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Research Progress of Solid Electrolytes in Lithium Batteries

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Abstract. With the continuous advancement of sustainable energy and the increasing demand for high-energy-density batteries, lithium-ion batteries have received much attention. However, lithium-ion batteries have problems such as leakage, short cycle life, and Lithium Dendrite Growth, which result in low battery safety and hinder their development. To address this issue, this article summarizes and classifies four different types of solid electrolytes, which can fundamentally alleviate safety problems such as thermal runaway. Research shows that oxide-based solid electrolytes have a wider electrochemical window and stronger stability compared to the other three types, making them more conducive to breaking through the limitations in energy density. However, the excessively high cost of this material has severely hindered its commercialization process. Polymer-based electrolytes, on the other hand, possess good flexibility and strong processability, but they are also prone to deformation, which affects the performance of the battery. The oxidative stability of halide electrolytes is good, and they can be matched with high-voltage positive electrode materials. However, their strict preparation conditions increase the operational difficulty and cost, and thus pose significant challenges in practical applications. Finally, the sulfide electrolyte has a high ionic conductivity similar to that of liquid electrolytes. However, the generation of many by-products leads to the problem of unstable interfaces. Additionally, this paper discusses the exploration of new materials and the application of artificial intelligence in the future development of solid electrolytes.

Keywords: Lithium-ion batteries, solid-state electrolytes, development prospects.

1. Introduction

With the continuous development of human industrialization and the continuous innovation of technology, the demand for energy in human society is increasing rapidly. At present, fossil energy remains the mainstream energy, among which coal holds the dominant position. However, this type of primary energy has the characteristic of being non-renewable, and its adverse effects on the environment are gradually increasing: First, the reserves of fossil fuels are limited. Excessive extraction will lead to resource depletion and land desertification. Second, burning fossil energy releases substances such as sulfur dioxide and nitrogen oxides that seriously harm human health, causing ecological problems like smog and acid rain. It will also release a large amount of carbon dioxide, intensify the greenhouse effect and pose a severe challenge to sustainable development. Therefore, electrification is becoming the direction of social development. It replaces coal with

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electricity to improve efficiency and reduce greenhouse gas emissions at the source [1]. Lithium-ion batteries, due to their high energy density and mature technology, have become the mainstream in the power battery market and are widely used in areas such as electric vehicles. However, this traditional liquid lithium-ion battery has significant safety issues: First, the liquid electrolyte contains flammable organic solvents such as ethylene carbonate (EC) and dimethyl carbonate (DMC), which can easily decompose at high temperatures, generating combustible gases and causing thermal runaway. The battery temperature will rise sharply, leading to serious consequences such as explosion. Secondly, during the charging and discharging process, the liquid electrolyte decomposes, generating gas that causes the battery to expand, thereby reducing its cycle life. Thirdly, on the negative electrode of lithium metal, lithium dendrites will form. During the charging and discharging process, these dendrites grow and pierce the separator, causing a short circuit and posing a significant bottleneck for the improvement of energy density [2]. To overcome the above problems, people gradually focused on solid-state electrolytes. Solid-state electrolytes replace the flammable organic electrolytes, fundamentally reducing the risks of thermal runaway and battery expansion, enhancing safety, and also minimizing the risk of harmful substances polluting the environment. They have demonstrated progress in recycling and environmental protection. Furthermore, the separated-free structure optimization of the battery has reduced its weight. The solid-state electrolyte can also inhibit the growth of lithium dendrites, thereby further enhancing the theoretical energy density. Additionally, the solid-state electrolyte can reduce the occurrence of electrode side reactions and increase the cycle life of the battery [3]. Although solid electrolytes have shown promising application prospects, they still encounter numerous challenges in practical applications: the ionic conductivity at room temperature is significantly lower than that of liquid electrolytes, the interface contact impedance is high, and the cost is high, all of which hinder the process of commercialization. Therefore, this article will focus on the currently mainstream solid electrolytes, aiming to summarize the advantages, disadvantages and application scopes of different solid electrolytes. This article reviews four types of solid electrolytes and proposes suggestions for future development, providing reference and inspiration for the research on solid electrolytes, and promoting the research and development of solid electrolytes.

