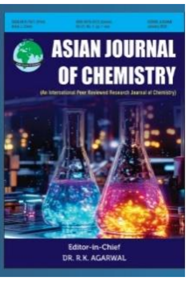
Asian Journal of Chemistry; Vol. 37, No. 10 (2025), 2342-2354

ASIAN JOURNAL OF CHEMISTRY

https://doi.org/10.14233/ajchem.2025.34419



REVIEW

Perovskite Anode-Based Gel Polymer Electrolytes for High-Performance Sodium-Ion Batteries: Design, Electrochemical Performance and Stability Analysis

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Received: 7 July 2025 *Accepted*: 19 August 2025 *Published online*: 30 September 2025 AJC-22117

Interest in sodium-ion batteries as an alternative to lithium-ion systems, especially stationary systems, has arisen due to the demand of safe, sustainable and scalable energy storage. The anode material and the composition of the electrolyte are some of the key aspects that define the performance of sodium-ion batteries. This review reports the integration of perovskite-based anode materials with gel polymer electrolytes (GPEs) to enhance the overall efficiency, safety and cyclability of sodium-ion batteries. Perovskite anodes exhibit promising characteristics for sodium storage due to their tunable redox-active sites, structural flexibility and high theoretical capacity. However, their practical application remains limited by challenges such as interfacial degradation, low ionic mobility and phase instability during electrochemical cycling. Compared to conventional liquid electrolytes, GPEs offer improved safety, enhanced thermal stability, superior ionic conductivity and better compatibility with electrode interfaces. This review also systematically examines the electro-chemical behaviour of perovskite anodes, recent advancements in GPE formulations and the synergistic interactions at the electrode–electrolyte interface. It also looks at the constraints, *e.g.* interfacial mismatches and structural degradation and optimization approaches, *e.g.* interfacial engineering, nanocomposite design and polymer modification. The study concludes by identifying future research and development directions, emphasizing scalable synthesis methods, environmental sustainability and the techno-economic viability of commercial deployment of sodium-ion batteries.

Keywords: Sodium-ion batteries, Perovskite anodes, Gel polymer electrolytes, Electrode electrolyte interface, Energy storage materials.

INTRODUCTION

The global transition toward sustainable energy sources has intensified research into next-generation energy storage systems, with sodium-ion batteries (SIBs) emerging as a promising alternative to lithium-ion batteries (LIBs) due to their abundant raw materials, more balanced geographical distribution and greater cost-effectiveness [1-3]. Although lithium-ion batteries (LIBs) currently dominate the energy storage market, concerns related to safety risks, rising material costs and the uneven global distribution of resources have highlighted their limitations. As a result, sodium-based chemistries are gaining increasing attention, particularly for large-scale and stationary energy storage applications [4,5]. The geographical and economic accessibility of sodium as a geographically diversified mineral and its affordability puts SIBs as the solution to economical energy storage on a large scale. Nevertheless, in spite of these advantages, sodium ions possess a bigger ionic radius than lithium, which is a drawback like slow diffusion kinetics and unfavourable intercalation characteristics. This, in its turn, influences the volumetric energy density and stability of electrode materials, thus requiring a substantial material innovation [3,6]. Such drawbacks require the development of new anode and electrolyte materials with improved electrochemical performance, thermal stability and cyclability of SIBs [7,8].

The anode is the most important part that defines the efficiency and cycle life of sodium-ion batteries. Conventional anode materials such as graphite and hard carbon face several limitations, including low initial Coulombic efficiency, insufficient electronic conductivity and mechanical degradation during cycling due to significant volume changes [6,9,10]. To address these issues, scientists have focused on other materials such as metal oxides, sulfides, phosphides and their combinations, in order to attain high capacity, improved cycling life and safety [11,12]. In this respect, perovskite-based materials, which can be described by the general formula ABX 3, have

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received significant interest due to their structural plasticity, extensive redox chemistry and adjustable physico-chemical characteristics [13,14]. They can make diverse substitutions at the A- or B-sites such that the researchers can customize parameters like ionic mobility, mechanical stability and electronic conductivity [15-17]. Research on $Na_{0.5}Bi_{0.5}TiO_3$ and related perovskite materials have demonstrated promising sodium ion intercalation capacity, along with favourable thermal stability and electrochemical reversibility, positioning them as potential next-generation anode materials for sodium-ion batteries [9,18,19]. However, the interaction between the anode (perovskite) and the electrolyte has a great effect on the performance of such practical systems. There are conventional liquid electrolytes usually containing sodium salts in organic solvents that have high ionic conductivity at the cost of great disadvantages such as volatility, flammability, dendritic sodium growth and small electrochemical windows that reduce both reliability and safety [9,20]. In order to curb these problems solid-state and gel polymer electrolytes (GPEs) have been suggested as safer and more stable alternatives.

Gel polymer electrolytes (GPEs) are a type of hybrid electrolyte, which unites advantages of both liquid and solid electrolytes *i.e.* high ionic conductivity of liquids and mechanical strength of solids. Such systems can be synthesized by immobilizing a liquid electrolyte in a polymer matrix, which increases electrode-electrolyte contact, lowers interfacial resistance and inhibits the dendrite growth [11,21,22]. Sodium-ion batteries are especially well-suited to such quasi-solid-state architectures, as safety and stability are important parameters of commercialization. Furthermore, compared with conventional liquid electrolytes, the thermal and electrochemical stabilities of GPEs can withstand the number of volumetric changes that occur during charge-discharge cycling and therefore experience higher total cycling life and energy efficiency [23-25]. Development Innovation in GPE development, including *in situ* crosslinking, dual-network structures, the inclusion of ionic liquids and surface modification approaches, has additionally enhanced the mechanical integrity and ion transport characteristics of those electrolytes [12,14,26].

