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Challenges and development of solid-state electrolyte interface of Lithium-ion batteries (LIBs) as energy storage tools for future development

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Abstract
The Lithium-ion battery (LIB) as a new energy storage method is widely used in many fields. The safety problems and efficiency problems are the key drawbacks to be solved currently. Traditional liquid-state lithium-ion batteries have problems such as low electrochemical window, dendrite growth, flammability of electrolyte which may cause explosion, and leakage. One improvement of electrolytes is using solid electrolytes to replace liquid electrolytes. Different from liquid-state lithium-ion battery, solid-state battery has many advantages such as wide electrochemical window, high energy density and high safety and so on. In this work, the specific structure of different batteries is introduced and the advantages and drawbacks of liquid-state lithium-ion battery and solid-state battery are given. Besides, this paper also analyzes the necessity and feasibility of solid-state batteries replacing traditional liquid-state batteries. Moreover, the problems which should be paid attention to and development trends of solid-state battery are also being discussed. It lays the foundation for further exploring of the industrialization prospect of solid-state lithium-ion batteries (LIBs).

Keywords: Solid-state, lithium-ion battery, solid electrolyte, electrode- electrolyte, interface, energy tools, future development

Introduction

The development of human society and the consumption of energy is constantly expanding. Traditional fossil energy resources are limited and the pollution caused by them is not negligible, which forces people to turn their attention to new energy sources. However, solar, wind and other new energy sources are seriously affected by weather, so the power supply is less stable. Therefore, a light and efficient energy storage tool becomes particularly important. Lithium-ion batteries (LIBs) are widely used in cell phones and computers as energy storage tools with low price, high energy density, low self-discharge, no battery crystallization effect, long service life and environmental friendliness. With gradual maturation of lithium-ion battery technology, a large number of new materials are gradually developed and the cost of lithium-ion battery is gradually reduced. Currently, lithium-ion batteries are also widely used in automobiles, ships, smart homes, medical devices and other industries. However, there are still many problems in lithium-ion batteries (LIBs). Firstly, how to quickly charge and discharge lithium-ion batteries (LIBs) so as to achieve energy storage as fast as fuel tank refueling. Second how to solve safety problems of liquid electrolyte. Moreover, battery performance parameters such as power density, thermal stability, service life are also widely concerned by the scientific community.

The present manuscript mainly deals with the battery optimized based on electrode materials include lithium-air battery, lithium-sulfur battery and sodium ion battery. These batteries have higher specific energy with a lower cost [1]. However, due to technical reasons, they have not yet been commercially produced. Battery optimized based on electrolytes materials include adding additives and using solid-state electrolytes. Compared with traditional liquid-state electrolytes, solid-state electrolytes are less flammable, less volatile, and less prone to leakage, so solid-state electrolytes are safer than traditional liquid-state electrolytes. In addition, solid-state lithium-ion battery has higher energy density, thermal stability and a longer service life. It can also solve the dendrite growth problem to a certain extent. Except for lithium-ion solid-state battery, new types of batteries such as sodium-ion battery combined with solid-state electrolyte is also a hot research topic.

This manuscript firstly introduces the traditional liquid-state lithium-ion battery, finds out the problems existing in practical application and analyses the solutions of replacing the traditional liquid-state battery by solid-state battery. Then, the composition and working principle of solid-state battery are introduced; different types and advantages of solid-state electrolyte are discussed. Finally, We find out the advantages of solid-state battery and the challenges faced in the future development. Recently, the role of surface modification in enhance electrochemical properties for battery applications [2], transition metal oxides nanomaterial as electrodes for sodium ion batteries [3] have already been studied.

Materials and Methods

Liquid lithium-ion battery: Anode materials, the first commercially available lithium-ion battery was made in 1985 by Nobel laureate Akira Yoshino, who used petroleum coke instead of lithium metal as the anode. This method increased discharge voltage from less than 2 V to 4 V [4]. The anodes of liquid-state lithium-ion batteries are usually made up of graphite which can be used to embedded lithium ions. For charge and discharge process, lithium ions are transferred into new lithium compounds by charging them, and the lithium compounds are changed into lithium ions by discharge process which restore the anode to its original structure. The specific capacity of graphite is 372 mAh/g in theory. Moreover, graphite has a low de-lithium potential and relatively high lithium ion diffusion coefficient. The category of non- metallic anode materials also includes graphene and carbon nanofibers [5]. Alloy anode material is another focus of current research, this material has high specific capacity and high energy density. includes Si, Ge [6]. Compared with the traditional graphite electrode, the specific capacity of the silicon anode can reach 4200 mAh/g, which makes silicon anode widely concern in the scientific community.

