

Frontier Electrolytes for Efficient Sodium-Ion Batteries

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Abstract. The considerable demand for renewable energy sources and the mounting concern about environmental pollution has created a new development opportunity in the field of secondary batteries, which can store chemical energy in place of electrical energy. In the 2010s, attention was drawn to sodium batteries. A sodium-ion battery (SIB) is a specific kind of battery that can be recharged, given that sodium is a plentiful element on Earth and is more stable than lithium batteries. Consequently, there has been a growing body of research and development activity in this field. This article will describe the electrolytes in SIBs, which significantly contribute to the electrochemical and safety characteristics of SIBs. The electrolytes discussed will include liquid electrolyte, solid electrolyte, and solid-liquid composite electrolyte. The most common use of electrolytes in an organic solvent comprises sodium salt (NaPF_6 or NaClO_4) and carbonate. This electrolyte has been successfully matched with a range of anodes and cathodes, resulting in satisfactory overall performance. Nevertheless, the obvious drawback is corroding sodium metal electrodes over extended periods of use and potential safety concerns.

Keywords: Sodium-ion battery; liquid-state electrolyte; solid-state electrolyte; solid-liquid composite electrolyte.

1. Introduction

As the utilization of fossil fuels by humanity has reached its limit, there is a growing focus on the exploration of alternative energy sources [1]. There is a pressing need for humanity to direct its attention towards the development of energy storage technology. Electrochemical energy storage stands out among the several methods of energy storage due to its inexpensive initial investment, high efficiency, and versatile application. The widespread implementation of SIBs in electrochemical energy storage must demonstrate comprehensive performance in terms of longevity and cost-effectiveness. A SIB operates by transferring sodium ions between the positive and negative electrodes, allowing for the storage and release of electrical energy. In comparison to traditional LIBs, SIBs exhibit a number of distinctive characteristics.

To illustrate, the battery exhibits high safety standards and is not susceptible to combustion. Moreover, it demonstrates improved charging and discharging capabilities at both high and low-temperature conditions. The operating temperature range of LIBs is from $-20\text{ }^{\circ}\text{C}$ to $55\text{ }^{\circ}\text{C}$, while sodium batteries are able to operate normally between $-70\text{ }^{\circ}\text{C}$ and $100\text{ }^{\circ}\text{C}$ [1,2]. Furthermore, their capacity retention rate is nearly 82.95% under cold conditions, which is sufficient to address the issues of charging difficulties for electric vehicles in winter and the inefficiency of high-cold regions' energy storage power stations [3]. Additionally, SIBs possess distinctive advantages in the commercial sector. The Earth's crust contains 2.75% sodium, a significantly higher concentration than lithium (0.0065%). Following the large-scale production of SIBs, the cost is expected to be approximately half that of lithium. Nevertheless, several challenges remain to be addressed, including low cost, charging rate, cycle life, and energy density.

Electrolytes' function as a medium for electrochemical reactions is of great consequence in influencing the thermodynamic and kinetic processes of interfacial reactions in electrode materials. Consequently, it has a major effect on the multiplicity performance and cycling stability of SIBs. In addition to their high conductivity and large sodium ion mobility number, electrolytes should be considered for their thermal stability, economic efficiency and the solid electrolyte interface's stability. As with LIBs, liquid, solid, and solid-liquid composite electrolytes are the three main categories into which SIB electrolytes fall. In general, the ionic conductivity of liquid electrolytes is much higher

than that of solid electrolytes, which are further subdivided into ester and ether electrolytes. This paper will provide an in-depth analysis of the electrolytes utilized in SIBs.

2. Operation Principle of SIBs

SIBs are rechargeable battery technologies that operate on a comparable principle to LIBs. As with all rechargeable batteries, SIBs are composed of four principal components: an electrolyte, a positive electrode, a negative electrode, and a diaphragm. During the charging process, it is the cathode to the anode that transfers sodium ions. Simultaneously, electrons are transported through an external circuit incorporated into the anode. The quantity of sodium ions transferred is directly proportional to the battery's charging capacity. Conversely, in the discharge phase, the ions undergo a reversal of the aforementioned process, whereby they return to the positive electrode. Consequently, a greater discharge capacity is attained with the transfer of each ion.