The potential synergy between perovskite-based anodes and high-performance gel polymer electrolytes (GPEs) warants particular attention, as their integration could effectively address critical limitations in sodium-ion batteries, including interfacial instability, poor ionic transport and limited cycling durability. Although both materials have been advanced individually, they are not well explored when combined. Theoretical and preliminary experimental works indicate that the combination of perovskite-based electrodes and GPEs may result in an improved interfacial compatibility, an accelerated sodium transport and the dendritic growth suppression, which are some of the most problematic aspects of SIB development [16,20,27]. However, such a combination also open new issues at the interface level such as phase mismatch, poor adhesion and interfacial instability all of which can degrade the long-term electrochemical performance [17,28]. Such interface issues require a deeper study of interface design and engineering. Surface functionalization, buffer layer incorporation and redox mediator additives are some of the techniques that have been suggested to reduce degradation, enhance ion diffusion and sustain long-term cycling integrity [22,29,30]. Moreover, the addition of ether-type solvents and fluorinated materials as well as ion conductive fillers into GPE matrix has been reported to improve electrochemical stability, safety and compatibility with sodium metal or sulfur-based chemistry [10,26,31]. Although such innovations have shown encouraging results at the laboratory scale, they need to be scaled into scalable manufacturing procedures to enter into commercial production. The significant issues are the high cost of synthesis of perovskite compounds, the scalability of the polymer processing, the sensitivity of some of the GPE components to the environment and the challenge of incorporating such systems into the existing battery manufacturing lines [19,27,32]. Therefore, it is crucial to achieve a multi-disciplinary-buoyant approach to SIB technology by ensuring materials design, electrochemical engineering and manufacturability are integrated, to break the limits of SIB technology. The combination of perovskite-based anodes with GPEs does not only overcome the drawbacks of the conventional SIB chemistries but also follows the general trend towards all-solid-state and intrinsically safe battery designs [11,16,24]. Similar advances in characterization methods, including operando spectroscopy, imaging and computational modelling and machine learning-directed material informatics are making it possible to better extrapolate interfacial behaviour, ionic pathways and degradation mechanisms [32,33]. Also, the world move towards sustainable energy storage is not only based on the technical benefits but also on geopolitical and environmental factors. The sodium-ion batteries that use earth-abundant, non-toxic and widely distributed raw materials provide a fair and sustainable energy storage system. With the increasing number of countries aiming to achieve carbon neutrality, electrify transport and install decentralized energy storage to integrate renewable energy, the need to have cheap and sustainable batteries is constantly growing [27,34,35].

The synergistic design of perovskite anodes and GPEs is a critical area of sodium-ion batteries in the next generation in this changing scenario. The combination of the two classes of materials has the potential of rewriting the criteria of energy density, cycling life and safety in sodium-based energy storage. This work aims to provide a comprehensive and critical analysis of recent advancements in perovskite-based anodes and gel polymer electrolytes (GPEs), highlighting the key scientific and engineering challenges associated with their integration, as well as potential pathways for future development. The current work fills the gap between the concepts of materials chemistry, electrochemistry and battery engineering and thus, contributes to the roadmap of developing safe, scalable and efficient sodium-ion energy storage systems that will be able to meet the current and future energy needs.

Fundamentals of perovskite anode materials

Structure and properties of perovskites: The perovskite materials are a wide range of compounds, usually expressed as ABX_3, in which the A-site is generally occupied by large monovalent or divalent cations (*e.g.* Na^+, Bi^{3+}), the B-site by smaller transition metal cations (*e.g.* Ti^{4+}, Ni^{2+}) and the X-site by anions such as oxygen or fluorine. This structure type creates a three-dimensional network of corner-sharing BX_6 octahedra and the A cation occupies the 12-coordinated cuboctahedral holes

(Fig. 1). It depends on the ionic radius which determines the geometric compatibility and stability of this structure by the use of the Goldschmidt tolerance factor [36]. Perovskite materials that have been extensively engineered are frequently the most efficient halide perovskites. Their exceptional performance is ascribed to excellent light absorption, low charge recombination rates, long carrier diffusion lengths, benign defect states and tunable characteristics [37].

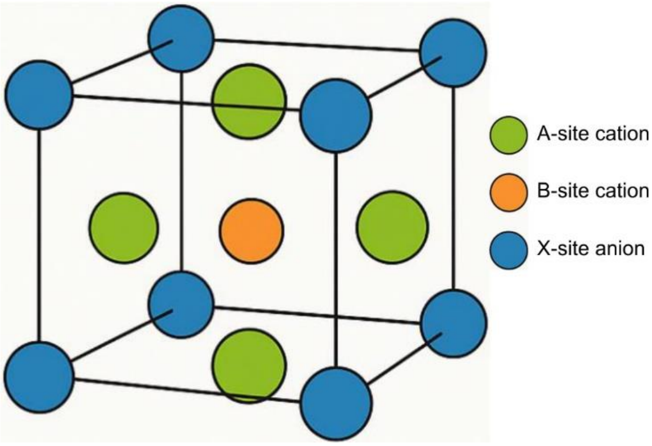

Fig. 1. Perovskite structure (ABX_3) showing A-site cations (green), B-site cations (orange) and X-site anions (blue), ideal for sodium-ion battery anodes

This architecture allows large compositional tunability where researchers could manipulate important properties like ionic conductivity, electronic transport and redox properties. The combination of these properties renders perovskites a good choice to be applied in electrochemical devices, particularly sodium-ion batteries (SIBs) that require sturdy, stable and ion-conductive anode structures [38]. This flexibility to accommodate multiple oxidation states and to accommodate multi-electron redox reactions, by virtue of the ability to replace ions at the A or B site without disturbing the crystalline backbone, makes perovskites capable of supporting charge storage capacities [39]. Besides, their open structures are highly compatible with accommodating large sodium ions as compared to their rigid structures. In perovskites, ionic channels and the interstitial holes allow the diffusion of Na^+ during charging and discharging and also provide better thermal stability and structural deformation tolerance [40].