Cathode materials: It is a lithium-containing cathode material, which makes it possible for lithium-ion batteries(LIBs) working without supplying lithium by anode [7]. This material is the most widely used commercial cathode material, because of its high charging and discharging efficiency, simple production process and recyclable. However, this material is unstable in high temperature, which is easy to cause safety problems. Compare with $LiCoO_2$ and $LiMnO_4$ has a higher thermal stability, a lower price, and a better lithium ion conductivity [8]. It is also being used in commercial production. However, the cubic crystal structure of $LiMnO_4$ will be transformed into tetragonal crystal structure after excessive embedding of lithium ions, and metal ions or other ions should be added to enhance the stability in the production.

Electrolyte: The electrolyte usually consists of organic solvents, lithium salts and trace additives. The purpose of organic solvents is to dissolve lithium salts and complex with lithium ions for transport. Electrolytes usually require high conductivity, high lithium ion mobility, high stability, high interfacial stability, and wide electrochemical window. Moreover, it is necessary for electrolytes to provide a stable solid electrolyte interface (SEI) layer. In practical application, an environmentally friendly and non-flammable electrolyte is more acceptable.

Separator: The separator is a diaphragm located in the electrolyte that separates anode from cathode. It has two main functions. First, a separator can prevent short circuit contact by separating the anode and the cathode. Secondly, when the temperature is too high, the diaphragm pores can be closed in time to prevent the passage of lithium ions, thus preventing the effect of excessive temperature explosion. Currently, commercial membranes are mainly olefin microporous membranes. This kind of membranes have strong mechanical and electrochemical stability, but the shortcoming is low thermal stability [9].

Working principle: The working principle the most common lithium-ion battery [10] which is made up of graphite and lithium metal oxide as anode and cathode. When the battery works in a discharging state, lithium embedded in graphite loses electrons to become lithium ions, which pass through the electrolyte solution and the diaphragm. Electrons are moved from anode to cathode which form a current and charge the outer circuit. When the battery working in a charging state, the lithium ions in cathode migrate to anode, which are stored in anode as lithium-graphite compounds.

Disadvantages of traditional liquid lithium-ion batteries:

Lithium dendrite growth: Dendrite growth is a normal occurrence of metals electroplating. During the charge process, lithium ions gain electrons from outer circuit and deposit in the anode to form different kinds of dendrites such as needle, whisker, or filament-like dendrites. The implications for lithium-ion batteries are destructive. First, the sites where dendrites are first formed are more likely to deposit, which leads to higher consumption of lithium ions and electrolyte. Second, seriously dendrites growth will cause lithium particles detach from the electrode. And these particles are tightly wrapped by SEI layer that causes the formation of "dead lithium". Besides, "dead lithium" leads to polarization, which causes the battery to deviate from the normal operating state. Most seriously, dendrite growth can break through the diaphragm, which may cause the contact of anode and cathode. It will cause the battery to be short-circuited, even may cause a fire.

Safety: Liquid-state electrolyte is usually an organic solvent, which is easy to volatilize, leak and burn. The first safety problem is the environmental impact of electrolyte leakage, because the organic solvents are polluting. Especially when lithium ion is used in cars and other vehicles, the safety has been paid more attention. Unlike mobile phones and computers, cars usually operate at high temperatures in summer, and there is a risk of collision. Lithium-ion battery(LIB) combines high energy materials and flammable electrolytes. Therefore, when the temperature exceeds the critical temperature, thermal runaway is easy to occur. Besides, one battery explosion in a lithium-ion battery pack can cause the entire battery packs to explode, which leads to an enormous disaster.

