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Ionic Transport in Solid Electrolytes: Conductivity, Interfacial Challenges, and Optimization Strategies

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Abstract. Solid-state batteries are different from traditional liquid lithium-ion batteries because they do not use flammable organic liquids for the electrolyte, and are therefore safer, with high energy density and a long cycle life. They are the main development directions for the next generation of energy storage systems and power sources. The ionic conductivity of the solid electrolyte affects the speed of charging and discharging for a cell, changes in internal resistance during operation, etc.; now, it has become one of the main technical bottlenecks preventing its industrial application. First, the whole structure and functions of solid-state batteries will be introduced in this paper. Then, it will gather the current methods for evaluating ionic conductivity and the present situation of research on ion transport paths. Finally, the technical attributes and applications of the four fundamental kinds of solid electrolytes will be introduced: polymer-based, oxide-based, sulfide-based and halide-based. Based on the systematic comparison of the strengths and weaknesses of all these links, this paper presents the current development stage of each link and points out problems that need to be solved to promote progress in the exploration and construction of high-performance solid-state batteries.

Keywords: Solid-state battery, ionic conductivity, solid electrolyte

1. Introduction

With the development of electric vehicles, portable electronic devices and large-scale energy-storage systems in recent years, people's demand for battery performance has increased, and thus the required standards have also been changing accordingly. Although there have been some advances in recent years, many problems with liquid lithium-ion batteries have not been solved yet. An electrolyte is required; since it is volatile and flammable, it is likely to leak, evaporate or be ignited by heat in bad weather. The above are serious safety problems of new-generation energy storage systems and power modules.

A solid-state membrane is used in the structure of solid-state batteries to conduct ions and prevent leakage; thus, it can be used instead of liquid electrolytes. Given that it has good chemical stability, a high-capacity anode and cathode material can be used to increase the energy density. Therefore, it has begun to be explored in the field of renewable energy and is now attracting attention from both research institutions and enterprises. At present, the industrial application of all-solid-state batteries

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faces many problems. The two main reasons for the low ionic conductivity and high electrochemical interface resistance of solid-state electrolytes are unknown. The reasons for this are due to all sorts of reasons, such as crystal lattice structure and order, phase transitions, grain boundary effects, surface characteristics, etc.; as a result, solid-state membranes have conductivity at room temperature that is many orders of magnitude lower than that of liquid electrolytes. The deficiencies mentioned above will have an adverse effect on the charging and discharging performance of the battery, significantly increase both the internal resistance and polarisation, and are thus not suitable for practical application [1].

Bibliometric analysis has been used to organise the information on the ionic conductivity of polyelectrolytes, inorganic salts (oxides, sulfides, halides), metal-organic frameworks and new solid-state electrolyte structures. The following four kinds of technical strategies have also been used in the study of these materials: Material design and defect remediation; Synthesis of multicomponent composites and interface modification; Advanced manufacturing techniques; and Data-driven modelling. In recent years, the four directions of research and their mechanisms of action for various types of electrolytes have been summarised in this paper: creation of constituents and structural defects, construction of multicomponent composite materials and improvement of interfaces, advanced manufacturing technology, and data-driven model construction. The above work has offered good scientific basis and technical support for the design and development of good solid-state electrolyte systems.