The most notable similarities between SIBs and lithium-ion batteries (LIBs) reside in the standard electrode materials and electrolytes used by them. Nevertheless, even though Na- as well as Li-ions are close to one other in the periodic table, their electrochemistry usually differs significantly. This difference can be explained by the reality that a Na ion's radius (95 pm) is around 1.5 times bigger than a Li-ion's radius (60 pm). As an example, sodium cobalt oxide (NaCoO_2), which is the direct Na-ion analog of LiCoO_2 ($\text{NaCoO}_2 = \text{Na}_x\text{CoO}_2$), shows significantly lower discharge/charge voltages and poorer electrochemical recycling ability than LiCoO_2 [4]. Nevertheless, new Na-ion cathode materials with improved efficiency have been effectively created, exemplified by $\text{Na}_{1.5}\text{VPO}_{4.8}\text{F}_{0.7}$ [5]. Considering the affordability of sodium salts and the potential to use inexpensive Al current collectors on the anode side (in contrast to the alloying tendency of Li with Al), a considerable number of electrode materials currently under development for SIBs are not economically viable for deployment in commercial cells. This is especially true considering how much their preparation actually costs. Despite the elevated capabilities exhibited by other anode materials, they are characterized by elevated desodiation voltages, resulting in reduced cell voltage in the corresponding full cell. Additionally, certain anode materials present safety concerns. For example, P exhibits a relatively high specific capacity and a relatively low desodiation potential, which renders it an attractive candidate as an anode material [6].

Here is a comparison of the lithium and sodium's physical characteristics that are important for their use in rechargeable batteries (Table 1).

Table 1. Lithium/sodium properties comparison [7].

Characteristics	$_{11}\text{Na}$	$_3\text{Li}$
Atomic weight [g mol^{-1}]	23	6.94
Mass-to-electron ratio	23	6.94
Ionic volume [$\text{\AA}^3$]	4.44	1.84
Shannon's ionic radius [$\text{\AA}$]	0.76	1.02
Melting point [$^{\circ}\text{C}$]	98	182
Voltage [V] vs SHE	-2.714	-3.045
Ratio of reserves (ppm in the earth's crust)	2830	6.5
Distribution	Everywhere	70% in S. America
Material abundance in earth's crust [ppm]	23000	20
Material abundance in water [mg L^{-1}]	11000	0.18
Price of carbonates [\$ per ton]	150	5000
Price of anode current collector [\$ per metric ton]	1885(Al)	5719(Cu)
Metal electrode theoretical capacity (mAh g^{-1})	1165	3829
Metal electrode theoretical capacity [mAh cm^{-3}]	1131	2062
Desolvation energy [kJ mol^{-1}] in	$\approx 40\text{-}70 \text{ kJ mol}^{-1}$ higher than Li	
Solvation energy [kJ mol^{-1}] (DEC)	148	208
Solvation energy [kJ mol^{-1}] (EC)	152	211
Solvation energy [kJ mol^{-1}] (PC)	158	218
First IE [kJ mol^{-1}]	495.8	520.2

3. Feasibility of Improving SIB Performance through Electrolytes

It is common practice in the area of energy storage by electrochemistry to accord less attention to electrolytes than to active materials such as electrodes when developing new technologies in this area. This is due to the fact that the characteristics of the latter determine the energy density (both in volume and mass terms) of the system and are, therefore, the most immediately striking. It would be remiss not to consider the electrolyte's function, which plays an essential part in determining the lifespan of the system, as well as the achievable performance with regard to rate capability, safety, and accessible capacity [8].