Benefits of perovskite anode in SIBs: Anodes made of perovskites have attractive advantages compared to conventional materials used to make anodes in SIBs (carbon). These benefits are increased structural and thermal stability, expanded redox windows and enhanced electrochemical reversibility. The perovskites are less prone to mechanical degradation undergoing Na^+ insertion/extraction than in carbonaceous anodes classes [41]. New progress in doping and structural optimization has resulted in the high-rate capability perovskite anodes. As an example, the redox activity and the charge transfer kinetics have been found to be improved through transition metal doping at the B-site [42]. Moreover, the perovskite lattice has ionic pathways that favour bulk diffusion processes that are beneficial in maintaining moderate to high current densities. Notably, materials such as $Na_{0.5}Bi_{0.5}TiO_3$ (NBTO) have demonstrated good cycling stability and reversible capacity, making them promising candidates for sodium ion battery anodes [43]. The partially occupied A-site of Na^+ and Bi^+ in NBTO is important in charge balance and facilitating sodium movement through the lattice and Ti^{4+} at the B-site allows redox cycling to be stable [44]. Bi_3TiNbO_9 is another potential candidate, which is a layered perovskite oxide with 2D structural characteristics, which offer a variety of sodium diffusion pathways and improved stability at the interface with the electrolyte [45]. The perovskite structures also allow easier voltage profiles during cycling, which decreases the electrode materials stress. This is critical in terms of enhancing lifetime and energy efficiencies in real-world applications including electric vehicles and grid storage [46]. The ability of such materials to undergo deep sodiation with minimal volume change significantly reduces the risk of electrode pulverization, a common failure mechanism observed in alloy type anodes [47].

Challenges: Although perovskite-based anodes have strong points, they have a number of challenges as well that inhibit their commercial application in SIBs. Top of these is their inherently low electronic conductivity that limits the rate performance. To counter this, scientists tend to use conductive additions such as graphene or carbon nanotubes to form composite electrodes [37]. But these methods increase complexity of processing and can affect long term stability. The other significant problem is that unstable solid electrolyte interphase layers are formed during the first charge/discharge cycles. These layers tend to be loosely bonded and may change with time resulting in higher interfacial resistance and lower Coulombic efficiency [38]. Also, Na^+ ions are larger than Li^+ and hence cause high lattice strain during insertion, which may result in structural distortion, amorphization or even mechanical breakdown of the electrode [39]. Cycling can also lead to hysteretic behaviour, capacity fade and voltage instabilities through multiphase transitions in complex compositions perovskites [40]. Furthermore, sodium-ion transport through these materials tends to be slower than lithium ions, which is why optimization of ionic diffusion pathways is one of the highest priorities in the current research [41]. Some perovskite anodes have not yet achieved high-rate performance, which is critical to modern energy applications, due to slow interfacial dynamics. The surface interaction between the perovskite and the electrolyte may lead to parasitic reactions, resistive film formation and subsequent delamination [43].

Recent advances in perovskite-based anodes: Recent research has addressed these shortcomings by resorting to a number of more innovative approaches, such as nanoscaling, compositing, doping and interface engineering. Nanoscale perovskites offer shorter sodium-ion diffusion paths and increased surface area that together increases rate capability and decreases polarization [37]. Besides, the hierarchical nanostructures are able to absorb the volume change better, enhancing the cycling stability [41]. More recent work on NBTO and doped derivatives has demonstrated enhanced performance by bandgap tuning, surface activation and modification of the electronic structure [44]. Bi_3TiNbO_9 is a high-potential candidate, because of its layered structure and high metal-oxygen covalency, which promotes the ability to store sodium firmly

[45]. Embedding of perovskites within carbonaceous matrices has resulted in composites that have improved conductivity, more robust structures with stronger electrode stability even upon cycling [46]. As an example, the insertion of O3-type layered cathode materials into perovskite structures has proved to have synergistic electrochemical impacts [47]. Furthermore, other practical aspects like processing methods, compatibility with electrolyte and cost-effectiveness are also being considered. The search of viable cathode anode combinations and optimization of full-cell designs, is starting to show which perovskites can be made to work well under realistic operating conditions [48]. Other than the electrochemical performance, environmental and sustainability factors are emerging. As an example, lead-based perovskites, although they provide high energy density, are ecologically and health-wise problematic. Emerging lead-recycling technologies offer the potential to enhance material circularity and reduce toxicity by converting spent lead-containing perovskites into reusable materials for battery and photovoltaic applications. Moreover, the successful results in the rational design of new materials are being achieved using theoretical methods like the density functional theory (DFT) and machine learning models that are used to predict potentially interesting perovskite compositions [49]. These computational models are gaining insights that are confirmed by multidimensional experimental methods, including operando spectroscopy and *in situ* transmission electron microscopy, which can provide real time images of structural and interfacial changes [50]. Recent discoveries of fluorinated perovskites such as $NaNiF_3$ are also illuminating sodium storage, since these compounds have the structural advantages of oxides with the high redox potential of fluorides, with distinct electrochemical properties [51]. In addition, lead-based perovskite anodes have also shown tremendous energy storage capability, although recyclability issues are being solved by the application of green chemistry [52,53].

Table-1 presents a comparative overview of the most frequently cited perovskite-based anode materials discussed in the text, highlighting their performance metrics and relevance to sodium-ion battery (SIB) applications. This enhances the technical depth and contextual understanding of the corresponding section.

Gel polymer electrolytes used in sodium-ion batteries

Composition of GPE: Gel polymer electrolytes (GPEs) are semi-solid systems which are composed of three main components *viz*. polymer matrix, sodium salt and a plasticizer or liquid solvent. The polymer host serves as a mechanical scaffold that holds the liquid phase in place and does not leak and maintains structural integrity, whereas the plasticizer makes polymer chains more mobile and improves ionic conductivity by promoting ion transport [54]. Poly(vinylidene fluoride-*co*-hexafluoropropylene) (PVDF-HFP), polyethylene oxide (PEO) and polyurethane (PU) derivatives are common polymer hosts due to their mechanical strength, good film-forming property and electrochemical stability [55]. The ion conduction mechanism in GPEs is not only limited to the dissociation of sodium salts, but also to the polymer ion interactions, crosslinking density and the availability of functional groups on the polymer backbone. These influence segmental motion and the solvation environment which in combination determine the sodium-ion transport properties. The spatial arrangement of sodium ions, polymer chains and PEG side chains highlights the role of functional groups in promoting solvation and facilitating ion hopping, which contributes to enhanced ionic conductivity [56].

Polydopamine-modified tough polyurethane represents an advanced biodegradable polymer employed in the fabrication of eco-friendly gel polymer electrolytes (GPEs) for the flexible sodium-ion batteries (SIBs) [57]. Also, mechanical properties, ionic conductivity and interfacial chemistry have been improved with fillers such as NASICON-type ceramics and sulfonamide-functional polymer nanoparticles [58,59]. Such composite approaches have shown significant potential in enhancing overall performance by ensuring efficient Na^+ transport and improved thermal stability.