2. Fundamentals and evaluation metrics of ionic transport

2.1. Basic structure of solid-state batteries

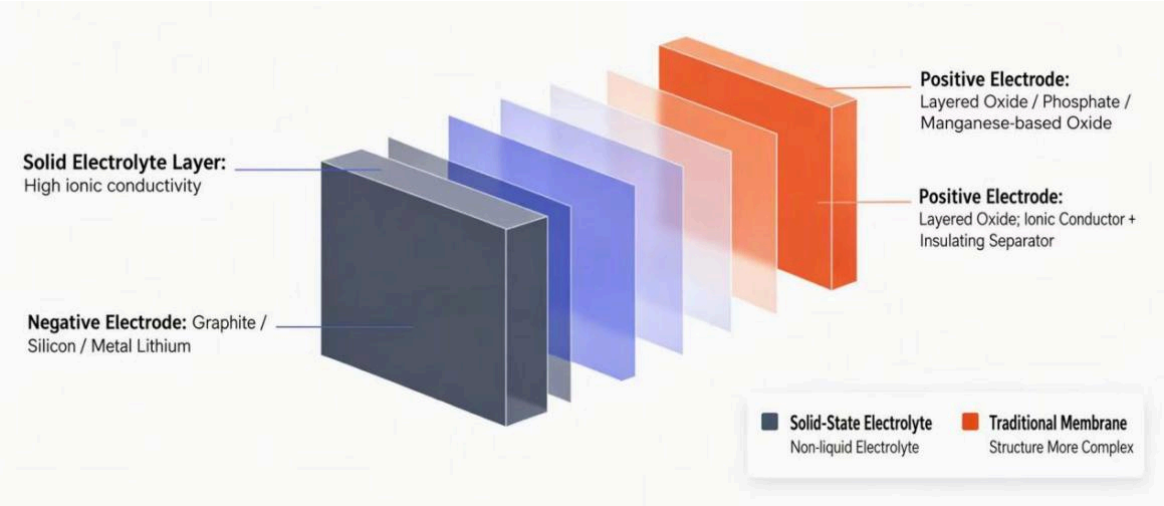

Figure 1. Three-layer structure comprising the cathode, solid electrolyte, and anode (photo credit: [2])

All-solid-state batteries are a new kind of energy-storage device that generally consists of cathodes, solid electrolytes and anodes. They do not use the traditional liquid electrolyte but have developed composite electrodes and inorganic solid-state interfaces. The three parts of these batteries are a high-efficiency cathode material, a high-performance solid-state electrolyte and a fine-grained anode material. High-efficiency cathode materials can be the traditional lithium-ion battery cathode materials, such as layered oxides and phosphates, or high-energy-density materials,

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such as sulfur-based cathodes or air cathodes. The choice of a high-performance anode material is very important; there are many kinds, such as graphite, silicon-based alloys, metallic lithium, etc. Given its very high theoretical capacity, the metallic lithium anode is expected to have excellent energy density [2]. Although a solid-state electrolyte can conduct lithium ions, there should still be no internal short-circuit at the same time. The first principle of all-solid-state battery innovation is the idea of a module, as shown in Figure 1.

2.2. Key conductivity indicators of solid electrolytes

Conductivity is a necessary indicator of the transport efficiency of lithium-ion in solid-state electrolytes, and it can be used to determine the discharge/charge rate and energy density of solid-state batteries. To determine the electrical properties of a material, one needs to obtain relevant data on the interface impedance, activation energy and lithium-ion diffusion coefficient, etc., and study how these factors are related to its application.

Interface impedance is the electrochemical coupling between the electrolyte and the electrode layer, and it varies greatly in solid-state structures. Given that the contact area of solid-solid interfaces in crystalline materials is small and they generally have microstructures of voids and defects, the contact resistance is generally much higher than that at solid-liquid interfaces in liquid-based batteries. Therefore, the efficiency of lithium-ion circulation will be relatively low. The activation energy for ionic conduction is the energy that needs to be supplied to move lithium ions in the electrolyte; a large value means that changes in the surrounding environment will have a significant impact on the speed of diffusion and thus alter the power requirements. Although the carrier density is also related to conductivity, this connection is not direct. Transfer coefficient refers to the amount of lithium-ion movement that can occur in a battery; that is, it is the ratio of the current carried by genuinely mobile lithium ions to the total conductive current [3]. Under ideal circumstances, it should be 1; then, the concentration polarization effect will be reduced, and the charging and discharging capacity will be higher.

Many ion-conducting paths are present in various solid electrolyte materials. The movement of ions in polymer frameworks is mainly due to thermodynamic chain rearrangement, and this is affected by changes in temperature. Given that it contains crystalline parts, the movement of polyethylene oxide (PEO) units is restricted at low temperatures, and thus its electrical conductivity is low. Oxide-based solid electrolytes are conductors that have structural vacancies or ionic sites, and lithium ions move by hopping in these lattice openings. Sulfide-based systems have a small activation energy and good electrical conductivity at room temperature because of the strong polarizability of sulfur atoms and the low energy of the lithium-sulfur bond. Halide-based electrolytes have various anion structures and can form three-dimensional or multi-dimensional high-performance, fast lithium-ion transport paths; therefore, they have good electrical conductivity.

3. Comparison of conductivity performance of mainstream solid electrolyte materials

3.1. Polymer solid electrolytes

The first constructed and most technically advanced solid electrolyte framework is the polymer-based system, which is generally prepared by solution blending and thermal curing. The two are a polymer base and lithium salts. Polyethylene glycol, polycarbonate and polyacrylonitrile (PAN) have good properties and are used widely. Polyethylene glycol-derived polymer electrolytes have

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good properties for practical use, such as high ionic conductivity and structural stability; therefore, they are now attracting the attention of many researchers around the world.