In the context of SIBs, the electrolyte is the ionic carrier of charge that is necessary for electrochemical reactions to take place. In order to achieve the optimal characteristics for an ideal electrolyte, a salt or solvent must fulfill the ensuing requirements: It ought to have excellent chemical stability, electrochemical conductivity, and ionic conductivity within a wide potential range, good thermal stability, a low cost, a straightforward process, a low toxicity and environmental compatibility.

The adoption of the following criteria for electrolytes would significantly enhance the possibility of improving battery performance through the use of more efficacious electrolytes. The additional

point to note is that a universal collection of characteristics needed for an SIB electrolyte is completely compatible with the lists usually created for LIB-oriented electrolytes. [9].

(1) The chemical stability of the cell is maintained throughout its operational lifetime, preventing any chemical reactions. These can be discovered inside the cells themselves, in the separators and electrodes that are used, or in the current collectors and packing materials that are utilized.

(2) electrochemically stable: the high and low onset potentials, which are broken down by oxidation and reduction, respectively, show a notable difference.

(3) thermally, the liquidus range's melting and boiling points are required to be located well outside of the operating temperatures, as defined by the internal parameters of the process.

(4) ionically conductive and electronically insulating-the primary objective is to sustain cellular operations by facilitating the transport of ions (in this case, sodium) while simultaneously attempting to minimize the self-discharge of the cell.

The aforementioned standards are nearly identical to those of LIBs, with the exception of a few specific requirements:

(5) The substance in question must exhibit a low level of poison and properly satisfy the necessary criteria for minimal risk to the ecosystem.

(6) The synthesis of the chemical compounds must be conducted sustainably, utilizing elements that are abundant in nature and employing synthesis processes with a low environmental impact in terms of energy consumption and pollution.

(7) The objective is to achieve the lowest total cost of materials and production [8].

4. Liquid-State Electrolytes

In the field of SIB liquid electrolytes, a crucial consideration is the interfacial aspects of the electrode process, which can be employed to enhance the safety and compatibility of these electrolytes with electrode materials and excipients. A liquid electrolyte can be defined as an electrolyte solution in the form of a liquid. The typical composition of such a solution comprises organic solvents, inorganic salts and additives. Organic solvents, including carbonates, display excellent solubility and stable electrochemical windows, thus providing an optimal environment for the dissociation of inorganic salts. The inorganic salts are typically sodium salts, exemplified by NaClO_4 , which undergo dissociation in the solvent to generate freely moving lithium ions and other anions that serve as charge-transfer mediators during battery operation [8]. Liquid electrolytes offer a significant advantage in terms of ionic conductivity, enabling batteries to perform at rapid charging and discharging rates and high current densities.

However, as a consequence of its fluid state, the electrolyte may present certain challenges, including potential leakage and volatilization. Additionally, it may exhibit a heightened susceptibility to instability and even combustion or explosion in extreme circumstances, such as elevated temperatures or excessive charging.

4.1. Organic Liquid Electrolytes

Both the electric field and the solvation effect are the main forces behind the transport of Na ions in organic liquid electrolytes, which together constitute the driving force for the flow of ions. By contrast, the driving forces within polymer and solid electrolytes are the migration of segments and vacancies. Therefore, it can be seen that the conductivity of Na-ions is noticeably higher in organic liquid electrolytes than it is in solid electrolytes, which is a consequence of the superior fluidity of the former [10]. The requirement for solvents is 1) Low viscosity for Na^+ migration, 2) High dielectric constant for dissolving sodium salts, 3) High chemical and electrochemical stability, and 4) High thermal stability and safety in harsh situations [11].

These different and sometimes contradictory requirements are not easily met by one single solvent. Ester solvents and ether solvents are the most common solvents in use at the moment in SIBs.

Excellent performance in battery applications is achieved with This pair of solvents that are organic. Table 2 shows the physical and chemical properties of some common organic solvents.

Table 2. Some common organic solvents - physical and chemical properties [12].