2. Classification and advantages/disadvantages of solid electrolytes

Solid-state electrolytes (SSEs) are materials that conduct ions rather than electrons in a solid state, and can be used to replace the liquid electrolytes in traditional lithium-ion batteries to achieve the transmission of lithium ions. Since the electrolyte acts as an organic solvent and has the characteristics of being flammable and volatile, this innovative energy storage technology significantly reduces the problems such as fire, leakage, easy decomposition, spontaneous combustion and even explosion caused by liquid electrolytes. The solid-state electrolyte can be matched with high-voltage positive electrode materials and lithium metal negative electrodes, and has good electrochemical stability, thereby improving the energy density and cycle life [4]. However, the ionic conductivity of solid electrolytes at room temperature ranges from 1×10^{-3} to 1×10^{-7} $S\cdot cm^{-1}$, which is significantly lower than that of liquid electrolytes. This results in a smaller number of ions passing through per unit time, posing a challenge in terms of battery capacity. The contact area between the solid electrolyte and the electrode is small, leading to the formation of voids and the formation of a space charge layer (SCL) with high resistance, thereby affecting the battery performance [5].

Solid-state electrolytes are mainly classified into four types based on their material types: oxide-based, polymer-based, halide-based, and sulfide-based. Each type of material has its own advantages

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and disadvantages in terms of ionic conductivity, chemical stability, and interface compatibility, and they have different application scopes. In the following sections, we will provide detailed explanations for each of these four types of solid-state electrolytes.

2.1. Oxide-based solid electrolyte

Oxide-based solid-state electrolytes are materials composed mainly of metal oxides and are used for ion transport in solid-state batteries. For example, sodium super ionic conductors (NASICON) (Li1+xAlxGe2-x(PO4)3, LAGP), garnets (Li7La3Zr2O12, LLZO), and sulfides (Li6PS5X, where X = Cl, Br, I) etc [6]. This type of electrolyte has rapid ionic conductivity, maintains its structure and performance even at high temperatures, has a low risk of thermal runaway, and exhibits excellent thermal stability.The oxide-based solid-state electrolyte has a wide electrochemical window, which means it can operate within a wider range of voltages, thereby increasing the voltage of the electrode materials and facilitating the improvement of energy density. Moreover, its high mechanical strength enables it to resist the volume changes of the electrode materials during charging and discharging, ensuring that the electrolyte maintains effective contact with the electrode. The oxides are not sensitive to the air and are easy to be processed, showing good chemical stability towards lithium metal [7]. However, there is an impedance between the oxide-based materials and the electrode materials, which will hinder the ion transmission at the interface, resulting in a high rigidity. This can be solved by artificially designing the SEI layer, constructing 3D conductive collectors, and other methods. Some oxide-based solid-state electrolytes have high costs, such as in terms of material costs. Nickel (Ni)-based materials are more expensive than stainless steel, but they can increase power density. In terms of manufacturing costs, when an additional thin diffusion barrier layer is added to prevent chromium evaporation, the cost increases. One possible way to reduce costs is to optimize the structure and decrease the thickness of the single battery [8]. Finally, oxide-based batteries are suitable for applications with high safety requirements. They can be used in types of solid-state lithium-ion batteries such as thin-film batteries, micro batteries, and high energy density batteries. They can also be used in solid oxide fuel cells (SOFCs). For more details, please refer to Figure 1. Metal-supported solid oxide fuel cells (MS-SOFs) compared to ES-SOFCs and AS-SOFCs have potential in terms of electrochemical performance improvement, stability, and industrial production. However, their complex manufacturing process and high cost prevent them from expanding the market scale. The oxide-based electrolyte can conduct oxygen ions at high temperatures to complete the electrochemical oxidation of the fuel.

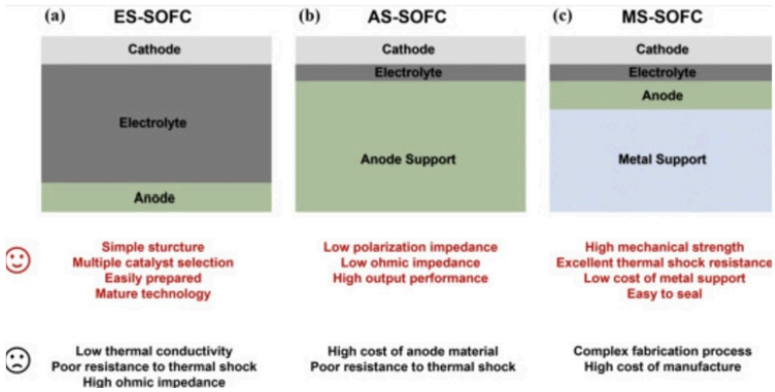

Figure 1. The advantages and disadvantages of the three common types of SOFCs (ES-SOFC, AS-SOFC and MS-SOFs) [8]