Benefits compared to liquid electrolytes: GPEs have a number of advantages over traditional liquid electrolytes that are critical to enhancing safety and electrochemical performance of SIBs. Liquid electrolytes are generally volatile, flammable and leak prone particularly when subjected to high temperatures. GPEs, however, are more thermally and chemically stable and this greatly reduces the possibility of fire and explosion in cases of device failure or thermal runaway [60]. Their semi-solid nature improves interfacial contact with electrode surfaces, reduces interfacial resistance and enhances electrochemical performance over extended cycling [61]. Moreover, GPEs are effective in suppressing the formation of sodium dendrites filamentous deposits that can penetrate separators and cause internal short circuits. By providing a more homogeneous and confined ion transport pathway, GPEs help reduce localized current density, which is a key factor in dendrite formation [62]. Furthermore, GPEs contribute to improved cycle life and Coulombic efficiency by minimizing solvent evaporation, electrolyte decomposition and parasitic side reactions [63]. Green products are also becoming more significant and GPEs manufactured using bio-derived polymers or non-toxic additives are in line with the world need of green energy technologies. Cellulose and chitosan-based biodegradable and biocompatible GPEs have also been investigated as a potential in low-cost and recyclable battery systems [64]. Moreover, nanoparticles with the sulfonamide functional group have been shown to stabilize high-voltage electrolytes and allow safe use with high-energy electrodes [65].

TABLE-1
ELECTROCHEMICAL CHARACTERISTICS OF REPRESENTATIVE PEROVSKITE ANODE MATERIALS IN SIBs

Material	Composition type	Specific capacity (mAh/g)	Voltage window (V)	Cycle stability	Ref.
$Na_{0.5}Bi_{0.5}TiO_3$	ABO_3 oxide	~200	0.01-2.5	>100 cycles	[13,18,45]
$NaNiF_3$	Fluoride perovskite	~160	0.01-2.2	~60 cycles	[25]
Bi_3TiNbO_9	Layered perovskite	~170	0.01-2.0	High reversibility	[45,46]

Challenges of GPEs: Although these are the advantages of GPEs, there are numerous challenges facing the widespread commercial application of GPEs. Among the major issues is that they have lower ionic conductivity in general than liquid electrolytes, especially at ambient temperatures. This constraint is usually enhanced in highly crystalline or high molecular weight polymers where the segmental movement is restricted, limiting the ability of ions to move [66]. The mechanical and thermal stability of GPEs should also be maximized to work at a wide temperature range and with different cathode and anode chemistries. Mismatch with high-voltage cathodes or unstable interfaces with emerging anode materials such as perovskite-based compounds can result in the formation of unstable solid electrolyte interphase (SEI) layers, leading to the premature battery degradation [67]. Interfacial mismatches often cause delamination, increased interfacial resistance and parasitic reactions at the electrode–electrolyte interface [68]. The other problem is the complicated synthesis and scalability of composite GPEs. Uniformity of dispersion of ceramic or polymer nanoparticles, regulation of crosslinking density and reproducibility of *in situ* polymerization methods are not trivial in mass production [69]. Reproducibility and structural homogeneity of large-scale GPE membranes is essential to the commercial viability and cost-effectiveness. In addition, GPEs are also required to have high safety and flame-retardant requirements, particularly in high-capacity, high-temperature applications. Polymer matrices should be able to withstand shrinkage, decomposition and ignition during abuse. Various flame retardant additives and siloxane-based polymers are currently under development to mitigate these interfacial and safety-related risks [70].

Recent developments in high-performance GPEs for SIBs: Over the past few years, a number of approaches have been identified to enhance GPE performance in sodium-ion battery systems. Among the most influential developments, the incorporation of sulfonamide-functional polymer nanoparticles is one of them. These materials enhance the mechanical strength of the electrolyte matrix as well as the conductivity of sodium-ions by establishing desirable conduction routes [65]. Such GPEs have been shown to be compatible with hard carbon and new high-energy anodes. High ionic conductivity and electrochemical stability of NASICON-type fillers have been used to integrate them into polymer matrices to form hybrid GPEs. These fillers can be used to fill the gap between liquid-like conductivity and solid-state safety, with better ion transport across a broad temperature range and better interfacial adhesion to electrodes [58]. This leads to a GPE that is effective in hot and cold conditions, which is essential in real world application [62]. Siloxane-based and flame-retardant polymer matrices are also becoming popular due to their twofold advantages of thermal stability and chemical insensitiveness [66,70]. These materials are particularly applicable in high end battery systems where safety is of great concern. Also, ionic liquids and deep eutectic solvents have been used as plasticizers or additives in GPEs to demonstrate potential improvements in electrochemical stability and ionic mobility as well as widened temperature operation windows [71]. Interface engineering is also another field that has been developed especially with *in situ* polymerization and crosslinking method. These methods assist in making more compact and integrated interfaces between GPEs and electrodes, which lower resistance and inhibit the formation of SEI [67]. Phase separation or leaching of liquid components can also be reduced by utilizing advanced crosslinking agents to result in longer operational life. Further studies have explored gel polymer electrolytes engineered to operate over a wide temperature range, maintaining flexibility and electrochemical performance from sub-zero to elevated temperatures [62]. This is particularly helpful to batteries that are used in automotive, aerospace and outdoor storage. Lastly, the coupling of conductive materials such as carbon nanotubes or conductive polymers with flexible biopolymer matrices has led to GPEs that are high rate capable and have outstanding mechanical stability. These materials are opening the door to the combination of flexible and wearable sodium-ion battery technologies [57]. Table-2 presents a consolidated overview of state-of-the-art GPE systems, highlighting the key attributes that make them suitable for modern SIBs. The referenced works are drawn from the GPE-focused section to ensure the consistency and relevance.

