

Solid-State Lithium-Ion Batteries: From Laboratory Research and Development to Industrial Application

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Abstract. With the development of science and technology, more and more "high technology" has been developed. New energy vehicles, mobile devices, and other technologies are booming, which also leads to an increasing social demand for high-performance energy storage equipment. In this paper, the structure of lithium batteries and solid-state batteries and their advantages as energy storage devices are explored. And it is found that lithium batteries have the characteristics of light weight, long service life, and high discharge voltage according to the literature review. Solid state battery has the advantages of high safety and high energy density, but it also has some problems, such as high cost, difficult recovery, and difficult large-scale production. If these problems are addressed, and make the fabrication cost of solid-state batteries reduced, the commercialization of solid-state batteries can solve problems such as insufficient battery life in new energy vehicles, low efficiency in sustainable energy storage, and short service life.

1 Introduction

Energy is the core support of human social activities and the material basis of economic development and civilization progress. Throughout the history of energy utilization, mankind has experienced the era dominated by "biomass energy" and "fossil energy" successively. However, the non-renewable nature of fossil energy and the greenhouse gases such as carbon dioxide (CO₂) produced in the use process have caused serious problems such as global environmental deterioration and intensification of the greenhouse effect. The climate crisis caused by excessive exploitation and consumption is a profound threat to human survival and development. In this context, the development of renewable energy such as solar, wind and tidal energy is imminent. But renewable energy has "regional distribution" and "intermittent supply" of inherent defects. For example, solar energy is limited by the sun's rise and fall in the east, and wind energy is limited by the season. The renewable energy scale application is highly dependent on efficient energy conversion and storage technology. Lithium ion battery as a kind of electric energy and chemical energy can achieve the reversible conversion of energy storage device, with its high energy density, long life and other advantages, become to solve the problems of the renewable energy supply and demand does not match one of the core technology.

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As "Internet Plus" and intelligent technologies improves by leaps and bounds, emerging fields such as large-scale power grid energy storage base stations, multi-functional intelligent devices and electric vehicles have put forward higher requirements for the performance of energy storage devices. Compared with traditional batteries such as lead-acid batteries and nickel-metal hydride batteries, lithium-ion batteries have become the preferred solution in the field of energy storage due to their "lightweight and high efficiency". In recent years, research institutions around the world have made important breakthroughs in solid-state battery technology. For example, Gao Xinyu has developed a composite solid-state electrolyte membrane to enhance the safety of solid-state batteries [1], Pranaya Keshari Nahak found that the polymer interlayer thickness has a great impact on the battery performance [2], Chanho Kim's team pushes the limits of the energy density of silicon sulfide solid-state batteries [3], but the technology from the laboratory to industrial applications, still faces interface stability, cost control, such as large-scale preparation of multiple challenges. Therefore, how to overcome the bottleneck of industrialization at the same time to promote the bottleneck, to promote solid-state lithium-ion battery's livelihood, is assigned to a key issue in society.

This article summarizes a large number of documents, comparing the lithium-ion battery with four common battery discharge voltage, cycle life, and energy density. It is found that lithium-ion batteries have the advantages of high energy density and long service life. Detailed explanations were provided for some solid-state batteries simultaneously, and the challenges faced by the high cost of large-scale production, the negative electrode can not be completely infiltrated into the electrolyte and so on.

2 Overview of sub-batteries

2.1 Working principle of the lithium battery

Currently, the structure of the lithium-ion battery used in daily life mainly consists of positive/negative electrode, electrolyte and diaphragm. Among them, the working principle of lithium cobaltate (LiCoO_2) // graphite lithium ion is the most typical, which relies on the intercalation chemistry theory of layered compounds, as shown in Fig. 1.

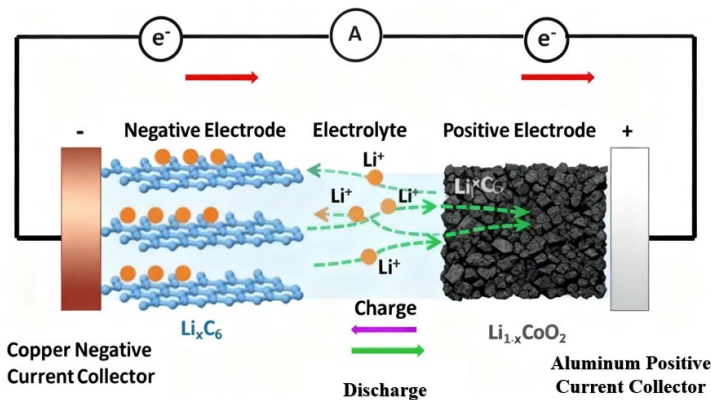


Fig. 1. Schematic diagram of the working principle of lithium-ion batteries.

