilities cause rapid capacity fading and poor cycle retention [6]. In contrast, graphite exhibits excellent mechanical stability, good conductivity, and a well-formed SEI, but its limited specific capacity restricts further enhancement of energy density [7].Silicon–graphite hybrid anodes provide a synergistic solution to these limitations. In such systems, graphite acts as a mechanical buffer to accommodate silicon's expansion while facilitating efficient electron transport and stabilizing SEI formation. Silicon, in turn, enhances the overall energy density of the cell. This complementary relationship has been shown to significantly improve cycle life, rate performance, and structural durability [1,7]. Liu et al. [1] demonstrated that hybrid Si–graphene composites can achieve excellent capacity retention and long-term cycling stability compared with conventional graphite anodes, while Wu et al. [7] emphasized the vital role of carbon in improving interface stability and mechanical resilience.

The design of high-performance silicon–graphite hybrids focuses on achieving uniform distribution, stable interfaces, and efficient mechanical buffering. Several advanced architectures—such as core–shell composites, porous matrices, and graphene- or carbon-coated hybrids—have been proposed to optimize these characteristics [1,6,7]. In core–shell structures, a conductive graphite or carbon coating encapsulates silicon nanoparticles to minimize direct exposure to the electrolyte, reducing SEI instability and mechanical stress [7]. Porous carbon frameworks provide void space to accommodate volume expansion, maintain electrical connectivity, and enhance electrolyte accessibility [4,6]. Graphene and carbon nanotube reinforcements further improve electron transport pathways and mechanical integrity [1,7].

Recent progress has also been made in the development of scalable synthesis methods such as high-energy ball milling, spray drying, and in situ carbonization. These approaches not only reduce cost but also enable tunable control of particle morphology and hybrid ratios, making them compatible with commercial electrode fabrication processes [10]. Furthermore, interface engineering using functional binders, surface coatings, and artificial SEI layers has proven effective in suppressing interfacial degradation and improving cycling life, particularly in high-loading electrodes designed for electric vehicles and grid-scale applications [2,5].

Experimental results have consistently shown that Si–graphite hybrids outperform pure silicon and pure graphite electrodes in both specific capacity and long-term cycling. For example, Si–graphene hybrid composites exhibit high capacity retention even after 500 cycles and deliver enhanced rate capability due to improved structural and electrical stability [1,7]. In sodium-ion systems, similar hybrid frameworks have shown promise in mitigating the challenges of larger ionic radii through mechanical flexibility and optimized electronic pathways [4,6].

This balance of capacity and stability positions hybrid anodes as a transitional platform between conventional graphite and emerging solid-state designs [9].

Despite considerable advancements, several barriers remain before Si–graphite hybrid systems can achieve large-scale industrial adoption. SEI instability remains a primary cause of performance degradation during extended cycling, as repeated volume changes induce fracture and reformation of the SEI [6,7]. High initial irreversible capacity loss remains problematic, particularly in silicon-

Material Type	**Specific Capacity (mAh g^{-1})**	**Cycle Life (cycles)**	**Key Advantage**	**Primary Limitation**
Pure Graphite	350–370	>1000	Excellent stability and low cost	Limited energy density [6]
Pure Silicon	3000–4200	<100	High theoretical capacity	Severe expansion, unstable SEI [4,6]
Si–Graphite Hybrid	800–1500	300–1000	Balanced capacity and cycle stability	Manufacturing complexity, SEI control [1,7,10]

table 1- Comparison of Graphite, Silicon, and Si–Graphite Hybrid Anode Materials

rich composites [4,7]. Mechanical stress at high areal loadings can lead to electrode cracking and reduced efficiency [4,6]. Manufacturing constraints persist, particularly in maintaining uniform dispersion and interfacial bonding at production scale [10].

Furthermore, achieving compatibility between hybrid anodes and advanced electrolytes such as gel polymer electrolytes (GPEs) and solid-state electrolytes (SSEs) is essential for improving interfacial stability, safety, and overall system performance [2,9].

Recent research directions emphasize integrating materials innovation with process scalability. Key areas include pre-lithiation and pre-sodiation strategies to enhance initial Coulombic efficiency and reduce irreversible losses [4,6]; multifunctional binders with self-healing or stretchable properties to sustain electrode integrity under stress [7]; AI-assisted material modeling for predicting optimal Si/C ratios and interface configurations [11]; and hybrid integration with GPEs and SSEs to improve thermal safety, interface contact, and electrochemical performance [2,9].

Patent and publication data show rapid growth in Si–graphite composite and hybrid electrolyte research, signaling a clear transition from academic exploration to pre-commercial maturity [11]. This convergence of scalable manufacturing techniques, advanced interface engineering, and predictive computational design defines the roadmap toward next-generation high-energy, long-life battery systems.

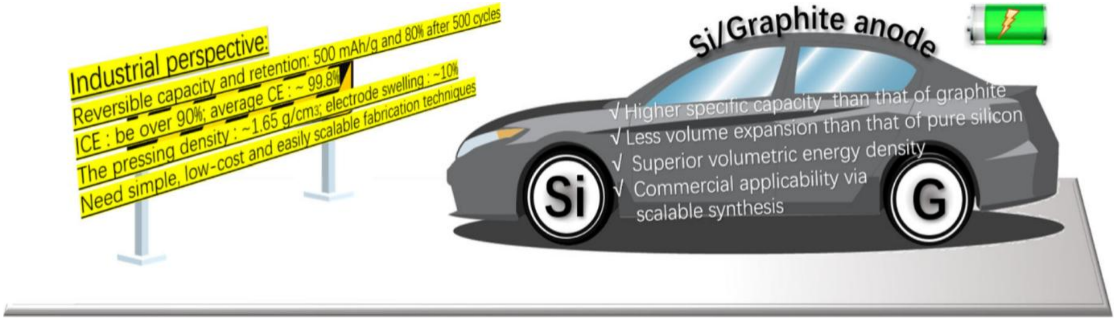

Figure2.:

illustrates the silicon–graphite hybrid anode concept as a dual-core engine, where silicon delivers capacity and graphite provides stability. Adapted from [7].Schematic illustration of "dual-core" (Si and graphite) -powered vehicle, showing many advantages and also obstacles from the industrial perspective that need to be addressed in the near future. CE, columbic efficiency; Si, silicon (7)

1.2 Gel Polymer Electrolytes: A Safer Ionic Pathway

Electrolytes play a decisive role in determining the performance, safety, and longevity of modern energy storage systems. Traditional liquid electrolytes, while offering high ionic conductivity, suffer from several limitations including flammability, leakage, and the tendency to promote lithium dendrite formation. These challenges have driven growing interest in gel polymer electrolytes (GPEs), which combine the ionic conductivity of liquids with the mechanical stability of solids, creating a safer and more adaptable platform for advanced batteries [2,5,8,9]. GPEs consist of a polymer matrix—such as PVDF-HFP, PEO, or PAN—swollen with a liquid electrolyte solution containing lithium or sodium salts. This hybrid design enables the system to maintain high ionic mobility while improving structural integrity and thermal safety. The polymer framework confines the liquid phase, minimizing the risk of leakage or flammability, and allowing GPEs to retain ionic conductivities in the range of 10^{-4}–10^{-3} S cm^{-1}, comparable to conventional liquid systems [2,5].

Recent advancements in materials design have further enhanced the functionality of GPEs. Cross-linked polymer networks, ionic liquid-based gels, and Nano fillerreinforced composites have demonstrated improved mechanical strength, enhanced electrochemical stability, and consistent performance even at ambient temperatures [8,9]. Nano fillers such as Al_2O_3, SiO_2, and TiO_2 not only strengthen the polymer framework but also promote faster ion transport and suppress dendritic growth, thereby improving electrode–electrolyte compatibility [5,8].

One of the most critical advantages of GPEs lies in their interface behaviour with high-capacity anodes such as silicon. The semi-solid nature of GPEs allows them to form a conformal contact with electrode surfaces, creating a uniform and flexible interface that can accommodate mechanical stress during cycling. This helps in stabilizing the solid–electrolyte interphase (SEI) and mitigating degradation mechanisms that commonly occur with rigid electrolytes [2,8].GPEs are also proving highly compatible with sodium-ion batteries, which require more flexible ion transport channels due to the larger ionic radius of sodium ions. The adaptability of GPEs supports efficient Na^+ transport and improved electrochemical stability, making them a strong candidate for cost-effective, large-scale energy storage [8,9].

From a manufacturing perspective, GPEs offer excellent process ability and are compatible with scalable fabrication techniques such as solution casting and roll-to-roll coating. This industrial advantage, coupled with their mechanical flexibility, positions GPEs as an ideal bridge between liquid-electrolyte batteries and solid-state systems. Their ability to serve as interfacial layers in hybrid battery architectures particularly between silicon–graphite anodes and solid electrolytes makes them a key enabler of next-generation, safe, and high-energy devices [2,5,9].In summary, gel polymer electrolytes represent a crucial advancement in battery design by combining safety, ionic mobility, and interface stability. Emerging research in self-healing gels, hybrid solid–gel systems, and AI-optimized formulations continues to extend the boundaries of this technology, accelerating its transition from laboratory research to practical, commercial applications [2,5,8,9].

1.3 Nanostructured Functional Composites

Electrode and electrolyte optimization have significantly improved battery performance, yet one of the most transformative advancements in recent years lies in the integration of nanostructured functional composites. These engineered materials, which combine inorganic nanoparticles and conductive polymer frameworks, serve multiple functions — enhancing ion transport, improving mechanical resilience, and stabilizing interfacial reactions [3,5,8,9,11].

When materials are engineered at the nanoscale, their surface-to-volume ratio increases dramatically, allowing faster ionic movement and greater electrochemical reactivity. This structural refinement not only shortens ion diffusion pathways but also provides abundant active sites for lithium or sodium insertion. Furthermore, elastic or porous nanostructures can absorb the mechanical stress caused by repetitive volume expansion, particularly in silicon-rich anodes, thus mitigating structural degradation during cycling [3,8]. Among various systems explored, manganese oxide (Mn_2O_3) based composites have demonstrated exceptional promise. When integrated with conductive polymer networks such as poly(thiophene-co-pyrrole), these hybrids exhibit remarkable synergy Mn_2O_3 contributes electronic conductivity and structural rigidity, while the polymer imparts elasticity and maintains continuous electron pathways [3]. This combination promotes uniform ion distribution, reduces interfacial impedance, and results in more stable solid–electrolyte interphase (SEI) formation during extended operation.