Mixing a perovskite anode with a gel polymer electrolyte in sodium-ion batteries

Compatibility of perovskite anode with GPEs: Incorporation of perovskite based anodes into GPE based SIB is a potent hybrid concept that harnesses the chemical flexibility of redox perovskites and mechanical flexibility and safety of semi-solid electrolytes. The purpose of this integration is to address the drawbacks of traditional solid-solid interfaces and volatile liquid electrolytes [72]. The efficacy of this strategy is highly contingent on the physicochemical compatibility as well as interfacial synergy between the perovskite material and the GPE matrix. Among the most extensively studied perovskite-based anodes are $Na_{0.5}Bi_{0.5}TiO_3$ and $NaNiF_3$. Their stability in the intercalation of Na^+, multi-valent redox potential and structural stability make them appealing to high-capacity and long-life SIBs [73]. These perovskites have a redox active Ti^{4+} and Ni^{2+}/Ni^{3+}, which gives a stable electrochemical platform when combined with a matching GPE. Nevertheless, difficulties come due to the incompatibility in the thermal expansion coefficients, dissimilarities in surface energy and inadequate wettability at the electrolyte-anode interface [74]. Recently, the application of nanocomposite GPEs in the form of ionic

TABLE-2
PERFORMANCE CHARACTERISTICS OF GPE SYSTEMS USED IN SODIUM-ION BATTERIES

GPE matrix	Salt/additive	Ionic conductivity (S/cm)	Operating temp. (°C)	Advantages	Ref.
PVDF-HFP + $NaClO_4$	Plasticizer + ceramic	$\sim 10^{-3}$	25-80	High mechanical + ionic stability	[55,57,60]
PEO + NaTFSI/NASICON	NASICON hybrid	$\sim 10^{-4}$	up to 90	Good electrode compatibility	[58,59,61]
PU + Ionic liquid	IL-doped GPE	$\sim 10^{-3}$	Broad	Eco-friendly and flexible	[64,65]

liquids and functionalized polymer nanoparticle to enhance interfacial bonding has been studied [75]. Not only are these GPEs more thermally and electrochemically stable, but also provide an increase in sodium-ion mobility at the interface. Also, the strategy of dual-ion channel polymer networks has been identified as a possible solution to develop mechanically adaptive interfaces that can ensure good contact with the perovskite surface during the dynamic cycling [76].

To enhance adhesion and minimize interfacial resistance 3D-structured GPEs with porous morphologies have been designed. These networks enhance the effective contact area between the anode and the electrolyte and allow uniform ion flow, which is crucial to stable cycling [77]. Better polymer-electrode interactions and ion solvation at the interface are achieved by functional polymer chains *e.g.*, those with sulfonic or ether groups [78].

Electrochemical performance: The combination of perovskite anodes and advanced GPEs has a tremendous effect in enhancing the electrochemical performance of SIBs. Improved interfacial contact results in improved sodium-ion diffusion, decreased polarization and stabilized long-term cycling voltage profiles [79]. These enhancements are manifested in increased capacity retention, improved rate performance and inhibited dendrite growth. Research has indicated that soft-tough composite GPEs offer mechanical compliance to compensate the rigidity of perovskite materials. This mechanical flexibility enables the electrode–electrolyte interface to accommodate the volume changes associated with Na^+ cycling, thereby preventing mechanical failure and SEI degradation [80]. Moreover, GPEs are used to form uniform and strong SEI layers that minimize the charge transfer resistance and enhance Coulombic efficiency [81]. These findings are essential in the optimization of GPE-perovskite combinations [82].

$NaNiF_3$ systems combined with sulfonamide-functional GPEs have high rate capability, low interfacial impedance and long cycle stability. The better Na^+ conduction is due to the better ion pathway and structural compatibility between the fluorinated perovskite and the GPE [83]. Also, the electrochemical structure is further strengthened by ionically conductive fillers, such as phosphate polyanions and NASICON-type ceramics, which enable safer and more efficient operation [84]. The composite architectures with interpenetrating networks, asymmetric layering and plasticized polymer matrices have proved especially useful in attaining high transference numbers, enhancing electrochemical stability and mechanical durability [85]. These designs assist in the preservation of electrochemical balance throughout the cell and avoid fading of capacity because of interfacial degradation or electrolyte breakdown [86].

Optimization strategies: To achieve high performance in hybrid SIB systems it is important to optimize the perovskite-GPE interface. The strategies are aimed at the anode surface and the composition of the electrolyte. GPEs can be functionalized to enhance ionic dissociation and wetting properties by modifying the functional groups, particularly to sulfonic acid, ether, amide or siloxane groups and form stronger bonds with perovskite surfaces [87]. The perovskite surface has been modified by atomic layer deposition (ALD) and carbon coating processes to increase electronic conductivity and protection against undesirable side reactions [88]. Also, an interphase layer of a well-designed engineering between the GPE and the anode can act as a buffer to regulate the transport of ions and reduce the instability of the interface [89]. The 3D GPEs with embedded ion-conducting networks have also been developed by researchers to facilitate stable diffusion of Na^+. Based on these soft-tough designs, they integrate strong mechanical scaffolds with ionic mobility to align with the electrochemical dynamics of perovskite anodes [90]. These asymmetric composite electrolytes help in reducing the volume changes, interface compatibility and increase battery life. Other materials such as metal-organic frameworks (MOFs) and polymer-ceramic hybrid membranes have also been added to the GPE systems to enhance structural stability and stabilize the sodium-ion transport, particularly in solid state applications [91,92]. Further, sodation of perovskite anodes and additives to electrolyte, like fluorinated solution or ionic liquid, have been found useful to minimize initial Coulombic inefficiencies and SEI formation [93]. In addition to materials engineering, lifecycle, environmental and economic factors are gaining prominence. Sustainable development goals and low-carbon objectives should be used in functional GPEs. Comparison of life cycle assessments of lithium-ion batteries with sodium-ion batteries demonstrates that the ecological impacts that sodium-based systems may have can be lower when coupled with recyclable GPEs and benign electrode materials [94-97]. The non-flammable, non-volatile composition of battery designs that employ perovskite anodes and GPEs can be especially beneficial because of the possibility of recovery and reuse [98]. Consequently, there is an increasing concern on the environmental impact of GPE synthesis, polymer sourcing and end-of-life recyclability of composite systems [99]. Such parameters will become more prominent in the choice of materials and design approaches in commercial SIB development [100]. Thus, sustainable design solutions and green chemistry methods should be incorporated into the future optimization pipelines of GPE perovskite hybrid batteries [101]. Table-3 summarizes the specific challenges and solutions at the electrode–electrolyte interface.