Solid-state polymers have a relatively high ionic conductivity above their melting or glass transition temperature, but they are generally too low at room temperature. Polymer-based electrolytes, such as polyethylene oxide (PEO), have an ionic conductivity of 10^{-6} - 10^{-5} S/cm at room temperature and do not meet the high-rate charging and discharging demands. As the ambient temperature reaches 60-80 °C, the increased thermal agitation of polymer backbone molecular units will open up ion-conducting pathways, and the conductivity will be about 10^{-5} - 10^{-4} S/cm; at this time, it will be close to or exceed that of some liquid electrolytes. The lithium-ion migration numbers in the above materials are generally between 0.2 and 0.5; thus, concentration polarization may occur and harm both the rate performance and cycle life of the battery [4]. As shown in Figure 2, the main source of energy for ion transport in polymer electrolytes is the thermal motion of molecular units.

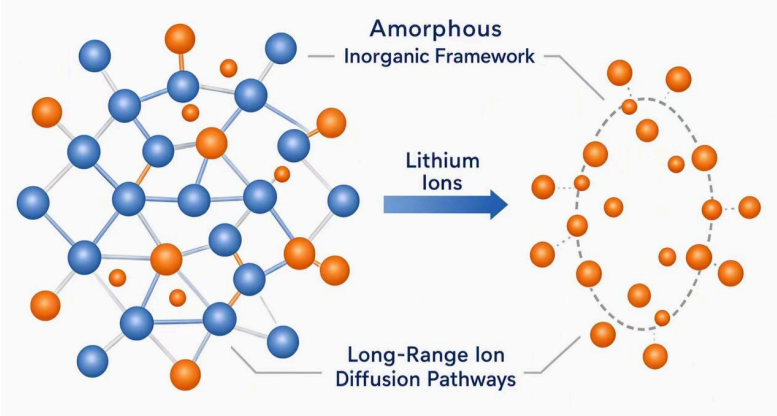

Figure 2. Polymer electrolyte: conduction by chain-segment motion (photo credit: [4])

3.2. Oxide solid electrolytes

Inorganic solid-state electrolytes are a type of crystalline material, and they can be divided into two main categories: crystalline oxides and amorphous glass oxides. Among them, some oxide materials have received extensive research in recent years due to their good ionic conductivity and thermal stability, such as garnet-type, NASICON-type and LISICON-type oxides; garnet-based oxide electrolytes have been particularly popular in recent times [4].

Research on the electrical conductivity of solid-state electrolytes has found that oxide-based materials have some advantages over polymers; at room temperature, their electrical conductivity is higher than that of polymers, and they also possess good thermal stability. Additions of La^{3+} have been made to increase the ionic conductivity of the NASICON-based material $LiZr_2(PO_4)_3$ to 0.72×10^{-4} S/cm at room temperature; at the same time, the activation energy has been reduced from 0.53 eV to 0.42 eV and the mobility of lithium ions is close to its maximum value (approximately 0.99). Structural disorder can be introduced in the garnet-based system by substituting some elements at the Zr site, as shown in the structure of $Li_6La_3Zr_{0.5}Ti_{0.5}Ta_{0.5}Nb_{0.5}O_{12}$, to increase the conductivity to 1.52×10^{-3} S/cm and reduce the activation energy to 0.18 eV. Solid-state electrolytes have a small activation energy for ion conduction compared with polymer-based ones. They have a small range of temperature-sensitive change, are more thermally stable and fire-resistant than polymer-based electrolytes, and therefore have good safety characteristics. As shown in Figure 3, ionic transport in oxide electrolytes is generally carried by the movement of crystal defects and vacancies.