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In addition to oxide-based nanostructures, conductive polymer gels and hybrid nanocomposites have been investigated for their ability to enhance both ionic transport and electrochemical durability. These multifunctional frameworks not only serve as mechanical buffers but also act as smart materials — capable of self-healing, stress dissipation, and even dielectric or photocatalytic functionality under specific conditions [5,9]. Such adaptability makes them particularly valuable for next-generation flexible and wearable battery systems.

Recent research further highlights the importance of Nano interfacial engineering, where the interphase between electrode and electrolyte is modified at the atomic or molecular level to suppress parasitic reactions and reduce interfacial resistance. Nanostructured coatings, functional fillers, and self-assembled Nano layers have been shown to stabilize SEI layers, improve electrode–electrolyte adhesion, and enhance ionic mobility [8,9]. These advances are especially relevant to hybrid systems integrating silicon–graphite anodes and gel polymer or solid-state electrolytes, where interfacial compatibility directly influences performance and cycle life [2,5,9].The growing body of patents and research on nanocomposite electrolytes and interlayers further signifies the transition of this technology from conceptual research to applied engineering [11]. Through innovations such as 3D nanostructured scaffolds, printable composite films, and adaptive interfacial materials, nanostructured composites are rapidly becoming an essential component of the evolving energy storage landscape.

In summary, nanostructured functional composites represent a crucial frontier in battery materials science. By combining high surface reactivity, mechanical resilience, and multi-functionality, they not only enhance electrochemical performance but also enable the design of safer, more flexible, and longer-lasting energy storage systems [3,5,8,9,11].

1.4 Electrochemical Impedance Spectroscopy (EIS)

Understanding how advanced battery materials interact and perform under operational conditions requires precise diagnostic tools. Electrochemical Impedance Spectroscopy (EIS) has emerged as one of the most powerful and versatile characterization techniques for probing electrochemical processes at both bulk and interfacial levels [8,9]. By applying a small alternating current (AC) perturbation and measuring the resulting voltage response across a frequency range, EIS provides valuable insight into ionic transport, charge-transfer resistance, diffusion dynamics, and interfacial stability within electrochemical cells.EIS is particularly critical for next-generation battery systems, including those employing silicon–graphite hybrid anodes, gel polymer electrolytes (GPEs), and solid-state electrolytes (SSEs). These architectures involve complex interfaces that strongly influence overall performance. Through impedance spectra—typically represented as Nyquist or Bode plots—EIS allows the deconvolution of various resistive and capacitive components associated with the electrolyte, electrode materials, and interphases[8].
A typical EIS profile consists of three major regions:

High-frequency response:
Corresponding to the bulk resistance of the electrolyte (R_b), which determines ionic conductivity.
Mid-frequency semicircle:
Representing charge-transfer resistance (R_ct) and interfacial capacitance at the electrode–electrolyte interface.
Low-frequency tail:
Associated with ion diffusion within electrodes or through SEI layers, often modelled as a Warburg element [8,9].

By analysing these parameters, researchers can quantify key electrochemical behaviours such as SEI formation quality, ion mobility, and interface degradation. In silicon–graphite hybrid anodes, for instance, EIS helps monitor the evolution of impedance associated with volume expansion and SEI reconstruction during cycling. Similarly, in GPE and solid- disconnection, and unstable solid–electrolyte interphase (SEI) formation. These structural instabilities cause rapid capacity fading and poor cycle retention [6]. In contrast, graphite exhibits excellent mechanical stability, good conductivity, and a well-formed SEI, but its limited specific capacity restricts further enhancement of energy density [7].or example, operando EIS enables the observation of interfacial changes during charge–discharge cycles, while AI-driven algorithms are increasingly being used to model impedance data and predict degradation mechanisms with high accuracy [11].

In hybrid and solid-state configurations, EIS has become indispensable for quantifying ionic conductivity (σ), interfacial charge-transfer efficiency, and thermal effects factors that dictate the overall energy and power performance of a cell. The method also facilitates optimization of new materials by correlating microscopic structure with macroscopic electrochemical response, guiding both academic research and industrial product development [8,9,11].

In summary, Electrochemical Impedance Spectroscopy is not merely a diagnostic technique but a strategic tool for design feedback and performance optimization. Its ability to dissect the complex interplay between materials, interfaces, and ion dynamics makes it essential for advancing hybrid, polymer-based, and solid-state battery technologies. As EIS continues to integrate with data-driven modelling and in situ monitoring, it is expected to play a defining role in accelerating the commercial realization of next-generation energy storage systems [8,9,11].

1.5 Innovation Trends and Industry Transition

The global battery industry is undergoing a rapid transformation, driven by the growing demand for high-energy, long-lasting, and safe energy storage systems. The focus has shifted from incremental material improvements to system level innovation, where materials, design, and manufacturing technologies evolve in a coordinated manner to deliver optimized performance [9,10,11]. This transition reflects a maturing industry moving from laboratory-scale discoveries to scalable and commercially viable energy storage solutions.

One of the most notable innovation trends is the integration of hybrid material systems such as silicon–graphite anodes, gel polymer electrolytes (GPEs), and solid-state electrolytes (SSEs) into unified architectures that balance energy density, safety, and manufacturing feasibility [9]. These composite systems combine the strengths of individual components while minimizing their inherent weaknesses, enabling the design of batteries that are both high-performing and durable. For instance, the combination of Si–C hybrid anodes with flexible polymer or solid electrolytes has demonstrated significant improvements in cycle life and safety compared to traditional lithium-ion configurations [10].

Another key area of advancement is interface engineering, where the atomic-scale design of electrode–electrolyte interfaces is used to control ion transport, reduce resistance, and improve long-term stability. Researchers are increasingly employing in situ polymerization, artificial SEI formation, and Nano layer coatings to mitigate interfacial degradation, enhance adhesion, and promote uniform ion flux [9,11]. These strategies are particularly valuable in solid-state and hybrid systems, where interface quality often dictates performance more than bulk material properties.

Simultaneously, manufacturing innovation is accelerating the transition from experimental prototypes to industrial deployment. Scalable fabrication techniques such as spray drying, high-energy ball milling, roll-to-roll coating, and 3D printing are being adopted to produce advanced electrodes and electrolytes with precise microstructural control [10]. These approaches not only reduce cost and material waste but also enable customized architectures tailored for specific applications such as electric vehicles, portable electronics, and grid storage.At the digital frontier, the integration of artificial intelligence (AI), machine learning (ML), and computational modelling is redefining how battery materials are designed, optimized, and tested. Predictive algorithms can now simulate material behaviour, identify degradation pathways, and recommend compositional modifications before physical synthesis, drastically reducing development time [11]. This data-driven approach complements experimental research, allowing for faster optimization cycles and more reliable performance forecasting.

Furthermore, the industry is witnessing a growing emphasis on sustainability and circular economy principles. Recycling and reprocessing of electrode materials, the use of bio-derived polymers for electrolytes, and the minimization of critical resource dependency (such as cobalt and nickel) are becoming integral to next-generation battery R&D [9,10]. This shift not only aligns with environmental imperatives but also enhances the long-term economic viability of large-scale battery deployment.

In summary, the battery sector is evolving from a materials-driven science to an integrated innovation ecosystem. The convergence of hybrid architectures, digital design tools, scalable manufacturing, and sustainability practices is paving the way for a new industrial paradigm. This collective movement signals a decisive step toward commercial readiness and global adoption of safer, smarter, and more efficient energy storage technologies [9,10,11].

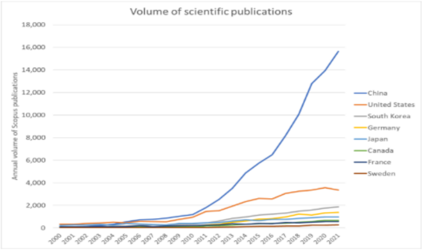


Figure 3. Development of paper volumes.

Figure3.:
Growth in publications and patent activity related to silicon–graphite hybrid anodes and solid-state electrolytes over the last decade. Adapted from [11].

1.6 Scope and Structure of This Review

This review presents a comprehensive discussion of emerging materials and design strategies that are shaping the future of rechargeable battery technology. It begins with an in-depth analysis of silicon–graphite hybrid anodes, emphasizing their structural optimization, electrochemical behaviour, and interfacial engineering approaches. Subsequent sections examine the development and performance of gel polymer electrolytes (GPEs), nanostructured functional composites, and solid-state electrolyte systems, highlighting their synergistic roles in enhancing energy density, safety, and long-term stability.

The review also integrates Electrochemical Impedance Spectroscopy (EIS) as a diagnostic framework to evaluate ionic transport, charge-transfer resistance, and interfacial stability in hybrid and solid-state systems. Furthermore, it outlines industrial innovation trends, covering scalable fabrication methods, data-driven material optimization, and sustainability considerations that are accelerating the commercialization of advanced energy storage systems [8–11].

Through this structured approach, the article aims to provide not only a consolidated understanding of recent scientific progress but also a forward-looking perspective on how material synergy, interfacial engineering, and digital design are redefining the landscape of next-generation battery technologies.

2. Silicon–Graphite Hybrid Anodes

2.1 Introduction to Silicon-Based Anodes

Among various anode materials investigated for next-generation lithium-ion and sodium-ion batteries, silicon stands out due to its exceptionally high theoretical capacity of approximately 4200 mAh g^{-1}, which is nearly ten times that of conventional

Figure4.:
Schematic illustration of the silicon–graphite hybrid anode concept. Silicon contributes high capacity, while graphite buffers expansion and maintains electronic pathways. Adapted from [1,7].