Solvents	Density/g cm ⁻¹	Boiling point/°C	Melting point/°C	Viscosity/ 10 ⁻¹ Pas 25 °C	Dielectric constant/ Fm- 125 °C	HOMO/ eV	LUMO/ eV
DMC	1.063	91	4.6	0.59	3.107	- 0.2488	- 0.0091
DEC	0.969	126	-74.3	0.75	2.805	- 0.2426	- 0.0036
EC	1.321	248	36.4	2.1	89.78	- 0.2585	- 0.0177
PC	1.200	242	-48.8	2.53	64.92	- 0.2547	- 0.0149
EMC	1.006	110	-53.0	0.65	2.95	- 0.2557	- 0.0062

The requirement for sodium salt is to be chemically stable and safe and to remain chemically inert to the solvent, electrode, collector liquid, and membrane. Therefore, Due to the sodium salt's high solubility and conductivity in the electrolyte, as well as its ease of preparation and environmental friendliness, the electrochemical performance of the electrolyte, including cycling stability, can be improved. Additionally, the use of greener raw materials in the production process contributes to the sustainability of the battery industry.

Sodium salts play an important role in electrolyte performance and act as an additional crucial element of organic liquid electrolytes. High solubility in organic solvents and limited amounts are prerequisites for additives. By carefully adding a tiny quantity of functional additives to the electrolyte of sodium-based batteries, some battery performance parameters, including reversible capacity, electrode or electrolyte compatibility, cycle performance, rate performance, and safety performance, may be enhanced. These chemicals are crucial to the functioning of SIB technology. Then, there are various kinds of additives, such as agents that reduce viscosity, scavengers of impurities, acidifiers, and so on.

4.2. Aqueous Electrolytes

The battery would be cheaper and more environmentally friendly by using an aqueous electrolyte. Contrary to the aliphatic and aromatic carbonates utilized in organic electrolytes, aqueous electrolytes have higher heat-sinking capabilities and ionic conductivity. This could be expected to make the battery perform better and reduce costs.

The use of aqueous electrolytes in secondary SIBs has been the subject of substantial research interest due to the inherent safety, cost-effectiveness and environmental compatibility of such solutions, which have the potential to offer a safer, more ecologically friendlier, and more sustainable method of storing energy. One of the main obstacles to using aqueous electrolytes in real-world situations is the electrolyte's limited electrochemical stability window (ESW) and the requirement to maintain the electrodes' structural and chemical integrity inside this ESW [13].

Aqueous rechargeable SIBs function according to a mechanism that bears resemblance to that observed in conventional non-aqueous batteries. As depicted in Fig. 1, the process involves the diffusion of sodium ions via the electrolyte and the transmission of electrons between the cathode and anode via the external circuit.

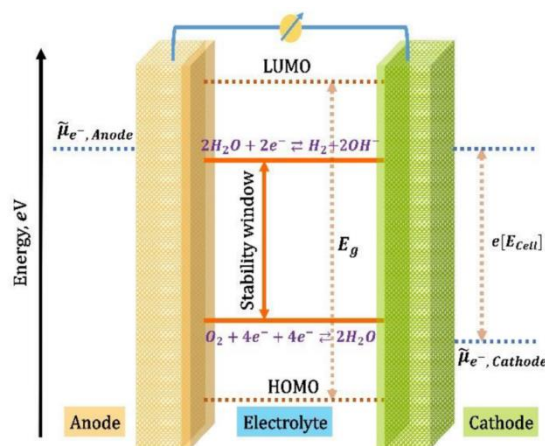


Fig. 1 Working principle of sodium ion battery [13].

At the same time, the following key electrolyte characteristics are necessary to obtain improved cycle stability and high energy density: 1) the electrolytic electrochemical stability window, 2) the chemical stability of the electrolyte's electrode material, 3) electrolyte ionic conductivity, 4) salinity and 5) interfering side reactions of dissolved oxygen in the water [13].