Stability and performance analysis

Electrochemical stability: Sodium-ion batteries (SIBs) require electrochemical stability as the basis of long-term operation and safety. The performance of integrated systems made of perovskite anode and GPEs relies on the presence of

TABLE-3
INTERFACIAL COMPATIBILITY STRATEGIES BETWEEN PEROVSKITE ANODES AND GPEs

Challenge	Mitigation strategy	Mechanism or component	Ref.
High interfacial resistance	Sulfonic/ether/amides on GPE	Enhances ion dissociation + bonding	[30,75,76]
Thermal and mechanical mismatch	Dual-ion channel GPE design	Improves ion movement + thermal adaptation	[27,77,86]
Delamination during cycling	Soft-tough composite GPEs	Provides better adhesion under stress	[28,87,90]

stable electrode-electrolyte interactions, reduced phase transitions and strong SEI formation [102]. Properly assembled, these components enable efficient and reversible insertion and extraction of Na^+ with little degradation. Researchers have found that the electrode-electrolyte interface, in particular the formation of SEI layer, plays an important role in determining the cycling performance of perovskite anodes. Phase transformation and lattice distortion are likely to occur during Na^+ insertion/removal in perovskite anodes and subsequently the electrochemical reversibility of the material [103]. Although GPEs are beneficial in decreasing the interfacial reactivity, the structural flexibility of perovskites should be maintained by controlled synthesis and compatibility with electrolytes [104]. Comparative studies of hard carbon and perovskite anodes have shown that hard carbon tends to experience more SEI build up and irreversible ion trapping when subjected to unstable liquid electrolytes [105]. It has resulted in the suggestion of customized electrolytes like GPEs that are able to deliver a consistent ionic field and inhibit parasitic reactions [106]. It has been found that in cathode development that reversible phase transitions and intergrowth engineering in layered oxides can be used to control the electrochemical stress and enhance long-term performance [107]. The same findings are applicable to the perovskite anodes, where the stable crystallographic behaviour is also necessary. More sophisticated interface stabilization such as surface passivation and defect engineering will be able to decrease oxygen escape and lattice collapse during prolonged cycling [108]. Recent developments have presented layered oxide cathodes with synthesized multifunctional structures that have synergistic properties at both cathode and anode interfaces, which allow balanced sodium transport and improved capacity retention [109]. The insights of these designs are used to determine how perovskite anodes can be structurally optimized to last longer in combination with GPEs [110].

Thermal stability and safety considerations: The thermal stability is a major component in defining the feasibility of SIBs, especially at high current, deep cycling or high temperatures. Although SIBs tend to be less reactive with organic solvents than lithium-ion batteries, they are nonetheless susceptible to thermal runaway in the event of the decomposition of polymeric or gel-based electrolytes [111]. The thermal decomposition processes include exothermic reactions of the electrolyte solvents, gas release due to salt breaking and SEI burning, which may cause a rapid increase in temperature [112]. GPEs have proved to be very promising in alleviating these risks by stabilizing the volatile components and lowering the flammability [113]. For example, when $NaNi_{1/3}Fe_{1/3}Mn_{1/3}O_2$ cathodes were tested in different electrolytes, it was found that polymer-based GPEs produced less heat and had a higher thermal resistance than conventional liquid electrolytes, mainly due to lower solvent volatility and stable polymer structures [114]. Safety-engineered GPEs with flame-retardant additives or thermally insulating backbones are under investigation as an alternative to the use of GPEs in military and automotive applications where there is a possibility of thermal abuse conditions [115,116]. Electrolytes in nanostructures with incorporated ceramic fillers have also been made to ensure the conductivity with minimal thermal degradation, which also ensures safer battery designs [117]. Battery safety measures should encompass both external thermal management systems and internal self-regulating materials capable of responding to abnormal temperature conditions. A potential possibility in the dissipation of heat during critical events is the incorporation of thermal buffers and phase change materials in GPE layers [118].

Degradation mechanisms and mitigation strategies: The SIBs performance deterioration is caused by multifaceted interconnected processes, such as SEI instability, electrode pulverization, ion trapping and electrical contact loss. These problems are particularly acute when the charge/discharge rates are high and the operating temperatures are high [119]. Structurally tunable perovskite-based anodes are however challenged by issues of ion-induced strain, surface reactivity and interfacial mismatch with the electrolyte [120]. GPEs are able to reduce degradation due to mechanical buffering and SEI growth inhibition. They should however be well engineeered to be compatible with high voltage cathodes and active anode surfaces. The same degradation has been mitigated in hard carbon anodes by pre-sodiation approaches and electrolyte additive optimization, which are currently being ported to perovskite-based designs [121]. More sophisticated mitigation methods involve the use of atomic layer deposition coatings, which create the protective and ionically conductive interlayers between the electrolyte and the electrode. These interlayers inhibit dendrite nucleation and they stabilize surface chemistry under long cycling [122]. General interfacial reconstruction methods, like the conversion of residual alkali on the cathode surface, have also been quite promising in improving air stability and reversibility at the interface [123]. The same is true of perovskite structures, which can degrade in the same way as layered oxide cathodes, by phase transition to inactive rock-salt structures, or irreversible oxygen loss. It has recently been demonstrated that bulk oxygen vacancies could be created by annealing treatments to increase sodium storage capacity and cycle stability [124]. Other solutions such as heterostructure engineering and defect passivation have also been effective in increasing the lifetime of electrodes [125]. The sustainability of these batteries is an issue in lifecycle. Lifecycle assessments (LCAs) stress that reducing degradation does not only increase battery usefulness but also enhances total carbon and economic performance [126]. Therefore, the structural engineering, interfacial chemistry and thermal management should be considered as a whole in the design of next-generation SIBs with perovskite anodes and GPEs.