However, the practical deployment of pure silicon anodes faces severe challenges. During lithiation and delithiation, silicon undergoes a volumetric expansion of nearly 300%, resulting in pulverization, loss of electrical contact, and unstable solid–electrolyte interphase (SEI) formation. These phenomena lead to rapid capacity fading and poor cycling stability [6]. Graphite, on the other hand, is structurally stable but limited by its low specific capacity and slower lithium intercalation kinetics. Bridging the gap between these two materials has given rise to a new class of anodes — silicon–graphite hybrids — which harness the high capacity of silicon and the mechanical and electrical stability of graphite [7,9].

2.2 Structure and Synergistic Mechanisms

The silicon–graphite hybrid architecture operates on a principle of synergy: silicon contributes high energy capacity, while graphite acts as a flexible, conductive buffer that absorbs mechanical strain and stabilizes the SEI. This hybrid structure ensures better stress accommodation, electronic conductivity, and cycle retention compared to either component alone [1,7].

In these hybrids, graphite also provides pathways for rapid electron transfer and uniform lithium-ion distribution, reducing localized stress and suppressing dendritic growth. Moreover, graphite's graphitic planes facilitate the formation of a stable SEI, which minimizes continuous electrolyte decomposition [6,9]. Silicon, in turn, increases the total energy density and ensures improved capacity retention when embedded within the carbon matrix.

Experimental and computational studies confirm that a balanced Si:C ratio is crucial. A silicon content of 10–30% often yields optimal performance, where volumetric expansion is well managed without sacrificing capacity [1,7]. Excess silicon leads to structural instability, whereas low silicon content fails to deliver substantial capacity gains.

2.3 Fabrication Approaches and Design Strategies

The fabrication of silicon–graphite hybrid anodes has evolved significantly in recent years, focusing on achieving homogeneous dispersion, robust interfacial bonding, and mechanical flexibility.

Prominent synthesis techniques include:
High-energy ball milling: A scalable and low-cost method for uniformly mixing silicon and graphite powders, enabling

nanoscale dispersion and intimate contact between components [10].Spray drying and in situ carbonization: These methods produce spherical composite particles with controlled porosity, enhancing both ionic and electronic transport [9].Chemical vapour deposition (CVD): Used to create core–shell structures, where a thin carbon layer or graphite shell encapsulates silicon nanoparticles, protecting them from direct electrolyte exposure and SEI fracture [7]. In addition, interface engineering plays a vital role in extending cycle life. The application of functional polymer binders (such as PAA, CMC, or alginate) and artificial SEI coatings (e.g., Al_2O_3, LiF, or Li_3PO_4 layers) has shown to effectively suppress mechanical degradation and electrolyte decomposition [4,9]. Advanced binders with self-healing or elastic properties can maintain electrode integrity even under repetitive expansion–contraction cycles [11].

2.4 Electrochemical Performance and Optimization

The hybrid design substantially enhances the electrochemical stability and rate capability of the anode. Optimized Si–C composites exhibit specific capacities ranging between 800 and 1500 mAh g^{-1} and can retain over 80% of their capacity after 500 cycles under moderate current densities [1,7,9].

Material Type	Specific Capacity (mAh g^{-1})	Cycle Life (cycles)	Key Advantage	Major Limitation
Graphite	350–370	>1000	Excellent stability, low cost	Limited capacity [6]
Silicon	3000–4200	<100	Ultra-high capacity	Volume expansion, unstable SEI [4,6]
Si–Graphite Hybrid	800–1500	300–1000	Balanced performance	Manufacturing complexity [1,7,9,10]

table 2- Comparison of Graphite, Silicon, and Si–Graphite Hybrid Anode Materials

The inclusion of graphitic buffering suppresses impedance growth and enhances lithium-ion diffusion. EIS measurements show lower charge-transfer resistance and improved interfacial kinetics for hybrid composites compared with pure silicon anodes [8,9]. The development of nanoscale voids, porous channels, and conductive carbon frameworks further facilitates ion and electron transport, leading to improved rate performance and reduced polarization [9,11].

2.5 Integration with Advanced Electrolytes

One of the most promising frontiers for silicon–graphite hybrids is their integration with advanced electrolytes such as gel polymer electrolytes (GPEs) and solid-state electrolytes (SSEs). These electrolyte systems provide enhanced thermal safety, wider electrochemical windows, and better interfacial stability, addressing the key limitations of liquid-based electrolytes [2,9].

GPEs, due to their semi-solid nature, form conformal interfaces with hybrid anodes, accommodating expansion while maintaining ionic conductivity [5,8]. Similarly, SSEs paired with Si–C anodes have demonstrated superior performance through interface engineering techniques that lower interfacial impedance and improve overall mechanical compatibility [9]. The co-optimization of hybrid anode composition and electrolyte type is, therefore, a pivotal factor in advancing next-generation high-energy-density batteries [10,11].

2.6 Challenges and Future Prospects

Despite the impressive progress, several challenges still hinder the large-scale commercialization of silicon–graphite hybrid anodes:SEI instability due to continuous expansion–contraction cycles [6]. Irreversible lithium consumption during initial cycles leading to capacity loss [4,7].

Complex manufacturing requirements for consistent particle dispersion and uniform coating [10]. Limited understanding of long-term degradation mechanisms under practical operating conditions [11]. Future research must focus on multi-scale modelling to correlate mechanical, electrochemical, and thermal behaviours, as well as AI-guided material discovery to identify optimal hybrid compositions. Developing scalable, eco-friendly synthesis routes and self-healing architectures will further accelerate the transition of these materials from the lab to commercial reality [9,11].

In conclusion, silicon–graphite hybrid anodes represent a critical advancement bridging conventional lithium-ion technology and future solid-state battery systems. Their unique combination of high energy capacity, mechanical resilience, and interfacial stability positions them at the forefront of the next generation of rechargeable energy storage technologies [9–11].

3. Gel Polymer Electrolytes (GPE)

3.1 Fundamentals and Rational

As the demand for safer and high-energy batteries intensifies, electrolytes have become the critical component defining system performance, safety, and lifetime. Traditional liquid electrolytes, although exhibiting excellent ionic conductivity, are inherently limited by flammability, leakage, and dendrite formation, posing serious challenges for large-scale and high-voltage applications [2,5].

Gel Polymer Electrolytes (GPEs) have emerged as a transformative solution that bridges the gap between liquid and solid systems. A GPE typically consists of a polymer matrix—such as PVDF-HFP, PEO, PAN, or PMMA—incorporating a liquid electrolyte within its porous framework. This semi-solid design combines the ionic mobility of liquids with the mechanical stability of solids, achieving ionic conductivities in the range of 10^{-4}–10^{-3} S cm^{-1} while significantly improving safety and flexibility [5,8,9]. The polymer host immobilizes the liquid phase and prevents electrolyte leakage, while the presence of plasticizers and inorganic fillers enhances both ionic transport and mechanical robustness. This makes GPEs ideal for flexible, wearable, and next-generation lithium- and sodium-ion batteries [2,8].

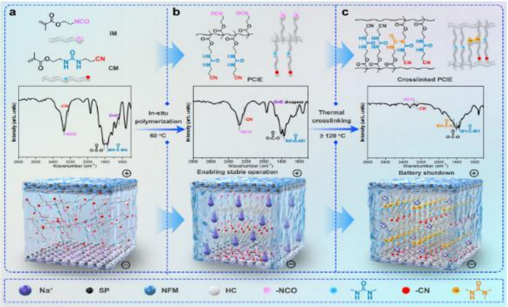


Figure 5.:
Schematic illustration of a gel polymer electrolyte (GPE) system showing Li⁺/Na⁺ transport through polymer matrix, liquid retention, and stable electrode–electrolyte interface. Adapted from [2,5,8].

3.2 Structural Design and Material Engineering

The performance of GPEs depends heavily on polymer selection, filler type, and crosslinking strategy. Common polymer hosts such as PEO provide efficient lithium-ion conduction through segmental motion, whereas PVDF-HFP offers excellent electrochemical stability and mechanical strength [5]. Blending these polymers or incorporating **ionic** liquids can widen the electrochemical stability window and improve interfacial contact.

To overcome the intrinsic low ionic mobility of polymers, researchers have incorporated nanofillers such as SiO_2, TiO_2, Al_2O_3, and graphene oxide (GO) into the GPE matrix [8,9]. These nanofillers not only enhance ionic conductivity by disrupting polymer crystallinity but also reinforce mechanical integrity, allowing the electrolyte to resist volume deformation under cycling. Functionalized nanofillers additionally promote Li^+ coordination, facilitating ion hopping across polymer chains.

Crosslinking strategies—such as in situ polymerization or UV-induced gelation—enable uniform ion pathways and prevent phase separation, resulting in enhanced thermal and electrochemical stability [9,10]. Such modifications yield GPEs that remain flexible and conductive even under elevated temperatures, making them ideal for high-performance and high-voltage battery systems.

3.4 Recent Advancements and Functional Innovations

The next generation of gel polymer electrolytes (GPEs) has evolved beyond basic ionic transport materials to become multi-functional, adaptive systems that respond intelligently to mechanical and electrochemical stress. Recent developments have introduced self-healing polymer networks capable of autonomously repairing micro cracks and maintaining interfacial integrity during prolonged cycling, thereby extending electrolyte lifespan. In parallel, ionic liquid–based GPEs have gained prominence due to their high ionic conductivity, negligible volatility, and non-flammable nature, making them ideal for applications requiring both thermal stability and safety [2,9].

Further innovation has emerged through the development of hybrid solid–gel architectures, which merge the flexibility and interfacial compatibility of GPEs with the mechanical rigidity of solid electrolytes. These composite systems offer improved electrochemical stability and mechanical durability, particularly suited for high-energy-density devices such as electric vehicles and grid-scale batteries [10]. Complementing these material advances, AI-assisted material optimization has recently been employed to predict ideal polymer–salt–filler ratios, balance ionic mobility with mechanical strength, and accelerate formulation discovery through data-driven modelling [11].