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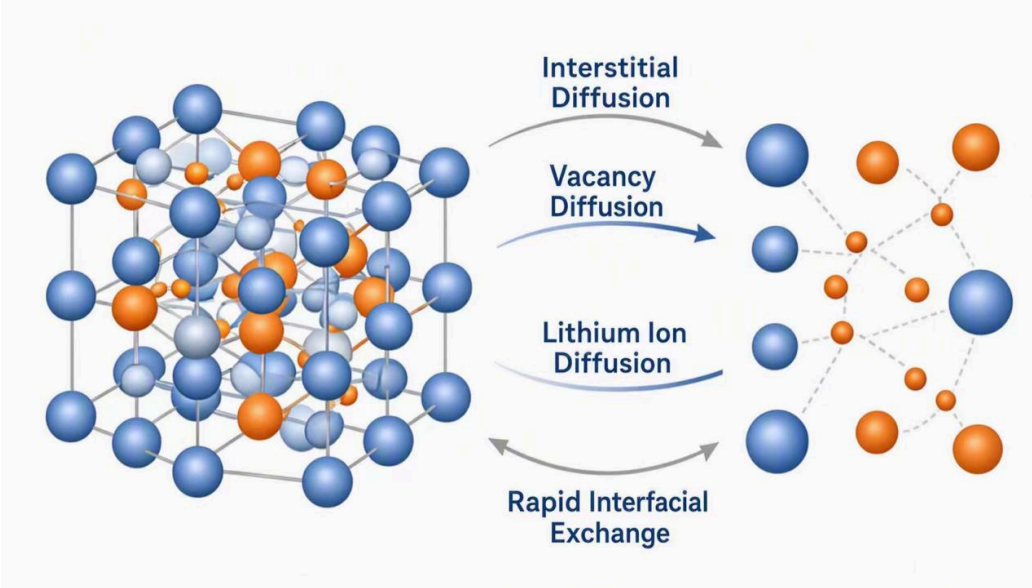

Figure 3. Oxide electrolyte: conduction through lattice defects (photo credit: [5])

3.3. Halide solid electrolytes

In recent years, halide solid-state electrolytes have gradually drawn the attention of scholars in various fields as new kinds of inorganic dielectrics. The basic parts are lithium, transition metals and all kinds of halogen-containing substances, such as fluorine, chlorine and bromine; Li_3InCl_6 and Li_2ZrCl_6 are typical cases. They have a good voltage plateau stability and good interfacial bonding, so they are suitable for use with high-nickel anode materials. They have effectively solved many of the problems of traditional sulfide and oxide electrolytes in actual use and are very suitable for the development of high-voltage battery systems [6].

Based on the investigation of ionic conductivity, most halide-based electrolytes have values of 10^{-5} - 10^{-4} S/cm. Although they are much better than the traditional oxide matrix, they have not yet achieved the high performance of optimised sulphide alternatives. Improve the nanostructure and add dopants to increase the electrical conductivity to more than 10^{-3} S/cm, so that it can meet the operating conditions of power batteries. Research has found that the ionic migration activation energy and a relative small amount of heat are relatively small in this group of electrolytes, but they are not always so. The lithium-ion transference numbers of these systems are generally above 0.9; therefore, there will be less interfacial polarisation, and the battery will have better cycle life and energy capacity.

3.4. Sulfide solid electrolytes

Due to their high energy-storage capacity, sulfide solid-state electrolytes have attracted a lot of attention in the field of solid-state batteries repeatedly. The first kind of system is based on sulfur-containing lithium salts, and by adding germanium, phosphorus or silicon, inorganic amorphous or polycrystalline phases can be formed, such as Li_2S-P_2S_5, Li_2S-GeS_2-P_2S_5 and Li_6PS_5Cl. Compared with the traditional oxide electrolyte, the sulfide electrolyte is relatively flexible, has good interfacial bonding, and can also be produced via a low-temperature cold-pressing method to reduce the total production costs [7]. Due to alterations in the chemical composition and activation energy of these materials, their electrochemical characteristics change with different temperatures, and thus they

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must be fully evaluated and optimised at various operating temperatures before being used in practice.

3.5. Performance comparison of mainstream solid electrolyte materials

All-encompassing and systematic tests were carried out on the electrical conductivity, utilisation spectra and general performance of the four main solid-state electrolytes, and the resulting data have been organised and analysed. The main indices that will be examined are the ionic conductivity at room temperature, lithium-ion velocity, service temperature limits, interface stability, high-voltage resistance and commercial viability; all this information is shown in Table 1.