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2.2. Polymer-based solid-state electrolyte

Polymer-based solid electrolytes are usually composed of a high-molecular-weight polymer matrix (such as polyethylene oxide (PEO), polyethylene glycol (PEG), etc.) and alkali metal salts (such as lithium ions, sodium ions, potassium ions, etc.). The polymer serves as the framework, providing channels for the ions to conduct and achieve ionic conductivity. Compared with inorganic solid-state electrolytes (ISEs), polymer-based solid-state electrolytes have better film-forming and processing properties. This is because polymers possess excellent flexibility, processing ability and film-forming capability, making it easy to prepare large-area and thin electrolyte membranes. This enables more possibilities for the shape of the battery. Compared with other electrolytes, the cost of polymers is usually lower and shows potential in large-scale production [9].

Furthermore, in today's society, many biodegradable polymers are being studied. If these polymers are used as electrolytes, it will be more environmentally friendly and in line with the development concept, contributing to sustainable development. However, polymers are generally amorphous at room temperature. For example, polymers like polyethylene oxide (PEO) have the characteristic of easy crystallization, resulting in low ionic conductivity at room temperature, which has a significant impact on practical applications [10]. Some polymers have narrow electrochemical windows and cannot be applied in high-voltage batteries. They are also prone to deformation, resulting in poor mechanical strength and affecting the stability of the batteries. Moreover, they cannot inhibit dendrite growth. Finally, this polymer-based solid-state electrolyte is suitable for various solid-state batteries, such as lithium-ion batteries, lithium-metal batteries, sodium-ion batteries, and potassium-ion batteries. Due to its good flexibility, it is applicable to portable electronic devices and flexible batteries.

2.3. Halide-based solid electrolyte

Halide solid electrolytes are ionic conductors composed of lithium, metal elements, and halogen elements, with the general formula Li—M—X. Lithium ions migrate within the octahedral structure formed by these elements. For example, such materials include Li_3YCl_6, Li_3InCl_6, and Li_2ZrCl_6, etc. The advantages of halide-based SSE include high ionic conductivity, exceeding 1 mS cm^{-1} at room temperature. For example, Li_3InCl_6 can reach 1.49 mS cm^{-1}, and Li_3YBr_6 can reach 1.7 mS cm^{-1}. It also has good oxidation stability and can be directly matched with high-voltage cathode materials. The synthesis of some halide solid-state electrolytes is simpler and can be prepared through methods such as the aqueous phase method (Li_3InCl_6) and solution method, which indirectly reduces the cost and is conducive to practical applications. Compared with oxide SSE, halides have better ductility and plasticity, which helps achieve close contact between electrode materials and thereby improve the problem of high interface resistance [11]. However, apart from fluoride, most halides have high hygroscopicity and are prone to react with water, generating corrosive gases (HCl/HBr), which leads to a decrease in ionic conductivity and affects the stability and other properties of the battery. The metal cations in halides are easily reduced to metallic elements, resulting in continuous interfacial reactions and battery short circuits. The preparation conditions for halides are very strict. Most halides need to be synthesized under an inert atmosphere, which increases costs and operational difficulties, and thus affects large-scale commercialization [12]. Halide-based solid-state electrolytes are suitable for uncoated cathodes because they have high oxidation stability and can be directly compounded with the cathode materials. They are applicable to high-voltage all-solid-state batteries and can be directly matched with materials with voltages above 4V (such as $LiCoO_2$, NCM, NCA,

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etc.). With the continuous development of sodium-based halide SSEs like Na_3YCl_6 and Na_2ZrCl_6, their potential has also been demonstrated in sodium-ion batteries.