Additionally, sodium-ion GPEs are gaining attention as sustainable and cost-effective alternatives for large-scale energy storage. Sodium-compatible polymer hosts with tailored side-chain chemistry have demonstrated efficient Na^+ transport and high thermal and chemical stability, supporting their potential in environmentally benign, grid-oriented applications [8].

3.5 Challenges and Future Directions

Despite rapid progress, several technical challenges limit the large-scale commercialization of GPEs:

Limited long-term stability due to solvent evaporation and polymer degradation over prolonged cycling. Reduced conductivity at low temperatures, restricting all-climate usability. Complex processing routes for large-area, defect-free membranes. Trade-offs between ionic conductivity and mechanical strength, which remain difficult to balance [9,10].Future development must focus on hybrid polymer frameworks, where covalently bonded inorganic clusters and flexible organic chains deliver both robustness and ion mobility. Advanced simulation models and operando characterization techniques such as EIS and Raman mapping will further elucidate ion transport mechanisms and guide molecular-level optimization [11].

In summary, Gel Polymer Electrolytes (GPEs) represent a cornerstone of safe, flexible, and high-performance energy storage systems. Through continuous innovation in polymer chemistry, Nano filler engineering, and hybrid design, GPEs are poised to play a central role in the commercialization of next-generation lithium- and sodium-ion batteries [2,5,8,9,10,11].

4. Nanostructured Functional Composites: Redefining Electrochemical Interfaces

The evolution of nanostructured functional composites represents a paradigm shift in the design of advanced battery materials, merging electrochemical performance with mechanical resilience and interfacial intelligence. These hybrid materials integrate inorganic nanophases within conductive polymer or carbon frameworks to create multifunctional architectures capable of concurrent ion transport, stress buffering, and electronic conduction [3,5,8,9,11]. By leveraging nanoscale control over morphology and composition, researchers have significantly enhanced active surface area, reduced charge-transfer resistance, and stabilized electrode–electrolyte interfaces — parameters that directly determine both energy density and lifetime in next-generation storage systems.

From a materials-design standpoint, Nano structuring transforms passive electrodes into dynamic, self-reinforcing systems. The large surface-to-volume ratio of nanomaterials accelerates ionic exchange and facilitates rapid electron percolation, while flexible polymer matrices accommodate volumetric strain during cycling. This synergy minimizes electrode pulverization and preserves structural integrity even under aggressive operating conditions a breakthrough that conventional microstructure materials cannot achieve [3,8].

From a fabrication standpoint, composite engineering enables unprecedented tunability. In systems such as manganese oxide (Mn_2O_3)–polymer nanocomposites, the inorganic phase provides electronic conductivity and structural stiffness, whereas the conductive copolymer imparts elasticity and uniform current distribution. The resulting hybrid maintains a stable solid–electrolyte interphase (SEI) and suppresses impedance build up during repeated cycling [3,5]. Parallel advances using graphene, carbon nanotubes, and amorphous carbon coatings have extended these benefits further by forming continuous electron pathways and distributing mechanical stress uniformly throughout the electrode matrix [8,9]. Functionalized Nano fillers chemically bond with polymer chains to create strong interfacial bridges, preventing delamination and phase separation. This nanoscale cohesion delivers remarkable improvements in both cycling life and rate capability.

From an integration perspective, nanostructured composites serve as the connective tissue between next-generation materials. Within hybrid battery architectures, they act as mechanically compliant interlayers that harmonize silicon–graphite anodes, gel polymer electrolytes (GPEs), and solid-state electrolytes (SSEs) [5,9]. Their ability to maintain continuous ionic pathways while mitigating mechanical mismatch ensures stable operation under high-energy, high-temperature conditions. This multifunctional adaptability is what differentiates nanocomposites from traditional fillers — they do not merely exist in the system; they *engineer the interface itself.*

From an industrial and research direction standpoint, emerging fabrication techniques such as sol–gel synthesis, electrospinning, and atomic layer deposition now allow precise manipulation of nanoscale interfaces. These methods enable hierarchical structures that combine porosity control, compositional uniformity, and surface functionalization. In parallel, machine-learning-driven materials discovery and computational interface modelling are redefining how researchers design these composites, cutting down experimental cycles and accelerating scalability [11].

In essence, nanostructured functional composites are no longer incremental improvements — they represent the convergence of chemistry, physics, and materials engineering into a single platform that intelligently balances energy density, safety, and durability. By enabling elastic interfaces, fast ion diffusion, and structural self-regulation, these composites are setting the foundation for the next generation of high-performance energy storage technologies [3,5,8,9,11].

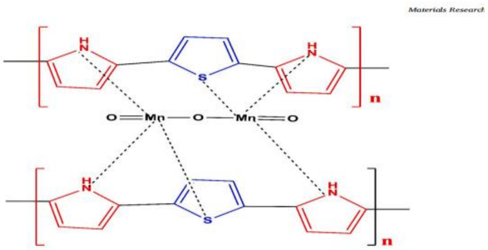


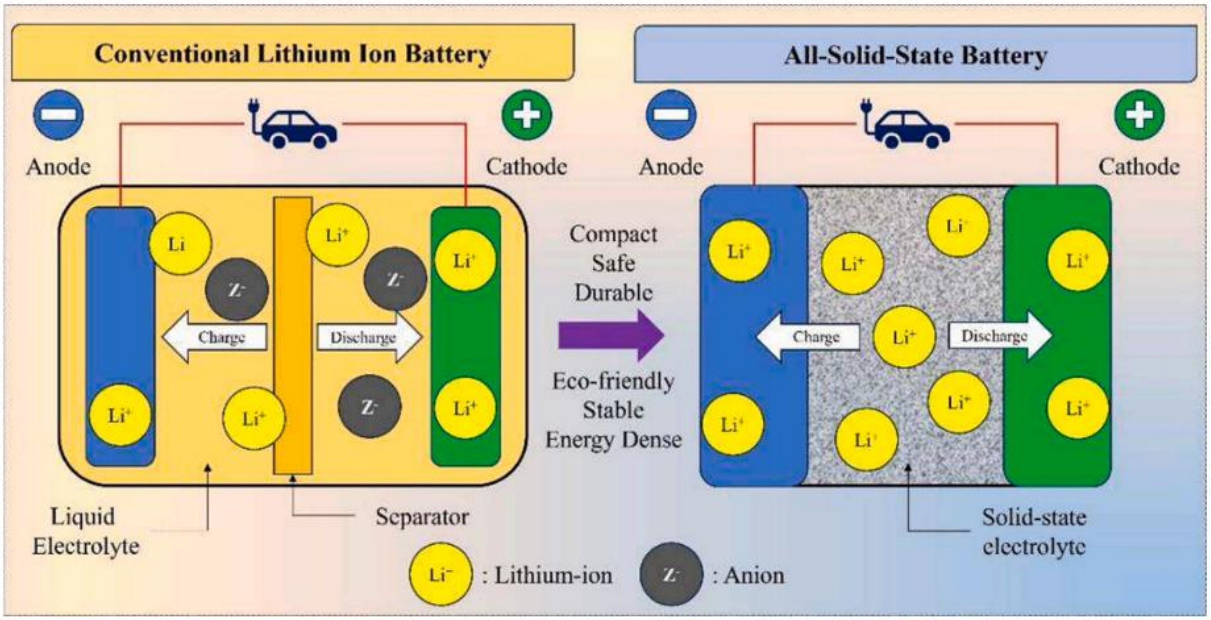
Figure 6.:
Illustration of Mn_2O_3/conductive polymer hybrid improving electrode–electrolyte interface stability. Adapted from [3].

These types of hybrids are particularly promising when paired with Si–graphite hybrids because they buffer volume change without sacrificing ionic transport. They also work well inside GPE matrices to enhance interface strength and stability [2,5,9].

5. Solid-State Batteries: Toward Safer and Higher-Energy Architectures

The global shift toward electrification and high-power mobility has intensified the need for energy storage systems that combine exceptional safety, high energy density, and prolonged cycle life. Among next-generation battery technologies, solid-state batteries (SSBs) have emerged as the most promising alternative to conventional liquid-electrolyte lithium-ion systems. Unlike liquid cells, where flammable organic solvents facilitate ion transport but risk thermal runaway and dendrite formation, SSBs employ **solid-state electrolytes (SSEs)** that enable safe, compact, and high-voltage operation

Figure 7.:
Comparison between conventional lithium-ion and solid-state battery architectures. Solid electrolytes replace the liquid component, enabling denser, safer cells. Adapted from [9]



5.1 Fundamentals and Design of Solid-State Electrolytes

From a materials-design perspective, the solid-state approach introduces a new dimension of electrochemical architecture. Solid electrolytes serve dual functions — ionic conductors and mechanical separators — providing both ion transport and dendrite suppression. Among the diverse electrolyte families, inorganic oxides, sulfides, and polymers represent the three major classes of SSEs, each offering distinct advantages and limitations.

As summarized in Figure, oxide-based SSEs (such as LLZO and LATP) exhibit excellent thermal and chemical stability sulfide-based electrolytes (such as $Li_{10}GeP_2S_{12}$*) provide superior ionic conductivity exceeding* 10^{-2} *S* cm^{-1}*, and polymer-based systems offer flexibility and improved electrode contact [8–10].* These materials collectively form the backbone of today's SSB research, balancing ionic mobility, process ability, and mechanical strength for next-generation energy devices.

5.2 Interface Engineering and Ion Transport Optimization

From a mechanistic standpoint, achieving stable and low-resistance interfaces between solid electrolytes and electrodes remains one of the defining challenges in SSB development. Unlike liquid systems that self-wet and conform, solid–solid interfaces suffer from poor physical contact, leading to voids, high interfacial impedance, and localized lithium deposition [8].To address these issues, researchers have developed advanced interfacial engineering strategies, including the introduction of buffer layers, atomic-scale coatings, and 3D structured electrode–electrolyte architectures. *As depicted in Figure 3, these designs improve interfacial contact, enhance ionic percolation pathways, and reduce charge-transfer resistance.* Typical coating materials such as Li_3PO_4, $LiNbO_3$, and Al_2O_3 have shown notable improvements in long-term cycling and impedance stability [9,10].