Table 1. Performance comparison of mainstream solid electrolyte materials

Electrolyte Type	Room-Temperature Ionic Conductivity (S/cm)	Room-Temperature Ionic Conductivity after Modification (S/cm)	Lithium-Ion Transference Number
Polymer Solid Electrolyte	10^{-6}~10^{-5}	10^{-5}~10^{-4} (inorganic filler doping) [3]	0.2~0.5
Oxide Solid Electrolyte	10^{-4}~10^{-3}	10^{-4}~10^{-3} (aliovalent ion substitution and medium-entropy design; e.g., LLZO reaches 1.52×10^{-3}) [3, 5]	≈1.0
Sulfide Solid Electrolyte	10^{-3}~10^{-2}	10^{-2} (process-parameter optimization and composition control) [5, 6]	0.8~0.9
Halide Solid Electrolyte	10^{-5}~10^{-4}	10^{-3} (elemental-composition control and nano-modification; e.g., $LiMOCl_4$ reaches 12.4 mS/cm) [6]	≥0.9

4. Optimization strategies for improving the conductivity of solid-state batteries

4.1. Optimization through material doping and modification

Strontium titanate is one of the materials for solid-state electrolytes, and its crystal lattice can be altered by adding various dopants or other phases. Reduce the activation energy for ion migration, increase the carrier concentration, and so on, to improve the performance of the four main types of solid-state electrolyte systems.

Most of the research on solid-state polymer electrolyte frameworks has focused on increasing the room-temperature ionic conductivity through the addition of inorganic filler doping. Based on the above empirical data, the addition of nanometre-sized inert ceramic particles, such as alumina, titanium dioxide and silica, to a polymer matrix can reduce the formation of crystalline regions and thus decrease both the total crystallinity and the regularity of the arrangement among polymer chains. Therefore, the thermal stability and mechanical strength of the electrolyte will be improved, so it can better withstand external mechanical stress and reduce interface wear caused by dendrite growth. The nanoparticles can form a network to conduct lithium-ion transport by spreading out the

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material more evenly; thus, they are able to reduce the distance that ions need to travel and improve the performance of the battery at room temperature [7].

In the research on oxide solid-state electrolytes, to increase the electrical conductivity, one can introduce specific substitutions that modify the valence of the ions. $LiZr_2(PO_4)_3$ is a NASICON-type material, and by substituting some Zr^{4+} ions with La^{3+}, the charge is reduced; thus, to maintain charge neutrality and ensure the thermodynamic stability of the rhombohedral crystal structure, some lithium ions (Li^+) must be redistributed. According to the above research, this method can increase the ionic conductivity from the initial 10^{-6} S/cm to 0.72×10^{-4} S/cm and reduce the activation energy from 0.53 eV to 0.42 eV [8]. To improve the modulation of lattice strain effects in garnet-type $Li_7La_3Zr_2O_{12}$, a medium-entropy design strategy has been applied, and some substitution of Zr with other elements such as Ti, Ta and Nb has also been carried out to regulate the strain effect. Therefore, the migration energy of lithium ions is about 0.18 eV, many low-energy displacement mechanisms have arisen, interfacial bonding has been strengthened, and overall performance has improved; as a result, the ionic conductivity at room temperature is 1.52×10^{-3} S/cm [9].

Optimisation of the Synthesis Process for Solid-State Electrolytes is to be carried out to Improve the Efficiency of Sulfide Compounds. $Li_7P_3S_{11}$ prepared by high-energy ball milling and combined with cold-state solid-state synthesis has an ionic conductivity several times higher than that of samples prepared by the traditional ball-milling method, and it was found that its crystal structure is in the conventional LPS type [10]. Ionic conductivity is a typical indicator of the performance of sulfide systems, and it is very sensitive to changes in the laboratory. Kalyk and others have shown that isostatic pressing, uniaxial pressure states, contact interface properties and electrochemical impedance spectroscopy (EIS) evaluation factors can all have some influence on the final conductivity results [11]. Therefore, a general and standardised characterisation plan needs to be established to guarantee the accuracy of the data obtained in the doping or functionalisation step.

Based on the above research, altering the chemical composition of the components in solid electrolytes can be done to improve their electrical characteristics. Recently, researchers have introduced a new type of lithium metal chloride, $LiMOCl_4$ (where M is Nb or Ta), which has been found to have a high ionic conductivity of 12.4 mS/cm at room temperature and is far better than that of traditional liquid electrolytes [12]. Further optimisation of the ball-milling conditions and the high-temperature sintering process can be carried out to improve the conductivity of the $Li_{2.25}Zr_{0.75}Fe_{0.25}Cl_6$ electrolyte and enhance its all-around electrochemical performance; thus, both the total operating efficiency and the reliability of this specific energy storage system will be improved [13].