Future perspective and challenges

Scaling up: A major challenge is the scale-up of sodium-ion batteries (SIBs) to commercial application. Despite the demonstrated potential in research of electrode materials, particularly perovskite-based anodes and gel polymer electrolytes (GPEs), scaling these technologies to avoid cost-effectiveness and performance trade-offs is complicated. This challenge is based on the problems of synthesis complexity, batch purity and absence of streamline processing infrastructure [127,128]. Phase transitions at low state-of-charge, as [127] points out, are also important to overcome and cathode stability is essential to enable long-term real-world operation. The use of manufacturing strategies that are compatible with current lithium ion battery (LIB) production lines, *e.g.* modular fabrication or drop-in process integration, are also being more investigated to lower capital costs and accelerate adoption [129]. As an example, Gaurav *et al.* [129] point out that halide perovskites, which are already processable in solar cells, could be modified to work in battery systems to simplify electrode processing. Nonetheless, scale-up can be economically sensitive as Zhang *et al.* [130] warn that halide perovskites are commonly synthesized with fine control over composition and morphology. Stability and environmental safety also create issues of industrial feasibility as reported by Mohan *et al.* [131]. It will be required to optimize GPE manufacturing processes, including solvent-free polymerization, improved mixing processes and green solvent recovery methods to guarantee reproducible performance in thousands of battery cells [132,133]. Moreover, vacancy or dopant incorporation in perovskite structures could be scaled, which could provide new routes to improve ion mobility without losing processability [134,135].

Cost *vs.* performance trade-offs: The high availability and low price of sodium render SIBs very appealing to cost-sensitive uses such as grid storage, backup and stationary energy infrastructure. Nevertheless, the relatively lower energy density and high cost of advanced materials such as perovskites and engineered GPEs are some of the factors that question the economic feasibility of SIBs [136]. Newton *et al.* [137] state that although sodium-ion chemistries are beneficial in terms of sustainability, they are not always cost-effective without lifecycle optimization. High-performance perovskite anodes are either rare or costly transition metals or complex synthesis, which restricts their large-scale application. Likewise, the addition of biodegradable polymers, ionic liquids and functional nanofillers to GPEs enhances safety and stability, but it may significantly increase the cost of materials [138,139]. It will be important to balance the energy efficiency and the material costs. Hasan *et al.* [140] propose the use of hybrid cell designs that strategically incorporate low-cost, mid-performance components to reduce costs while preserving key battery specifications. In general, this trade-off can be overcome by optimizing materials and simplifying the design, *e.g.* by using single-layer pouch cells or modular GPE coating systems. Moreover, cell packaging, thermal management and electrolyte reuse advances can also potentially lower the overall system costs [141,142].

Potential breakthroughs in perovskite chemistry and GPEs engineering: Perovskite chemistry is still developing, expanding to include halide, double perovskites and hybrids. Such new systems provide tunability of ionic conductivity, redox properties and structural stability. Wang *et al.* [136] highlighted the increasing focus on heterostructure engineering and interface tuning to achieve superior electrochemical performance. As an example, halide perovskites have already shown superior ion migration properties in supercapacitors and can be modified to SIBs to achieve high-rate and stable performance [129,130]. Furthermore, Zahra *et al.* [132] and Choon & Lim [133] explained the performance of perovskites can be optimized to high-capacity SIB anodes by defect chemistry, especially oxygen and cation vacancies. The defects can help in the control of volumetric changes and the improvement of sodium ion transport. In the electrolyte side, GPEs are experiencing the developments in backbone functionalization, nanoporous matrices and dual-ion pathways. Wang *et al.* [135] and Newton *et al.* [137] report composite GPEs that combine several transport channels and supramolecular motifs to achieve robustness to dendrite formation and interface degradation. The combination of these developed GPEs with perovskite structures provides new avenues of synergistic enhancements in both safety and rate capability.

Sustainability and environmental considerations: The future of battery technology is dependent on sustainability. The ecological benefits of sodium-ion systems, based on more common and less hazardous materials, are inherent in comparison with lithium-based systems. SIBs are typically better in terms of carbon footprint, resource depletion and toxicity than lithium iron phosphate (LFP) and nickel manganese cobalt (NMC) chemistries according to life cycle assessments (LCA) [139,143]. The strategies of circular economy like biodegradable GPEs, low-energy processing and efficient recycling loops are becoming popular. According to Banerjee & Joo [142], SIBs are the key to the realization of decentralized, environmental friendly energy storage, particularly in emerging economies. Phogat *et al.* [144] & Nekahi *et al.* [145] also demonstrate that sodium-ion chemistries may satisfy sustainability criteria without using conflict minerals or water-intensive mining. In the meantime, the environmental burden of electrode manufacturing has been decreased by the development of green anode technologies, including lignin-derived hard carbon [146,147], wood-based precursors [148] and biomass-derived hosts [149]. Fan *et al.* [150] and Ghimbeu *et al.* [151] show that renewable hard carbon materials can provide high plateau capacities and outstanding cycle life. Cui *et al.* [152] underline that in case process innovations and circular supply chains are implemented in full, SIBs would become a pillar of sustainable, scalable and equitable energy access in the global context.

The combination of perovskite-based anodes and gel polymer electrolytes (GPEs) is a major breakthrough in the area of sodium-ion batteries (SIBs) that offers an encouraging pathway to a trade-off between safety, performance and scalability of energy storage. The commonly hung up potential of this review has been the synergy expected to be achieved between ion-conductive, flexible GPEs and structurally versatile perovskites. The two elements, however, require their interactions to be critically understood along with the issues of materials and design in general. Perovskites, general formula ABX_3, can be tuned to have desirable physico-chemical characteristics by A or B-site substitution to produce a desired sodium-ion mobility and structural stability during repeated cycling. As an example, $Na_{0.5}Bi_{0.5}TiO_3$ and $NaNiF_3$ have shown stable Na^+ intercalation, good redox behaviour and stability through phase transitions [6,7]. The Ti^{4+} and Ni^{2+}/Ni^{3+} redox flexibility in such structures further provides multi active sites to increase its energy storage capability. Nevertheless, the perovskite anodes have limitations associated with interface degradation, poor Coulombic efficiency in the first cycles and thermal and electrochemical instability [24,25].