5.2.1 Polymeric Electrolytes

Polymer-based solid electrolytes are built on flexible organic backbones capable of solvating lithium or sodium salts. Common host matrices include PVDF-HFP, PEO, and PAN copolymers. These systems exhibit good mechanical flexibility, are easy to process, and ensure intimate electrode contact—a key advantage in mitigating interfacial resistance.

Polymer electrolytes can be categorized into two main classes:

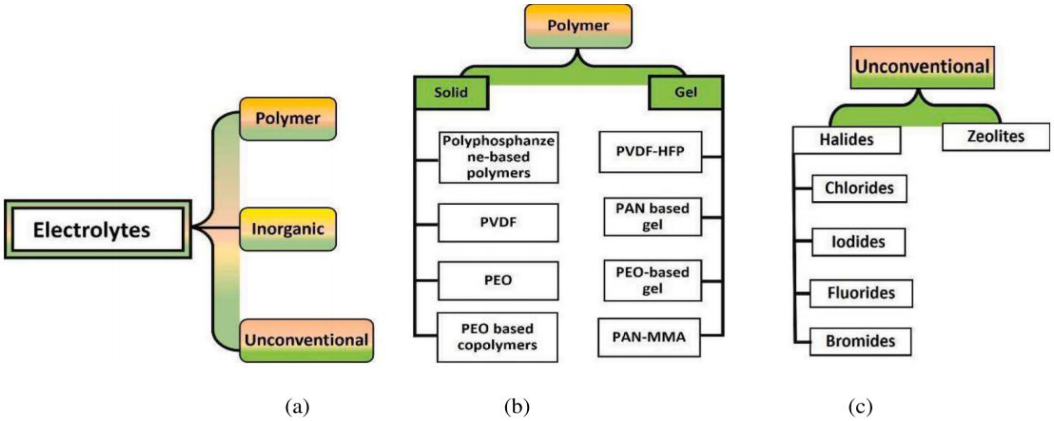


fig 8- Overview of Polymer, Inorganic, and Unconventional Electrolyte Systems

Dry solid polymers: systems such as PEO-LiX or PVDF-based materials, which rely on polymer segmental motion to enable ion conduction.

Gel polymer electrolytes (GPEs): hybrid systems containing a trapped liquid phase that enhances ionic conductivity. (See Section 3 for details on GPE performance.) The downside of polymer systems is their lower room-temperature conductivity compared to sulfides or oxides, which limits power performance. However, their process ability, cost-effectiveness, and compatibility with Si–graphite anodes make them ideal interlayers or hybrid components in commercial cells [2,5].

5.2.2 Inorganic Electrolytes

Inorganic solid electrolytes are currently viewed as the backbone of high-performance solid-state systems due to their superior ionic conductivity and chemical stability. They are generally divided into:

Sulfide-based electrolytes: e.g., $Li_{10}GeP_2S_{12}$ and argyrodite compounds. These materials offer liquid-like ionic conductivity (10^{-3}–10^{-2} S cm^{-1}), soft mechanical properties for good interface contact, and scalability potential. Their main limitation is moisture sensitivity, which complicates handling and processing.

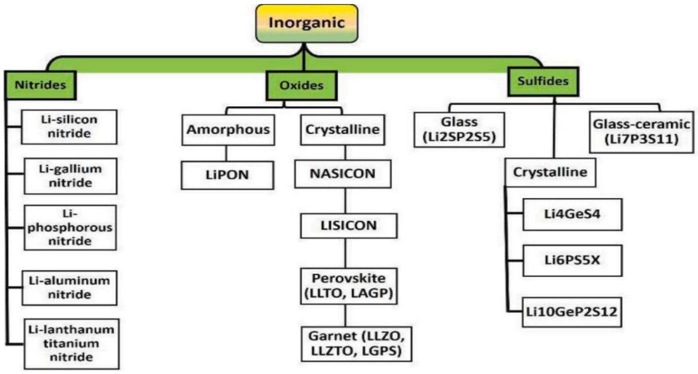
Oxide-based electrolytes: e.g., garnets (LLZO), NASICON, and perovskites. These offer high thermal and chemical stability but are brittle, making it difficult to maintain intimate electrode contact. Innovative sintering, coating, and pressure-assisted fabrication techniques are being explored to address these challenges.

Nitride-based electrolytes: e.g., LiPON films, which are thin and stable but typically lower in conductivity and more expensive to manufacture.Inorganic systems provide excellent stability at high voltages, which is crucial for pairing with both silicon–graphite anodes and advanced cathodes. However, achieving low-resistance interfaces remains one of the main engineering hurdles [9].

5.2.3 Unconventional Electrolytes

Alongside the mainstream polymeric and inorganic families, there is a growing interest in unconventional systems such as halides, zeolites, and composite structures.Halide electrolytes have recently attracted attention for their high electrochemical stability with high-voltage cathodes and excellent compatibility with moisture, unlike sulfides. Zeolite frameworks and other nanoporous structures allow ion-selective pathways, improving safety and offering tunable architectures for specific ion chemistries. Mixed-matrix systems, combining halides with polymers or oxides, are being designed to combine the mechanical flexibility of polymers with the high conductivity of inorganic frameworks [11].

These unconventional electrolytes are still largely in the research phase but are



(d)

fig 9- Classification of Inorganic Solid Electrolytes rapidly gaining momentum due to their versatility and potential to overcome existing limitations.

5.2.4 Hybrid and Composite Architectures

No single electrolyte family fully satisfies the competing demands of conductivity, stability, and manufacturability. Hybrid architectures — such as polymer/ceramic composites or multilayer stacks are emerging as a practical solution:
A ceramic backbone ensures high ionic conductivity and structural stability.
A polymer shell or interlayer improves mechanical compliance and electrode contact.
The combination improves overall cell performance, reduces interface resistance, and supports larger electrode loadings.
These composite strategies are now being integrated into pilot-scale production lines by several research groups and industrial players. Their compatibility with silicon–graphite anodes and interfacial nanocomposites makes them central to scalable solid-state designs [9,11].

5.2.5 Material Selection and Design Trade-offs

Each electrolyte class presents distinct engineering trade-offs:

Sulfides: best-in-class conductivity, sensitive to processing.
Oxides: excellent stability, but brittle.
Polymers: flexible and scalable, but lower conductivity.
Halides/Unconventional: emerging, promising balance of properties.

The choice of electrolyte is increasingly application-driven. For example, EV and grid applications prioritize high energy density and thermal safety, favoring sulfide or hybrid solid–polymer stacks, while wearable and flexible electronics may prefer pure polymer or polymer-rich hybrids due to form factor flexibility.

5.3 Fabrication Strategies and Industrial Translation

From a fabrication standpoint, the processing of SSBs has advanced from laboratory-scale cold pressing to scalable, industrially relevant techniques. Tape casting, spark plasma sintering, aerosol deposition, and co-lamination now allow large-area film formation and multilayer stack integration under controlled densification conditions [10,11]. These methods reduce interfacial defects, enable uniform ion conduction, and maintain mechanical strength at lower processing temperatures a critical factor for compatibility with existing electrode manufacturing lines.

From an integration standpoint, hybrid designs that combine silicon–graphite anodes, solid electrolytes, and gel polymer interlayers have shown exceptional promise. These semi-solid architectures maintain flexibility while achieving enhanced safety and energy density, effectively bridging the gap between liquid-based lithium-ion and fully solid-state platforms [5,9].

5.4 Technological Outlook and Industrial Feasibility

From a technological and industrial perspective, SSBs are rapidly transitioning from concept to commercialization. Several leading companies including Toyota, Solid Power, and Quantum Scape have demonstrated functional SSB prototypes with energy densities exceeding 400 Wh kg^{-1} and robust cycle performance [11]. Beyond lithium, efforts are expanding toward sodium- and magnesium-based solid-state systems that promise resource sustainability and cost-effectiveness.

Concurrently, the convergence of machine-learning-driven electrolyte design, operando electrochemical characterization, and predictive modeling of interfaces is accelerating SSB optimization. Techniques such as *operando EIS*, *synchrotron tomography*, and *molecular dynamics simulations* provide real-time insights into ion transport and degradation processes, bridging the gap between experimental observation and computational prediction [10,11].

In essence, solid-state batteries embody the next evolution of electrochemical energy storage — an architecture that simultaneously delivers safety, performance, and scalability. With continued innovation in interface chemistry, composite electrolyte engineering, and large-scale fabrication, SSBs are poised to redefine the future of portable electronics, electric vehicles, and grid-scale energy storage [2,5,8,9,10,11].

6. Characterization Techniques and Electrochemical Evaluation

Characterization forms the analytical foundation of battery research, linking material structure to electrochemical behaviour. For advanced hybrid and solid-state systems, detailed examination of phase composition, morphology, and interface chemistry reveals how atomic-level design translates into measurable performance.

6.1 Structural and Morphological Characterization

Structural characterization identifies the crystallographic and chemical features that define functional performance. X-ray diffraction (XRD) confirms crystallinity, phase purity, and the presence of secondary compounds in silicon–graphite anodes, nanocomposites, and solid electrolytes. Fourier-transform infrared spectroscopy (FTIR) provides complementary evidence of chemical bonding and polymer–filler interactions, such as characteristic C–O, C=O, and Si–O–Si vibrations in gel polymer electrolytes and oxide composites.

Morphological analysis through field-emission scanning electron microscopy (FESEM) and transmission electron microscopy (TEM) reveals particle distribution, porosity, and interfacial homogeneity. These techniques help evaluate how nanostructuring or surface modification influences electrode texture, interfacial contact, and potential diffusion pathways. Together, they establish a clear link between physical structure and electrochemical stability.

6.2 Electrochemical Characterization Electrochemical testing translates material architecture into quantitative performance indicators. Cyclic voltammetry (CV) measures redox reversibility and identifies potential-dependent reactions

Estimated ionic conductivity (σ_dc) values derived from bulk resistance (R_b) using different evaluation methods. (a) σ_dc values for PEO-salt systems. (b) σ_dc values for PMA-salt systems. The dotted lines indicate linear regressions. Data adapted from [8]

Within electrodes and electrolytes. Galvanostatic charge–discharge (GCD) profiles yield information on specific capacity, rate capability, and Coulombic efficiency, reflecting how well the active material sustains repeated cycling.