4.2. Construction of composite electrolyte systems

A Way to Achieve Many Different Properties is to Use Multiphase Electrolyte Compositions. Generally speaking, the basic structure of such frameworks is a stretchable polymer, and to form a good interface with inorganic fillers that have high electronic conductivity, these fillers are added; therefore, they are called "flexible matrix + conductive phase hybrid" structures.

Polymer/oxide hybrid electrolyte frameworks have a flexible polymer substrate and good interfacial bonding. Thus, it will reduce the contact obstruction caused by the stiffness of traditional oxide electrolytes, and it is hoped that damage from dimensional change in the battery during discharge over time can be avoided. Oxide reinforcements are added to increase the total conductivity of these structures by forming a good ion-conducting path and to solve the problem of low conductivity in the base polymer electrolyte at room temperature. Anik and others have carried

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out research and found that the BJAM method can increase the packing density and improve the homogeneity of the NaSICON/PEO hybrid sodium-ion solid electrolyte; thus, its overall electrochemical performance is enhanced [14].

Sulfide electrolytes that can be cured at room temperature have good electrical conductivity and are also suitable for flexible production in polymer-based sulfide composite frameworks. During the synthesis stage, it is necessary to control the size distribution of particles. Although mechanical ball milling can reduce the size of particles and improve the strength of interparticle bonds, excessive grinding may cause lattice defects and thus affect ion transport. In the optimisation of particle refinement, one must consider the stability of electrical conductivity to achieve an optimal balance between function and durability [15].

Composite oxide-halide structures have good synergy; high-temperature thermal stability is due to the different characteristics of the oxide matrix, and high-voltage resistance is provided by an excellent halide matrix. Lee and others have found that when sintering a glassy electrolyte with the composition of Li_2O-B_2O_3-Al_2O_3 at 520 °C, grains form and good interfacial contact is achieved with the electrode substrate; as a result, the lithium-ion mobility at room temperature is 3.075×10^{-5} S/cm [16]. Kurihara's group has improved the amorphous structure of the Li_2O-Al_2O_3-GeO_2-P_2O_5 system, and through low-temperature annealing in the range of 400-700 °C, NASICON-type solid electrolytes with conductivities of 10^{-5} - 10^{-4} S/cm have been obtained; in addition, the addition of MgO has also increased the total conductivity of this system significantly [17]. Sastre and others have conducted many studies on the garnet-type $Li_7La_3Zr_2O_{12}$ compound and have provided comprehensive analyses of the reasons for changes in its bulk and thin-film conductivity. This paper has provided a theoretical basis for the use of oxide-based solid-state battery electrolytes and put forward some optimisation ideas [18].

Recently, data-driven methods with machine learning have been applied to the design of composite electrolytes and prediction of electrical conductivity and have shown good results. Gamal and others have applied machine learning to the problem of electrochemical impedance spectroscopy. PCA was used to reduce the dimensionality of the data, and then the SelectKBest method for feature selection was applied. Based on the above, they have constructed a very accurate prediction model for ionic conductivity in solid-state electrolyte systems using the ElasticNet regression method, and the R^2 value is 0.9876. This paper has added to the existing knowledge on the reasons for changes in the performance of composite electrolytes and offers many practical suggestions for the intelligent development of related materials [19].

4.3. Research outlook

With the development of the car industry, so too has the demand for high-energy-density batteries that are safe and have a long cycle life in large-scale energy storage applications. Existing liquid lithium-ion batteries have many technical problems. Solid-state batteries, as a leading alternative, are based on solid electrolyte substrates and have achieved excellent protection and very good energy storage; therefore, they have rapidly become a focus of research at home and abroad as well as companies [20]. The four main directions for the development of solid-state batteries will be to improve electrical conductivity, increase thermal stability, enhance interface compatibility and promote industrial application.

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5. Conclusion

This paper will first present the general structure of solid-state batteries and then provide specific data on the electrical conductivity, temperature range, interface properties, etc., of four common solid-state electrolytes: polyvinylidene fluoride (PVDF), oxides, sulfides and halides. All of them have different advantages and disadvantages in actual use. The electrolytes listed above have different strengths and weaknesses and can work well together. Polymer-based electrolytes have good manufacturing conditions and excellent mechanical properties, but at room temperature, their ionic conductivity is relatively low; thus, they are generally used in intermediate- and low-temperature energy storage devices. Oxide-based electrolytes are preferred because they have good thermal stability and a high lithium-ion mobility; thus, they can operate normally at various temperatures, but they are inherently brittle and difficult to produce in large quantities.

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