2.4. Sulfide-based solid-state electrolyte

Sulfide-based solid-state electrolytes are solid-state electrolytes in which the main anions are composed of sulfur elements. They mainly include sulfide glasses (such as the Li_2S - P_2S_5 system), sulfide glass ceramics (such as $Li_7P_3S_{11}$), and crystalline sulfides (such as thio-LISICON, LGPS, Argyrodite, etc.). The sulfide solid electrolyte has a high ionic conductivity, reaching up to 10^{-2} S cm^{-1}, which is close to that of liquid electrolytes. Sulfide is a soft material and exhibits excellent mechanical properties. Its good ductility and low hardness enable the improvement of the interface contact between the electrodes, and this material itself has a very low interface resistance with the lithium metal anode. Sulfide solid-state electrolytes can be prepared by various methods, such as ball milling, hot pressing, and liquid-phase synthesis, and the preparation methods are usually simpler than those of other types of solid-state electrolytes (such as oxide SSE) [13]. Such electrolytes still face significant challenges, such as their tendency to react with water to produce H_2S, a toxic gas that poses a major safety hazard in subsequent processing and is a major issue throughout the battery's entire lifecycle. Its electrochemical window is narrow, which severely limits its application in high-pressure batteries. Besides, there is also a problem of unstable interface. Further speaking, it is prone to form a space charge layer (SCL) with the positive electrode interface and react with sulfate, polysulfide and other reaction by-products. It may also form a mixed conductive layer with the lithium metal negative electrode, thereby continuously decomposing the electrolyte. As shown in Figure 2, sulfide solid-state electrolytes will undergo electrochemical side reactions in lithium metal batteries and the formation of SEI layer can be observed. The relevant optimization strategies are given in the figure, such as alloying the lithium negative electrode with a less active metal or designing and constructing an artificial SEI layer. The good flexibility of this material also leads to its insufficient mechanical strength, making it prone to lithium dendrite penetration. Finally, the sulfide SSE is suitable for both silicon-based anodes and lithium metal anodes because it can accommodate their volume changes, and thus has the potential to achieve breakthroughs in energy density, making it suitable for applications in electric vehicles and portable electronic devices, and enhancing safety [14].

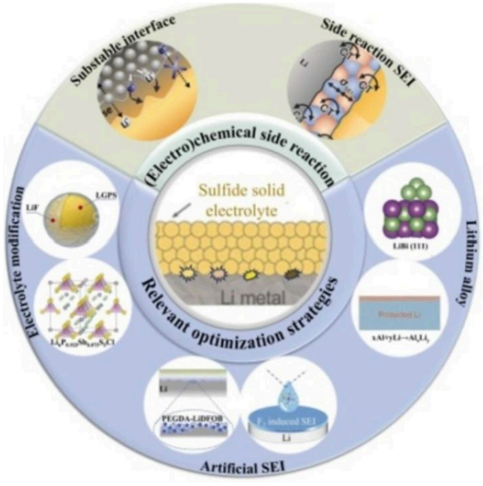

Figure 2. Interface side reactions of sulfide solid-state electrolytes, formation of the SEI layer, and improvement strategies [15]

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3. Prospects of solid electrolytes

Solid-state electrolytes include oxide-based solid-state electrolytes, polymer-based solid-state electrolytes, halide-based solid-state electrolytes, and sulfide-based solid-state electrolytes, etc. Among them, the ionic conductivity of most materials still needs to be improved, and the problem of lithium dendrite penetration remains a significant issue. Secondly, its contact resistance with the electrode material is relatively high. Therefore, it is an important direction to develop materials with higher mechanical strength and ionic conductivity. Solid-state electrolytes, as a key component of next-generation energy storage batteries, tend to innovate in material development. Incorporating the application of artificial intelligence, simulating and analyzing material screening, and predicting performance, have made the development of solid-state electrolytes full of potential.

4. Conclusion

This article conducts a systematic study on solid-state electrolytes, revealing that different solid-state electrolytes currently exist due to their distinct properties, and are applicable to various fields. Moreover, the main challenges faced by these solid-state electrolytes also vary. The oxide solid-state electrolyte has high safety and can be applied in thin-film batteries. However, it faces problems such as high resistance against the electrode materials and the need to use nickel as the material, which leads to higher costs. Polymers with good flexibility can be used in flexible devices. However, some polymers tend to crystallize at room temperature, such as polyethylene oxide (PEO), and their flexibility also leads to the problem that lithium dendrites are more likely to penetrate. The high oxygen stability of the halides enables them to directly react with the positive electrode materials, making them suitable for high-voltage all-solid-state batteries. However, most of the halides are prone to react with water to produce corrosive gases, and the metal cations within them are easily reduced to elemental forms, causing battery short circuits. Sulfides are most likely to achieve breakthroughs in energy density and be applied in the electric vehicle sector. However, they react easily with water to produce toxic gases, which is inconsistent with the green development concept. Moreover, they have problems such as unstable interfaces and a narrow electrochemical window. From this, it can be seen that the commercialization of various solid electrolytes is still hindered by multiple factors such as cost, conductivity, interfacial resistance, and toxic by-products. Therefore, in the future, this industry must continuously develop and research new materials, deepen the relationship and influence of materials on solid electrolytes, in order to achieve large-scale commercialization. At the same time, the research mentioned the assistance of artificial intelligence, which is conducive to accelerating the development speed of solid electrolytes and achieving breakthroughs and applications sooner.

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