Results and Discussion

Composition of Solid-state battery

Anode materials: The graphite anode of liquid lithium-ion batteries, lithium metal can be used in solid-state batteries as an anode. Compared with other electrodes, lithium metal has higher specific capacity and lower electric potential. However, the dendrite growth of lithium is still a difficult problem in lithium ion-lithium elemental cycle of solid-state battery. Dendrite growth not only consumes lithium, but also causes short circuit and other risks. In addition, the nature of lithium elemental is active, which is easy to react with oxygen and water in the air. So, it will directly lead to the cost of battery assembly greatly increased. At present, the best method is to alloy lithium with other metals, so as to reduce the activity of lithium ion, inhibit dendrite growth and side reactions, and increase the stability of the interface. For example, the lithium ions can be used to alloyed with Group IV and Group V elements as cathode to improve the performance of solid-state battery [11]. Although the alloy has many advantages, it may lead to electrode expansion and electrode pulverization in cycle, which will damage the electrode. In order to solve this problem, new alloy materials, composite alloy materials, ultra-fine nano-alloy and other new materials are also being developed.

Cathode materials: The cathode of a solid-state battery usually used a composite electrode which includes active substances, binders and conductive agents. Currently, cathode materials of all-solid-state batteries are usually oxides such as $LiCoO_2$ and $LiFePO_4$ with an olivine structure and $LiMn_2O_4$ with spinel structure. Interface resistance is a problem that positive electrode materials need to face. When the electrolyte is sulfide, the attraction of the cathode of oxide to Li^+ is much stronger than sulfide electrolyte due to the large chemical potential difference. It will result in a large amount of Li^+ moving to cathode and interface electrolyte will be lack of lithium. If cathode is a mixture conductor, the concentration of Li^+ at the oxide will be diluted by electron conduction, which results in an increased impedance at the interface. The current solution is to add a layer of ionic conductive oxide between the cathode and the electrolyte to efficiently inhibit space charge layer generation, reduce interface impedance, improve overall battery performance and increase power density [12]. Besides, cathode own conductivity, energy specific capacity and other factors are also the key to the battery performance. Ternary cathode materials and new cathode materials such as sulfide are also important research field because of their high specific volume.

Solid electrolyte: Lithium-ion battery(LIB) with high power density has attracted so much attention as a research hotspot. How to enhance the power density of lithium-ion battery has become a pressing issue for resolution. Lithium metal has the highest power density of anode materials. If it can be commercialized, the performance of batteries will be further improved. The large-scale application of graphite electrode, people have made a lot of research on lithium metal as a negative electrode material. However, lithium metal will cause serious security risks due to dendrite growth. Moreover, for liquid-state lithium metal battery, the organic solvent molecules are small, flammable and volatile. Especially in an external extrusion environment, it is easy to leakage and even cause a fire. Unlike liquid electrolytes, solid electrolytes have a better mechanical property such as flexibility and machinability. Moreover, solid electrolyte is safe because it does not contain flammable and volatile carbonic ester solvent. The presence of solid electrolyte reduces the dendrite growth of lithium metal and improves power density and service performance of lithium-ion battery, which causes a fluctuating chemical potential during transmission. Therefore, it will result in low conductivity at room temperature [13]. Moreover, solid-state batteries are facing high impedance at electrode electrolyte interface. Based on the composition of solid electrolytes, electrolytes can be generally split into inorganic, polymeric and composite electrolytes.

Types of solid electrolytes

Inorganic solid electrolytes: Inorganic electrolytes can be split into oxide and sulfide inorganic electrolytes according to their composition. Oxide solid electrolyte can be split into crystal state and glass state (amorphous). Crystalline electrolytes, also known as conductive ceramics, mainly include perovskite type, Na Super Ionic Conductors (NASICON) type, Li Super Ionic Conductors (LISICON) type and garnet type. These types of electrolytes are characterized by high strength, high hardness and high chemical stability. Moreover, it can be stable in the air, which is conducive to mass production of solid-state batteries. Current researches focus on improving the conductivity of room temperature ions and the compatibility of electrodes. In terms of conductivity, it is mainly improved through element replacement and heterovalent element doping. While the compatibility of electrodes is mainly improved through element replacement and adding buffer layer. The main research of glass-state electrolytes is lithium phosphorous oxynitride (LiPON) type electrolyte in thin-film battery. The material has excellent performance, with ionic conductivity up to 2.3×10^{-6} S/cm. Moreover, it has a wide electrochemical window and excellent thermal stability. So, LiPON is the standard electrolyte material for all-solid-state thin film batteries, which has already been applied commercially. Ionic conductivity of LiPON films depends on the amorphous structure of film material and the content of N. The increase of N content can improve the ionic conductivity [14]. Sulfide inorganic electrolyte is similar to oxide inorganic electrolytes. Due to S^{2-} has a larger ionic radius and stronger polarization than O^{2-}, it can reach the goal of increasing lattice volume and expanding size of lithium ion channels. Moreover, S^{2-} weakens attraction and binding between skeleton and Lithium, thus increasing the concentration of carriers and exhibiting greater ionic conductivity. Both crystalline and amorphous electrolytes are faced with an interface contact problem. Inorganic electrolyte is a rigid material, and the contact form between inorganic electrolyte and porous electrode is point-to-point contact. The contact area is limited, which restricts the battery's rate performance and power density. By contrast, polymer electrolytes are soft and viscous which can be used to solve the contact problem.