4.3. Ionic Liquid Electrolytes

Ionic liquid electrolytes refer to liquids that are at room temperature and contain only cations and anions, with no other molecular species present. In combination with ionic liquids, SIBs will have a wider operating temperature range and improved safety, which will allow the creation of batteries with superior performance with a wider range of applications [14]. In particular, replacing the electrolytes made of organic solvents that are frequently utilized in commercial systems with ILs is an alternative to consider, as their intrinsic volatility poses a safety risk during battery operation. It has better thermal stability than organic solvents but has poor transport characteristics [15].

5. Solid-State Electrolytes (SSE)

The distinctive element of a solid-state battery is called a solid-state electrolyte (SSE), which is a solid ion conductor and electron insulator substance. There are three main categories into which SEs may be divided: Inorganic solid electrolytes (ISE), solid polymer electrolytes (SPE), and hybrid combinations of these two, and SPE and ISE will be introduced. Outstanding safety is the main advantage of solid-state electrolytes. By eliminating the liquid component, solid batteries fundamentally eliminate the possibility of leaking and burning. In addition, solid-state batteries offer greater energy densities and longer cycle lives because solid-state electrolytes are better equipped to prevent side reactions on the electrode surface and minimize the loss of active material. However, there are still some challenges, characteristics like high interfacial resistance and poor ionic conductivity, which limit their use in high-volume applications.

5.1. Inorganic Solid Electrolytes (ISE)

The most extensively researched ISEs are oxides, sulfides, and boron hydrides. In general, these ISEs exhibit notable ionic conductivity, admirable thermal stability, and satisfactory mechanical characteristics despite the lack of adequate physical contact with the sodium anode and electrode materials [16]. They tend to exhibit poor compatibility and stability with the electrode because they are fragile. They are challenging to scale up from the research to the commercial scale and have a fast-growing interfacial resistance. There are some examples of ISE include Lithium Phosphorus Oxynitride (LiPON), cubic-phase $Li_7La_3Zr_2O_{12}$ (c-LLZO), $Li_{1+x}Al_xGe_{2-x}(PO_4)_3$ (LAGP), $Li_2S-P_2S_5$ (LPS) and $Li_{10}GeP_2S_{12}$ (LGPS) [17].

The following characteristics of the ideal electrolyte should be present: the electrical conductivity of approximately $10^{-2} \text{ S cm}^{-1}$ over an extensive range of temperatures (from ambient to moderate), minimal electronic conductivity, superior heat resistance, and a wide window for the electrochemical potential stability of at least 5 V vs. Na^+/Na to enhance voltage of the cell and circumvent unintended interfacial interactions between Na metal anodes and high-voltage cathodes. It has no chemical reaction with the Na-containing cathode or the Na metal anode. Furthermore, it must be dense and robust enough to prevent the formation of dendrites. Additionally, it should be straightforward to process into a thin film. It must also be affordable, safe for the environment, and easily scalable for industrial manufacturing on a big scale [18].

5.2. Solid Polymer Electrolytes (SPE)

An ion-conducting salt solution in a polymeric host material is the solid polymer electrolyte (SPE), which conducts ions via the polymer chains. SPEs have been shown to exhibit excellent interfacial contact and are manufactured on a large scale. However, it has also been shown that SPEs have drawbacks with regard to low ionic conductivity at ambient temperature, inadequate mechanical strength, poor stability in the presence of Na anodes, and excessive thickness [16,19].

Thankfully, polyethylene (PE) film, which serves as the foundation for permeable poly (ethylene oxide)-bis(trifluoromethanesulfonyl)imide (PEO/NaTFSI) to create PE-PEO/NaTFSI SPEs, has been studied by Jinbo Zhang et al. SPEs with outstanding $25 \mu\text{m}$ thickness, lightweight (2.1 mg cm^{-2}), great mechanical strength (tensile strength = 100.3 MPa), and high flexibility were the end product. Additionally, as shown in Fig. 2, the SPE demonstrated enhanced interfacial stability of the sodium metal anode and an ionic conductivity of $9.4 \times 10^{-5} \text{ S cm}^{-1}$ at 60°C [19].