GPEs have become the emerging prospects as electrolytes since they provide both mechanical stability of solid-state systems and ionic conductivity of liquid-based equivalents. Their semi-solid nature does not allow leakage and dendrite formation and keeps in close contact with the electrode [8,9]. Numerous changes, including the addition of sulfonic, ether or amide groups to polymer matrices, have made a major difference in terms of ionic conductivity, thermal stability and interface stability [30,94]. In addition, it has been useful to employ nanostructured or dual-ion channel polymer architectures to facilitate improved electrode-electrolyte adhesion and low polarization [27,86]. Nevertheless, integrating perovskite anodes and GPEs is a complex affair, as well. Of major importance is interfacial compatibility. An electrochemical performance could be damaged by developing a mismatch between thermal expansion coefficients, wettability and chemical activity. Sodium-ion diffusion can be hampered by the formation of an unstable or resistive solid electrolyte interphase (SEI) leading either to irreversible capacity loss [28,32]. Nanocomposite GPEs (including ionically doped GPEs or GPEs with functionalized polymer nanoparticles) have been studied and have been shown to increase interfacial bonding and electrochemical stability [21,26,83]. By electrochemical performance measurements, it can be seen that perovskite-GPE systems can keep their capacity retention and Coulombic efficiency during long cycling, especially under the appropriate interface engineering. Composites in GPEs, such as soft-tough combination, which fit the surfaces of anode particles, have lower strain upon volume increase and encourage SEI stabilization and increased reversibility [28,90]. As an example, anodes made of $NaNiF_3$ and optimized GPEs had lower polarization and enhanced rate capacities due to the smoother ion transport channels [25,82]. The XRD, SEM and electrochemical impedance spectroscopy *in situ* and *ex situ* characterizations have provided insights toward the phase evolution, lattice strain and resistance formation at interface. Such analyses indicate that reversible alloying/dealloying in Bi-based perovskites and strong crystalline retention are the factors that lead to improved electrochemical performance when combined with flexible GPEs [29]. However, the long-term stability remains undermined by degradation processes such as interfacial delamination, structural phase collapse and electrolyte decomposition, unless these processes are addressed by rational design. It is thus important to have mitigation strategies. Coatings that have been used on perovskite anodes have included atomic layer deposition (ALD) to minimize the surface reactivity to promote stable SEI development [23,107]. A major advantage in universal interfacial reconstruction methods (*e.g.* converting residual alkali at the electrode surface) is that these enhance air stability and electrochemical reversibility, features necessary to ensure the practical scope of a method [118]. At the same time, high transference number and porosity engineered polymeric GPEs contribute to the mechanical integrity and uninterrupted ionic pathways [27,86]. Such aspects of safety as thermal stability are also critical. Even with the fact that GPEs pose lesser risk of flammability than liquid electrolytes, the potential of thermal runaway still exists. The entrapment of volatile solvents in polymer matrices is one way of inhibiting the evolution of gases and heat production at high temperatures [37,87,112]. The additional use of ceramic nanofillers, hybrid organic inorganic composites and multi-functional dopants is proposed to improve GPE thermal properties without reducing ionic mobility [89,91].

Another important dimension is pointed out by a life cycle perspective. Sodium-ion technologies and in particular systems based on the use of abundant and those with low degrees of toxicity, are less environmentally adverse and more sustainable than the lithium-ion chemistries [31,136,145]. Progress has also been made in developing GPEs using natural compounds and biodegradable polymers such as lignin, cellulose, *etc*. [147,148]. Not only do they increase sustainability, but also follow the principles of circular economies, which focus on low-energy production, recyclability and reduced carbon footprint [100,141]. Although promising progress is made, there is still a bottleneck of scalability. The synthesis of both perovskite and GPE is frequently multi step with strict limits on purity and stoichiometry. Scaling these processes up through performance shall be achieved by means of modular, scalable manufacturing plans. It has been suggested that drop-in compatibility with current lithium-ion battery fabrication lines, comprising slurry-casting, calendaring and roll-to-roll coating, can lower capital cost, as well as the uptake of the technology towards commercial use [39,152]. Also, cost-performance trade-offs should be taken into consideration. Developed perovskites (especially high cost/rare element doped ones) can negate cost technologies of sodium. Likewise, GPEs with high performance, in terms of ionic liquids, ceramic nanofillers or functional polymers, could increase the cost of production. Performance gains have to pay off their cost in terms of large energy density, safety or cycle life [39,97] as demonstrated by the techno-economic analyses.

The future of breakthroughs is in chemistry and engineering. Halide perovskites with their excellent electronic tunability in photovoltaics could also be an opportunity in ion storage, particularly when designed to reduce vacancy related instability [129,130,133]. Similar GPE development, such as dual-ion channel engineering and multifunctional biopolymer hybrids, are also likely to overcome current constraints on ionic conductivity and mechanical properties [94,96,132]. Comprehensively, the perovskite-GPE combination in SIBs is in a revolutionary starting point. On the one hand, the combination is set to address long-term concerns of safety, cycle life and capacity retention. On the one hand, tuning interface chemistry, scaling up production and making it affordable is a daunting task. Handling such multi technological issues will need interdisciplinary efforts featuring materials research science, electrochemistry, modelling and sustainability research.

Conclusion

Perovskite anodes and gel polymer electrolytes (GPEs) are the promising direction toward safer, higher efficient and scalable sodium-ion batteries (SIBs). Perovskites have tunable structure, rich redox chemistry and high electrochemical performance, whereas, GPEs have mechanical stability, thermal resistance and higher ionic conductivity. Each of these parts has contributed to the development of SIB technology; collectively, they can revolutionize the next-generation energy storage systems. Nevertheless, there are main obstacles, which have to be overcome: interfacial mismatch, structural degradation of perovskites and low ion transport at room temperature, despite the great potential. Recent findings have identified a number of promising approaches to address these challenges that includes nanoscale structuring, interfacial engineering, sophisticated polymer design and multifunctional composite structures. Also, focus on sustainable low-cost materials and processing technologies contributes to the popularity of these systems as commercial and grid-scale applications. The development of co-designing electrode-electrolyte interfaces, large scale synthesis methods optimization and a profound knowledge of interfacial electrochemical dynamics should be the priorities of future work. The SIBs based on perovskite and GPE have high potential as a technologically feasible and eco-friendly option because global demand is increasing in sustainable and cost-effective energy sources. As long as innovation and interdisciplinary cooperation continue, the hybrid architecture will assist in closing the gap between laboratory success and industrial implementation.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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