Electrochemical impedance spectroscopy (EIS) evaluates internal resistances and interfacial impedance. Variations in semicircle diameter or Warburg tail slope indicate the efficiency of ion transport and the stability of the solid–electrolyte interphase. Collectively, these electrochemical analyses provide a functional perspective that complements structural characterization, confirming whether observed microstructural refinements lead to tangible electrochemical benefits.

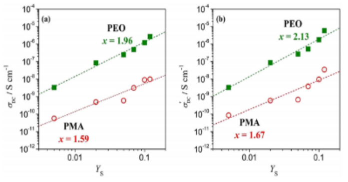


Figure 8: (a) σ_{DC} values estimated from R_b values and (b) imprecise σ_{DC}' values estimated from R_b' values for (■) PEO-salt and (O) PMA-salt systems. The dotted line represents the linear regressions after Equation (4).

Table 2: R_b and σ_{DC} values of PEO-LiClO$_4$ estimated using different approaches.

Approach used	PEO + 0.5 wt. % of LiClO$_4$ R_b (Ω) [σ_{DC} (S cm^{-1})]	PEO + 11 wt. % of LiClO$_4$ R_b (Ω) [σ_{DC} (S cm^{-1})]
Manual graphical	2.81×10^6 [3.32×10^{-9}]	2.41×10^3 [5.55×10^{-6}]
Mathematical regression	2.80×10^6 [3.33×10^{-9}]	2.40×10^3 [5.57×10^{-6}]
EIS software (Nova, 2018)	2.82×10^6 [3.31×10^{-9}]	2.49×10^3 [5.37×10^{-6}]

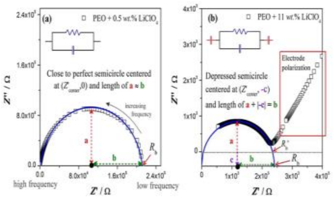


Figure 7: Nyquist plots for SPEs. (a) a close to perfect semicircle following Debye response, (b) a depressed semicircle with non-Debye response. The solid blue curve is the semicircle fitting of the impedance data estimated from Equation (3).

Figure 10:

Typical Nyquist plot illustrating high-frequency intercept (bulk resistance), semicircle (interfacial charge transfer), and low-frequency tail (diffusion or SEI behaviour). Adapted from [8].

6.3 In-Situ and Operando Analysis

In-situ and operando techniques capture the dynamic evolution of materials during actual operation. In-situ XRD tracks real-time lattice expansion, phase transitions, and structural degradation under cycling conditions. In-situ Raman spectroscopy monitors vibrational changes that reflect polymer segmental motion or electrode surface reactions. Operando EIS provides continuous impedance mapping, offering insight into aging behaviour and degradation mechanisms.

These approaches bridge the gap between laboratory characterization and device-level diagnostics, enabling researchers to correlate transient structural changes with electrochemical signatures. They also facilitate predictive modeling and early-stage failure detection, which are vital for industrial validation and large-scale deployment.

6.4 Data Correlation and Performance Interpretation

Integrating multiple characterization results allows comprehensive interpretation of structure–property–performance relationships. Crystallinity, surface roughness, and interfacial contact directly affect ionic conductivity, diffusion kinetics, and overall energy efficiency. Correlating XRD, FTIR, FESEM, and EIS data helps identify the dominant limiting mechanisms in hybrid systems, guiding further optimization in composition or processing. Such correlations also inform machine-learning models and computational frameworks used for predictive materials design, where experimental descriptors serve as input features for data-driven screening of new electrode–electrolyte pairs.

To strengthen the connection between electrochemical response and physical structure, it is crucial to combine complementary techniques that examine both dynamic electrical behavior and static material properties. While electrochemical impedance spectroscopy (EIS) provides frequency-dependent resistance data, it does not reveal structural or chemical evolution. Therefore, additional tools are employed in parallel to correlate impedance shifts with real-time physical changes, as summarized in table below Complementary Electrochemical & Structural Techniques While EIS provides a dynamic frequency response, it doesn’t give structural information. To correlate impedance behaviour with physical changes, other tools are used in parallel:

table 3- Characterization Techniques for Electrochemical Performance and Interfacial Analysis

Technique	What It Measures	Why It Matters in This Context
Galvanostatic cycling	Cycle stability, capacity retention	Tracks overall electrochemical performance under real conditions
Cyclic voltammetry (CV)	Electrochemical stability window, redox peaks	Assesses compatibility of electrolytes and interface reactions
X-ray diffraction (XRD)	Structural phase transitions	Identifies changes in electrode or electrolyte structure during cycling
Raman / FTIR spectroscopy	Chemical bonding, SEI composition	Detects formation of new phases or degradation pathways
SEM / TEM	Morphology and interface imaging	Reveals cracking, delamination, or uniformity of interfacial layers

By combining these techniques, researchers can directly correlate electrochemical signatures with underlying physical mechanisms, transforming raw measurements into mechanistic understanding. This multi-modal approach ensures that performance improvements are not merely observed but scientifically explained a crucial step toward designing durable, high-efficiency energy-storage systems.

6.5 Emerging Trends and Industrial Diagnostics

The next generation of characterization combines traditional tools with intelligent data interpretation. High-throughput spectroscopy, automated image analysis, and AI-based pattern recognition are transforming raw data into actionable insights. Industrial battery-management systems now incorporate embedded diagnostic modules that mimic laboratory characterization — miniature impedance sensors, temperature-mapping arrays, and voltage-response analytics enable real-time health monitoring.

This convergence of laboratory analytics and operational diagnostics marks a major step toward self-aware, predictive, and safer energy-storage platforms capable of continuous optimization throughout their service life.

6.6 Summary

Characterization and electrochemical evaluation transform empirical observation into quantifiable understanding. By integrating structural, morphological, and electrochemical insights with real-time diagnostics, researchers can trace every performance parameter back to its material origin. This holistic framework ensures that improvements in composition, interface engineering, or fabrication directly translate into higher capacity, stability, and safety for next-generation hybrid and solid-state batteries.

7. Challenges and Future Perspectives

Despite tremendous progress in hybrid and solid-state battery systems, including advances in silicon–graphite anodes, gel polymer electrolytes, and nanostructured composites, several *interrelated barriers* continue to limit commercial scalability. The translation from laboratory prototypes to industrial systems is hindered by a combination of interfacial instability, mechanical fragility, electrochemical

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degradation, and manufacturing complexity [11–13]. Overcoming these bottlenecks requires coordinated innovation in materials chemistry, interface engineering, and large-scale processing strategies, supported by predictive modeling and sustainable manufacturing practices [12,14].

7.1 Interfacial and Electrochemical Instability

One of the most persistent challenges lies in maintaining stable interfaces between electrodes and solid electrolytes. Even when bulk ionic conductivity is high, *electrode–electrolyte contact degradation* and *uncontrolled SEI evolution* often dominate overall impedance and restrict cycle life [12,15]. Sulfide- and oxide-based solid electrolytes exhibit reactivity toward metallic lithium and silicon-rich anodes, producing interphases that evolve dynamically under cycling conditions. This leads to impedance growth, polarization, and capacity fading. Mechanical stress caused by silicon volume expansion exacerbates these effects, promoting microcracking and delamination. Recent studies suggest that chemically stable, passivating interlayers, combined with self-healing GPE matrices, can mitigate these effects by accommodating mechanical strain and suppressing parasitic reactions [16,17]. Stabilizing electrode electrolyte interfaces through thermodynamically compatible formulations remains the key enabler for achieving reliable, high-performance solid-state batteries [12,15].

7.2 Mechanical Integrity and Manufacturing Scalability

Mechanical integrity directly impacts the long-term reliability and manufacturability of solid-state and hybrid systems. Brittle ceramic electrolytes and rigid interfaces are highly susceptible to fracture under repeated cycling or thermal stress, while flexible polymer electrolytes often lack the mechanical strength required for industrial lamination and roll-to-roll fabrication [12,17]. Advanced composite designs that combine ceramic fillers with polymer matrices can provide a balance between elasticity and modulus, allowing for defect-tolerant architectures suitable for large-area processing [14,16]. From a manufacturing perspective, scalable synthesis methods such as tape casting, solvent-free extrusion, and in-situ polymerization are emerging as viable solutions to replace laboratory-scale vacuum deposition [13,17]. However, ensuring structural homogeneity and interfacial adhesion across large-format cells remains an unsolved engineering problem. Integrating additive manufacturing and automated coating technologies may bridge this gap, offering precision and reproducibility critical for industrial deployment [12,17].

7.3 Sustainability, Energy Efficiency, and Green Transition

Beyond material performance, sustainable manufacturing and lifecycle impact are now at the center of solid-state battery research. The global shift toward low-carbon energy systems has intensified pressure to reduce greenhouse gas emissions from material synthesis and processing [13]. Conventional solid-state electrolyte fabrication often involves high-temperature sintering, solvent-intensive casting, or rare-metal precursors, all of which inflate energy demand and environmental footprint [14]. To align with future climate-neutral manufacturing targets, researchers are exploring low-temperature synthesis, aqueous processing routes, and biopolymer-based gel electrolytes that minimize toxic solvents and waste generation [12,15]. Incorporating renewable energy inputs during production and recycling critical elements such as lithium and cobalt will be essential for aligning solid-state technologies with net-zero policy frameworks [13,14].

7.4 Future Roadmap and Outlook

The next decade of solid-state battery innovation will be shaped by *synergistic progress in materials science, process engineering, and digital design frameworks*. Several studies forecast that future breakthroughs will depend on the convergence of mechanically optimized architectures with AI-driven material discovery and predictive modelling of electrochemical interfaces [15–17]. Roadmaps toward 2035 envision scalable, dendrite-free, and thermally stable systems that integrate smart diagnostics, automated manufacturing, and closed-loop recycling [16]. Interdisciplinary collaboration—uniting chemists, data scientists, and manufacturing engineers—will be crucial for closing the gap between experimental success and real-world adoption [12,13]. With continued focus on sustainable synthesis, reproducible interfaces, and intelligent optimization, hybrid and solid-state batteries are poised to evolve from promising prototypes to cornerstone technologies in the global transition toward electrified mobility and renewable energy integration [14–17].