Polymer solid electrolytes: Polymer electrolytes are mainly divided into gel (semi-solid) polymer and all solid-state polymer electrolyte. Gel polymer is a mixed system in which a large amount of liquid is contained by a small amount of solid-state polymer. Therefore, fluidity of the electrolytes made by gel polymer does not completely loss. The advantage of this material is high conductivity and high migration number, which is approximately equal to liquid electrolyte. Similar to liquid electrolytes, gel electrolytes have better antioxidant properties. Although the material retains the advantages of liquid electrolyte to a great extent, some of the disadvantages of liquid electrolyte cannot be overcome. For example, gel electrolyte can solve the safety problem caused by fluid leakage to some extent, but the flammability of electrolyte cannot be solved. Common gel polymer electrolytes include polyvinylidene fluoride (PVDF), hexafluoropropylene (HFP) polymethyl methacrylate (PMMA). All solid polymer electrolyte is a pure solid electrolyte without liquid components. This kind of electrolytes transport lithium ions through molecular chains of the thermal or electric field motion. Therefore, the solid electrolyte material has low conductivity at room temperature, and it is more suitable for working in a high temperature. Nowadays, a large amount of researches are based on how to solve the power density, conductivity, charging capacity and stability of all-solid-state batteries at low temperature. This kind of electrolytes mainly include polyethylene oxide (PEO), PVDF and chlorinated polyethylene (CPE). PEO has become the most commonly used solid polymer electrolyte among them due to high salt solubility and electrode interface compatibility. At high temperature, the molecular chain of polymer PEO will have a high thermal motion, especially in the amorphous region. The molecules in the PEO compound can bond with lithium ions to build a complex. And the transfer of lithium will base on the thermal motion of the complex. At present, polycarbonate-based, polyester-based and polysiloxane-based polymer electrolytes are also the focus of researches [15].

Composite solid electrolyte: Composite solid electrolyte consists of polymers, lithium salts and inorganic fillers. In this way, composite solid electrolytes combine benefits of inorganic solid electrolytes with polymer electrolytes, like high ionic conductivity, mechanical strength, flexibility, safety, and high electrode interface contact. The inorganic filler can reduce glass transition temperature of the composite electrolyte system, reduce crystallization of polymer and effectively increase ionic conductivity of the system. There are certain groups at the surface of inorganic fillers that can interact with the anions in lithium salt and help to dissociate lithium salt. Moreover, the mixture of polymer and inorganic filler can provide a better transport channel for lithium ions, which is suitable for fast migration of lithium ions [16]. Inorganic fillers can be specifically split in two different parts. The first part is inert materials which do not contain lithium, and lithium ions cannot be transferred inside, such as: Al_2O_3, TiO_2, ZnO, etc. The other one is the active material containing lithium, and lithium ion can be transported within it, such as $Li1.5Al0.5Ge1.5P_3O_{12}$ (LAGP), $Li1.3Al0.3Ti1.7P_3O_{12}$ (LATP), $Li6.4La3Zr1.4Ta0.6O_{12}$ (LLZTO).

Advantages of solid-state battery

Safety: Safety has always been the core issue to promote the development of solid-state batteries. In particularly, lithium-ion batteries are being developed from small batteries like mobile phone battery to large batteries like car batteries, because the size of batteries is increasing and the content of combustible electrolytes is increasing. Security problems are becoming more serious. However, low- carbon and environmentally friendly life makes people have to use lithium-ion batteries instead of traditional fossil energy. Solid electrolyte provides a method to solve the safety problem because it is not flammable. Moreover, solid electrolyte can reduce dendrite growth of lithium ion to a certain degree, which provides a theoretical basis for lithium metal anode. Generally, solid-state battery provides a guarantee for the safety of lithium-ion batteries.