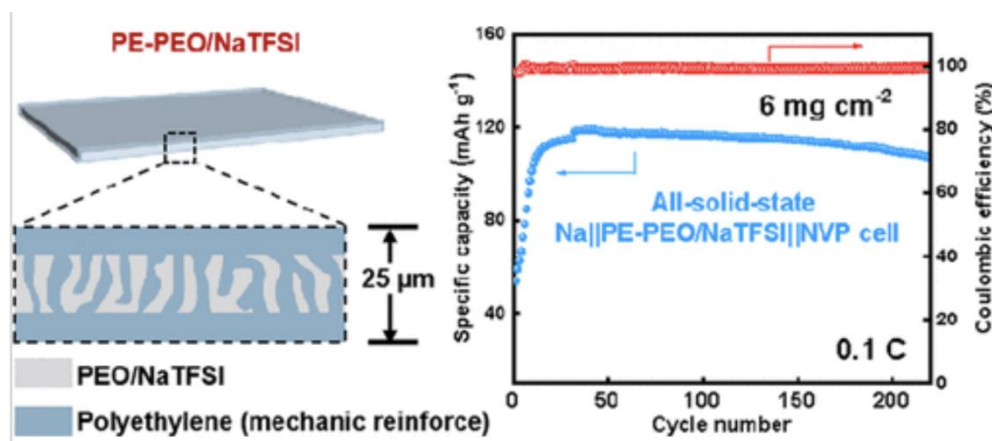


Fig. 2 Cross-sectional view of PE-PEO/NaTFSI SPE and comparison of various types of data, including specific capacity, cycle number and coulombic efficiency [19].

6. Solid-Liquid Composite Electrolytes

A solid-liquid composite electrolyte is called a gel polymer electrolyte (GPE). It is able to be used both as a diaphragm and as an ionic conductor in SIBs with a high degree of safety and is mainly composed of polymers, plasticizers and sodium salts. Owing to its broad electrochemical window, superior mechanical characteristics, good thermal stability, high ionic conductivity at ambient temperature, and good electrode compatibility during cycling, GPEs have attracted more and more research attention. However, in comparison to liquid electrolytes, there is a notable reduction in conductivity. The latest studies demonstrate that the electrolyte's crystal structure and chemical makeup have a significant impact on cell performance in a variety of ways. For SPEs, the ionic conductivity at ambient temperature is typically less than $\approx 10^{-5} \text{ S cm}^{-1}$. As a transition between liquid and all-solid electrolytes, GPE provides an acceptable ionic conductivity [20].

7. Conclusion

In order to optimize the performance of an SIB electrolyte, it is essential to consider not only its high conductivity and high sodium ion mobility number but also its thermal stability, economic efficiency, furthermore of the solid electrolyte interface's durability. The SEI layer formed by the electrolyte on the surface of different electrodes has varying compositions and properties, which consequently impact the electrochemical stability of SIBs. Consequently, there is a necessity for industrial production to focus on electrolyte and electrode matching. A series of fundamental experiments on electrolytes in different systems should be conducted through advanced characterization and simulation calculations to explore the ionic size of the solvent, the size of the solvated ions, and other parameters that may affect the sodium storage mechanism in different structural materials. Further research must be conducted with a more concentrated focus on the ionic transport properties of the electrolyte. The creation of sodium compounds at the electrode surface is closely related to the type of contact, which will manifest distinct properties such as ionic conductivity and mechanical strength. Hence, the development of commercially viable electrolytes for SIBs must adhere to the aforementioned criteria to ensure the efficient utilization of SIBs in the marketplace.

The electrolyte regulates the battery's overall performance, which includes safety, cycle life, energy density, multiplication ability, and electrochemical window. It also affects the interface chemistry. It is hoped that further high-quality electrolytes will eventually be developed, allowing sodium batteries to be enhanced and improved in all aspects of performance.

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