8. Conclusion and Future Outlook

The exploration of silicon–graphite hybrid anodes, gel polymer electrolytes, nanostructured composites, and solid-state systems reveals that the future of energy storage lies not in individual breakthroughs but in the integration of multiple innovations. Each improvement, from stable interfaces to flexible polymers and nanoscale architectures, is part of a larger narrative that redefines how the world stores and consumes energy. Despite remarkable progress, this domain still contains untouched potential, especially in connecting the chemistry of advanced materials with industrial-scale functionality. The interdependence between electrochemical performance, mechanical adaptability, and environmental responsibility remains a frontier that demands deeper and more coordinated exploration.

As the global energy landscape shifts toward electrified mobility and renewable power generation, the urgency of developing sustainable, high-performance storage systems has never been greater. Batteries now serve as the backbone of this transformation, bridging the intermittency of solar and wind energy while enabling the transition away from fossil-fuel dependency. Hybrid and solid-state technologies, with their superior stability and energy density, are poised to play a defining role in this global transition. Their success, however, depends on overcoming the challenges that separate laboratory achievement from real-world implementation.Industrial scalability and manufacturability remain among the most critical hurdles. Traditional fabrication methods must evolve into precision-controlled, data-optimized processes capable of ensuring reproducibility and reducing material waste. Emerging solutions such as additive manufacturing, solvent-free processing, and in-situ polymerization have the potential to transform production efficiency while maintaining structural integrity. The integration of digital manufacturing tools and automated diagnostics will mark a new era where materials discovery, prototyping, and production are tightly interlinked.

Equally important is the rise of advanced diagnostics and intelligent characterization. Real-time, in-situ analysis of interfacial chemistry and structural evolution offers unprecedented insight into failure mechanisms and long-term performance. When coupled with machine learning and predictive modelling, these diagnostics evolve from observation to optimization enabling the design of materials and devices that adapt, heal, and evolve with use. This convergence of analytics and materials science signifies a paradigm shift from reactive to proactive battery design.The future of energy will not be shaped by chemistry alone, but by conviction the belief that scientific progress can coexist with environmental responsibility. Every ion that moves within a cell carries more than electric charge; it carries the shared intent of humanity to build cleaner cities, sustainable industries, and a more resilient planet. The continuing evolution of hybrid and solid-state batteries reflects this transformation in thought where materials science, data-driven design, and sustainable engineering converge to redefine what energy truly means. The challenge before us extends far beyond storing power efficiently; it is about reimagining how civilization generates, conserves, and values it. When that vision is realized, and every home, vehicle, and network runs on the silent precision of these advanced systems, it will stand not only as an engineering achievement but as a defining chapter in how science and humanity learned to progress together.

REFERENCES :

1. High-Performance Dual-Ion Battery Based on Silicon–Graphene Composite Anode and Expanded Graphite Cathode Guoshun Liu † , Xuhui Liu † , Xingdong Ma, Xiaoqi Tang, Xiaobin Zhang, Jianxia Dong, Yunfei Ma, Xiaobei Zang, Ning Cao and Qingguo Shao https://doi.org/10.3390/molecules28114280

2. Smart gel polymer electrolytes enlightening high safety and long life sodium ion batteries Li Du1,2,3,4, Gaojie Xu 1,2,3 Chenghao Sun1,2,3, Yu-Han ZhangLang Huang1,2,3 https://doi.org/10.1038/s41467-025-57964-7

3. Manganese (III) oxide-infused poly(thiophene-*co*-pyrrole) nanocomposites for optical, dielectric, and photocatalytic applications S. Sankar , M.T. Ramesan Centre for Polymer Science and Technology, Department of Chemistry, University of Calicut, Calicut University P.O., Malappuram, Kerala 673 635, India https://doi.org/10.1016/j.materresbull.2025.113649

4. A Brief Overview of Silicon Nanoparticles as Anode Material:A Transition from Lithium-Ion to Sodium-Ion BatteriesAlireza Fereydooni, Chenghao Yue, and Yimin Chao The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/smll.202307275

5. .Multifunctional Nanostructured Conductive Polymer Gels: Synthesis,Properties, and Applications Fei Zhao,†,§ Ye Shi,†,§ Lijia Pan,*,‡ and Guihua Yu*,† Materials Science and Engineering Program and Department of Mechanical Engineering, The University of Texas at Austin, Austin,Texas 78712, United States Collaborative Innovation Center of Advanced Microstructures, School of Electronic Science and Engineering, Nanjing University, Nanjing 210093, China DOI:10.1021/acs.accounts.7b00191 Acc. Chem. Res. 2017, 50, 1734−1743

UNDER PEER REVIEW

6. Advancements in Graphite Anodes for Lithium-Ion and Sodium-Ion Batteries Kai Xiong | Tianshuang Qi | Xiong Zhang State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology https://doi.org/10.1002/elan.202400318

7. The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries Jingxing Wu1 | Yinliang Cao3 | Haimin Zhao3 | Jianfeng Mao1 | Zaiping Guo1,2 DOI: 10.1002/cey2.2

8. Basics of teaching electrochemical impedance spectroscopy of electrolytes for ion-rechargeable batteries – part 1: a good practice on estimation of bulk resistance of solid polymer electrolytes https://doi.org/10.1515/cti-2020-0011 Received June 13, 2020; accepted February 19, 2021; published online March 8, 2021

9. A comprehensive review of solid-state batteries Aniruddha Joshi a , Dillip Kumar Mishra a,* , Rajendra Singh b , Jiangfeng Zhang a , Yi Ding c a Department of Automotive Engineering, Clemson University, SC, USA b Department of Electrical and Computer Engineering, Clemson University, SC, USA c Ground Vehicle Systems Center, U.S. Army Combat Capabilities Development Command, USA https://doi.org/10.1016/j.apenergy.2025.125546

10. Research progress in the preparation of sodium-ionbattery anode materials using ball milling Liwen Zhang,†a Shandong Huang,†a Yihong Ding *a and Tianbiao Zeng*bDOI: 10.1039/d4ra08061k

11. Battery Research and Innovation—A Study of Patents and Papers Hans Pohl 1,* and Måns Marklund https://doi.org/10.3390/wevj15050193 Li C, et al. An advance review of solid-state battery: Challenges, progress and prospects. Elsevier BV Sep. 01, 2021. https://doi.org/10.1016/j.susmat.2021. E00297.

12. Xia S, Wu X, Zhang Z, Cui Y, Liu W. Practical Challenges and Future Perspectives of All-Solid-State Lithium-Metal Batteries. Elsevier Inc Apr. 11, 2019. https://doi.org/10.1016/j.chempr.2018.11.013.

13. Magento T, Bhoyate SD, de Souza FM, Mensah-Darkwa K, Kumar A, Gupta RK.Developing practical solid-state rechargeable Li-ion batteries: Concepts,challenges, and improvement strategies. Elsevier Ltd Nov. 25, 2022. https://doi.org/10.1016/j.est.2022.105688
Kim JG, et al. A review of lithium and non-lithium based solid state batteries. Elsevier BV May 15, 2015.https://doi.org/10.1016/j.jpowsour.2015.02.054.Battisti L. Energy, power, and greenhouse gas emissions for future transition scenarios. Energy Policy Aug. 2023;179. https://doi.org/10.1016/j.Enpol.2023.113626

14. Slamerˇsak A, Kallis G, O'Neill DW. Energy requirements and carbon emissions for a low-carbon energy transition. Nat Commun Dec. 2022;13(1). https://doi.org/10.1038/s41467-022-33976-5

15. Mauger A, Armand M, Julien CM, Zaghib K. Challenges and issues facing lithium metal for solid-state rechargeable batteries. Elsevier B.V; 2017. https://doi.org/10.1016/j.jpowsour.2017.04.018

16. Yim CH, Houache MSE, Baranova EA, Abu-Lebdeh Y. Understanding key limitingfactors for the development of all-solid-state-batteries. Chemical Engineering Journal Advances Mar. 2023;13. https://doi.org/10.1016/j.ceja.2022.100436.

17. Wu D, Wu F. Toward better batteries: Solid-state battery roadmap 2035+.Elsevier BV Apr. 01, 2023. https://doi.org/10.1016/j.etran.2022.100224.

18. Koerver R, et al. Chemo-mechanical expansion of lithium electrode materials-on the route to mechanically optimized all-solid-state batteries. Energy Environ Sci Aug. 2018;11(8):2142–58. https://doi.org/10.1039/c8ee00907d\

19. Z. Zeng, V. Murugesan, K. S. Han, X. Jiang, Y. Cao, L. Xiao, X. Ai,H. Yang, J.-G. Zhang, M. L. Sushko, J. Liu, "Non-flammable electro lytes with high salt-to-solvent ratios for Li-ion and Li-metal batteries"Nature Energy 3 (2018): 674–681, https://doi.org/10.1038/s41560-018-0196-y

20. 43. Y. X. Yao, X. Chen, C. Yan, X. Q. Zhang, W. L. Cai, J. Q. Huang, Q.Zhang, "Regulating Interfacial Chemistry in Lithium-Ion Batteries by aWeakly Solvating Electrolyte**", Angewandte Chemie InternationalEdition 60 (2020): 4090–4097, https://doi.org/10.1002/anie.202011482

21. 44. Q. Zheng, Y. Yamada, R. Shang, S. Ko, Y.-Y. Lee, K. Kim, E.Nakamura, A. Yamada, "A cyclic phosphate-based battery electrolytefor high voltage and safe operation", Nature Energy 5 (2020): 291–298, https://doi.org/10.1038/s41560-020-0567-z

22. 45. J. Hou, L. Lu, L. Wang, A. Ohma, D. Ren, X. Feng, Y. Li, Y. Li, I.Ootani, X. Han, W. Ren, X. He, Y. Nitta, and M. Ouyang, "Thermalrunaway of Lithium-ion batteries employing LiN(SO2F)2-based concen□trated electrolytes", Nature Communications 11 (2020): 5100, https://doi.org/10.1038/s41467-020-18868-w

23. 46. J. Choi, H. Jeong, J. Jang, A. R. Jeon, I. Kang, M. Kwon, J. Hong,M. Lee, " Weakly Solvating Solution Enables Chemical Prelithiation ofGraphite–SiOxAnodes for High-Energy Li-Ion Batteries", Journal of the American Chemical Society 143 (2021): 9169–9176, https://doi.org/101021/jacs.1c03648.