Durability: Durability is another big issue for lithium-ion batteries. Mobile phones always have a decreasing battery capacity as they grow in life, which forces people to replace their phone batteries. For large batteries, the durability of lithium-ion batteries is also unsatisfied. Electrolyte decomposition and electrode side reaction are the main factors affecting battery life. First, lithium-ion batteries store energy by combination of a highly oxidizing cathode and a highly reductive anode to produce a high battery voltage. This will have a tendency to decompose the electrolyte through an electrochemical process. The presence of solid electrolytes can reduce this situation. Besides, solid electrolytes can improve battery life by inhibiting side reactions on electrodes. At room temperature, lithium ions are the only particles which can move in solid electrolyte, so nothing else can diffuse to the electrode surface for oxidation-reduction reactions. Therefore, solid electrolyte can effectively inhibit the side reaction on electrodes and provide high durability for solid-state batteries.

Larger electrochemical window: The small electrochemical window of organic liquid electrolytes restricted the range of choice for electrode materials. Chemical window refers to the potential difference between oxidation and reduction reactions. Both electrodes must be inert to electrolytes. Which means that the oxidation potential needs to be higher than the lithium ion embedding potential in cathode, and the reduction potential must be lower than the lithium metal potential in anode. Generally, solid electrolytes can withstand higher electrochemical windows which are around 4-5 V. Therefore, solid electrolytes are more compatible with more electrode materials. Solid electrolyte can extended the acceptable electrochemical window by adding interphase coating between electrodes and electrolyte [17].

Power density: Power density is a measurement of how fast batteries can provide stored energy. Although solid electrolytes have been considered to be low power density, solid electrolytes have higher potential to increase power density. Firstly, the solid electrolyte can overcome the concentration gradient caused by ion flow in the liquid electrolyte. Because in solid electrolyte, negative charge is generally bound to the framework, which cannot flow easily [18]. Moreover, in liquid electrolytes, lithium ions are solvated, so desolvation is a necessary step before they are inserted into the interlayer galleries of electrodes. But the process of desolvation requires a high activation energy, which limits electrode reaction kinetics. However, for solid electrodes, there is no need to be desolvated, so it has greater potential in terms of power density. With the continuous development of new materials, a large number of solid electrolytes with high ionic conductivity are being discovered. They give solid-state batteries a big breakthrough in power density and make the solid-state battery reach a high power density comparable to lithium-ion batteries.

Challenges of solid-state battery: According to the latest research, the lithium dendrite growth produces a strong growth stress up to 130 MPa [19]. It illustrates that the solid-state electrolytes cannot prevent dendrite growth effectively. Therefore, it is a challenge for increasing the service life of solid-state battery. Besides, the interface resistance is also a challenge for solid-state battery. Point to point contact between solids is the main problem of interface resistance which should be improved. Moreover, the performance of solid-state battery in cold environment is also a serious problem. Because the mechanism of lithium ion conduction is using solid-state molecular chains to complex lithium ions and conduct lithium ions by molecular chain thermal motion. To solve this problem, electrolyte materials need to be improved continuously. After solving all the problem, the final challenge is to make cost-reduction programs. The low cost, high performance and high safety battery is the development trend of battery in the future development

Conclusion

Challenges of Solid-state Lithium-Ion Battery (LIB) is the main method to improve the performance and safety of liquid-state lithium-ion battery. Besides, solid-state electrolyte is core part of all-solid-state lithium-ion battery, which receive extensive concern. The development of solid-state electrolyte with wide electrochemical window, high ionic conductivity, high ionic mobility and small contact resistance between electrode and electrolyte has a great significance for solid-state batteries. In this paper, performance of traditional liquid-state lithium-ion battery is analyzed. Besides, its composition and working principle are investigated. It is found that there exist some problems such as narrow electrochemical window, lithium ion dendrite growth and less safety. Meanwhile, it finds out the solution of replacing the traditional liquid-state battery with solid-state battery, introduces the composition and working principle of solid-state battery, and finds out advantages of inorganic, polymer and composite solid electrolyte respectively. In addition, this article evaluates the overall advantages of solid-state batteries and challenges which they will face. In the future, all-solid-state batteries will break through technology and production bottlenecks. And with the continuous improvement of battery material performance, solid-state batteries will improve further in terms of energy density and safety. Eventually, solid-state batteries will be industrialized and become one of essential energy storage tools for future development.

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