24. 47. B. Nan, L. Chen, N. D. Rodrigo, O. Borodin, N. Piao, J. Xia, T.Pollard, S. Hou, J. Zhang, X. Ji, J. Xu, X. Zhang, L. Ma, X. He, S. Liu,H. Wan, E. Hu, W. Zhang, K. Xu, X. Q. Yang, B. Lucht, C. Wang,"Enhancing Li+ Transport in NMC811j jGraphite Lithium-Ion Batteries at Low Temperatures by Using Low-Polarity-Solvent Electro□lytes" Angewandte Chemie International Edition 61 (2022): e202205967, https://doi.org/10.1002/anie.202205967

25. 48. Y. Zou, Z. Ma, G. Liu, Q. Li, D. Yin, X. Shi, Z. Cao, Z. Tian, H. Kim,Y. Guo, C. Sun, L. Cavallo, L. Wang, H. N. Alshareef, Y. K. Sun, and J.Ming, "Non-Flammable Electrolyte Enables High-Voltage and WideTemperature Lithium-Ion Batteries with Fast Charging", Angewandte Chemie International Edition 62 (2023): e202216189, https://doi.org/101002/anie.202216189

26. Q. Cai, H. Jia, G. Li, Z. Xie, X. Zhou, Z. Ma, L. Xing, W. Li,"Outstanding performances of graphite j jNMC622 pouch cells enabledby a non-inert diluent",

UNDER PEER REVIEW

Journal of Energy Chemistry 81 (2023): 593602, https://doi.org/10.1016/j.jechem.2023.02.044

27. 50. X. Xu, X. Yue, Y. Chen, and Z. Liang, “Li Plating Regulation onFast-Charging Graphite Anodes by a Triglyme-LiNO3 SynergisticElectrolyte Additive”, Angewandte Chemie International Edition62 (2023): e202306963, https://doi.org/10.1002/anie.202306963

28. 51. R. Huang, G. Wei, X. Zhou, J. Zhu, X. Pan, X. Wang, B. Jiang, X. Wei, and H. Dai, “Targeting the low-temperature performancedegradation of lithium-ion batteries: A non-destructive bidirectionalpulse current heating framework”, Energy Storage Materials 65 (2024):103173, https://doi.org/10.1016/j.ensm.2024.103173

29. W. Cai, C. Yan, Y. X. Yao, L. Xu, X. R. Chen, J. Q. Huang, Q.Zhang, “The Boundary of Lithium Plating in Graphite Electrode for Safe Lithium-Ion Batteries”, Angewandte Chemie International Edition 60 (2021): 13007–13012, https://doi.org/10.1002/anie.202102593

30. 54. G. Song, Z. Yi, F. Su, L. Xie, Z. Wang, X.-X. Wei, G. Xu, C.-M.Chen, “Boosting the Low-Temperature Performance for Li-Ion Batteries in LiPF -Based Local High-Concentration Electrolyte”, ACS Energy Letters 8 (2023): 1336–1343, https://doi.org/10.1021/acsenergylett.2c02903

31. 55. S. Liu, B. Gu, Z. Chen, R. Zhan, X. Wang, R. Feng, Y. Sun,“Suppressing dendritic metallic Li formation on graphite anode under battery fast charging”, Journal of Energy Chemistry 91 (2024): 484–500, https://doi.org/10.1016/j.jechem.2024.01.009

32. 56. P. Roy, S. K. Srivastava, “Nanostructured anode materials forlithium ion batteries”, Journal of Materials Chemistry A 3 (2015): 2454–2484, https://doi.org/10.1039/C4TA04980B

33. 57. B. P. Thapaliya, A. S. Ivanov, H.-Y. Chao, M. Lamm, M. Chi, H. M.Meyer, X.-G. Sun, T. Aytug, S. Dai, and S. M. Mahurin, “Molten saltelectrochemical upcycling of CO2 to graphite for high performancebattery anodes”, Carbon 212 (2023): 118151, https://doi.org/10.1016/j.carbon.2023.118151

34. 58. R. Z. L. Zhu, “Impacts of graphite morphology on rate performanceof Li-ion battery”, Batteries 48 (2018): 113–116, https://link.cnki.net/doi/10.19535/j.10011579.2018.02.012

35. J. A.-R. N. A. Laziz, A. Darwiche, J. Toufaily, A. Outzourhit, F.Ghamouss, and M. T. Sougrati, “Li- and Na-ion Storage Performance ofNatural Graphite via Simple Flotation Process”, J. Electrochem. Sci. Technol 9 (2018): 320–329, https://doi.org/10.5229/JECST.2018.9.4.320

36. 60. H. M. H. D. K. Naranpanawa, T. H. N. G. Amaraweera, N. W. B.Balasooriya, A. N. B. Attanayake, and H. W. M. A. C. Wijayasinghe,“Development of vein graphite by optimizing the NaOH concentrationin alkali roasting-acid leaching process for the anode application inrechargeable Li-ion batteries”, Ionics 29 (2022): 129–144, https://doi.org/10.1007/s11581-022-04819-6.

37. A. Lähde, J. Välikangas, A. Meščeriakovas, T. Karhunen, S.-M.Meščeriakovė, O. Sippula, S. Leinonen, U. Lassi, J. Jokiniemi, “Effect ofhigh temperature thermal treatment on the electrochemical perform□ance of natural flake graphite”, Journal of Materials Research 39 (2024):944–954, https://doi.org/10.1557/s43578-024-01282-z.

38. 62. M. Ko, S. Chae, J. Ma, N. Kim, H.-W. Lee, Y. Cui, J. Cho, “Scalablesynthesis of silicon-nanolayer-embedded graphite for high-energylithium-ion batteries”, Nature Energy 1 (2016): 16113, https://doi.org/10.1038/nenergy.2016.113.

39. E. M. K. H. Huang, and J. Schoonman, “Graphite–metal oxidecomposites as anode for Li-ion batteries”, Journal of Power Sources 97(2001): 114–117, https://doi.org/10.1016/S0378-7753(01)00597-3

40. J. Sun, G. Zheng, H.-W. Lee, N. Liu, H. Wang, H. Yao, W. Yang, Y.Cui, “Formation of Stable Phosphorus–Carbon Bond for Enhanced Performance in Black Phosphorus Nanoparticle–Graphite CompositeBattery Anodes”, Nano Letters 14 (2014): 4573–4580, https://doi.org/10.1021/nl501617j

41. L. Wang, Y. Zhu, C. Guo, X. Zhu, J. Liang, Y. Qian, “Ferricchloride-Graphite Intercalation Compounds as Anode Materials for Li-ion Batteries”, Chem Sus Chem 7 (2013): 87–91, https://doi.org/101002/cssc.201300874

42. N. Kim, S. Chae, J. Ma, M. Ko, and J. Cho, “Fast-charging high energy lithium-ion batteries via implantation of amorphous silicon nanolayer in edge-plane activated graphite anodes”, Nature Communications 8 (2017): 812, https://doi.org/10.1038/s41467-017-00973-y

43. 68. P. Li, J.-Y. Hwang, and Y.-K. Sun, “Nano/Microstructured Silicon–

44. Graphite Composite Anode for High-Energy-Density Li-Ion Battery”,ACS Nano 13(2019):2624–2633, https://doi.org/10.1021/acsnan9b00169

45. D. Ruan, L. Wu, F. Wang, K. Du, Z. Zhang, K. Zou, X. Wu, and G.Hu, “A low-cost silicon-graphite anode made from recycled graphite of spent lithium-ion batteries”, Journal of Electroanalytical Chemistry 884 (2021): 115073, https://doi.org/10.1016/j.jelechem.2021.115073

46. S. Xiao, X. Li, T. Li, Y. Xiang, J. S. Chen, “ Practical strategies forenhanced performance of anode materials in Na+/K+-ion batteries”,Journal of Materials Chemistry A 9 (2021): 7317–7335, https://doi.org/10.1039/D0TA12417F

47. 71. Y. Jiang, Z. Q. Niu, G. Offer, J. Xuan, H. Z. Wang, “Insights into theRole of Silicon and Graphite in the Electrochemical Performance ofSilicon/Graphite Blended Electrodes with a Multi-Material PorousElectrode Model”, Journal of the Electrochemical Society 169 (2022): 020568, https://doi.org/10.1149/1945-7111/ac5481

48. 72. Y. Kim, G. Park, J.-C. An, H.-J. Sun, J. Shim, “Chemical Deposition of Tin Nanoparticles on Highly Porous Carbon-Coated Graphite and Its Electrochemical Characterization as an Anode for Lithium Ion Batteries”, Transactions on Electrical and Electronic Materials 22(2021): 603–611, https://doi.org/10.1007/s42341-021-00352-x

UNDER PEER REVIEW

49. 73. Y.-F. Huang, Y.-C. Yang, Y.-Y. Tseng, H.-Y. Tuan, “Two dimen□sional MnPSe3 layer stacking composites with superior storageperformance for alkali metal-ion batteries”, Journal of Colloid andInterface Science 635 (2023): 336–347, https://doi.org/10.1016/j.jcis.2022.12.082.

50. W. Yan, Z. Chen, S. Ma, S. Chen, Y. Lu, M. Wang, L. Chen, Q.Huang, B. Wang, Y. Su, J. Wang, N. Li, and F. Wu, “Unraveling the relationship between the mineralogical characteristics and lithium storage performance of natural graphite anode materials”, Carbon 227(2024): 119270, https://doi.org/10.1016/j.carbon.2024.119270