When lithium cobalt oxide (LiCoO_2) // graphite lithium ion is charged, the Co^{3+} in the positive electrode is oxidized to Co^{4+} to generate $\text{Li}_{1-x}\text{CoO}_2$ and release electrons, which are then transferred to the negative electrode through the external circuit. Meanwhile, the lithium

ion escapes from LiCoO_2 and uses the electrolyte in the battery to penetrate the membrane and reach the graphite surface of the negative electrode. It is then combined with electrons transferred through an external circuit and embedded between the graphite layers to form the Li_xC_6 compound.

In the discharge process, the lithium ions are removed from LiC_6 and release electrons to turn into graphite material again. The removed lithium ions pass through the diaphragm along the electrolyte and return to the positive electrode surface. The released electrons return to the cathode along the external circuit. Both then combine with $\text{Li}_{1-x}\text{CoO}_2$ on the surface of the positive electrode to form LiCoO_2 .

2.2 The characteristics of lithium-ion batteries

In addition to lithium-ion batteries, the common electrochemical energy storage technologies include nickel-hydrogen battery, Li-ion and nickel-cadmium cell. According to the data of lithium battery and other common batteries, such as Table 1, it can be found that lithium battery has the characteristics of high energy density, long life and large working voltage. Lithium-ion batteries have become the most common commercial batteries due to these significant performance advantages [4].

In terms of energy supply, the energy density of lithium-ion batteries can reach up to 300Wh/kg, which makes them have less weight while having the same amount of energy as nickel-metal hydride batteries; At the same time, the discharge voltage of lithium batteries is 2~3 times that of other batteries, which can meet the demand of high power appliances for high voltage.

In terms of use, the charge and discharge cycle life of lithium batteries is far greater than that of other batteries on the market, which greatly saves the cost of battery use. When idle, lithium battery almost will not cause damage to electrical equipment.

Table 1. The four secondary battery performance of the type.

Type of battery	Voltage of discharge (V)	Density of energy (Wh/kg)	Effect of memory	Monthly discharge (%)	Cycle life
Lead-acid battery	2	30-50	none	~10	~300
Nickel-cadmium battery	1.2	50-60	Yes	~20	~1000
Nickel hydrogen battery	1.2	60-80	Yes	20-30	~500
Lithium-ion battery	3.6	150-300	none	<5	>1000

The earliest commercialization of lithium cell was in the 1990s. Since then, lithium-ion battery technology has become increasingly mature and gradually entered People's Daily life, but there are many problems in the use of Li-ion, some of which have not been solved until today.

First, the safety of traditional lithium batteries is poor. On the market at present the circulation of lithium battery is one of the traditional lithium-ion batteries, most of the use of liquid electrolyte, prone to leakage [5]. In the use of Li-ion, more or less mechanical damage will lead to damage to the shell, then the electrolyte inside the battery may leak to the use of people or equipment damage.

Secondly, so far, the energy density of liquid lithium-ion batteries has reached a bottleneck, which cannot meet the current demand for energy density in future development [6].

2.3 Solid-state lithium-ion battery

2.3.1 Overview of solid state lithium-ion batteries

Solid-state battery is mainly composed of three core parts: positive electrode material, Anode materials and solid electrolyte, which is one of the most important part of the solid electrolyte, among which solid electrolyte is one of the most important components, its main role is to block positive and negative electrodes and ensure the normal transfer of ions. With the traditional lithium-ion battery, the biggest difference of solid battery is that the flame retardant solid electrolyte was used instead of the original liquid electrolyte, which makes the solid battery have high safety.

The cathode materials of Li-ion mainly include layered structure, cationic disordered rock salt, olivine structure and spinel structure. The cationic unordered rock salt is expected to be the cheapest of the anode materials.

Disordered Rocksalt Cathodes, (DRXs), General formula for the $\text{Li}_{1+x}(\text{MM}')_{1-x}\text{O}_{2-x}\text{F}_y$, belongs to the cubic dense packing structure, and its unique point lies in the layered structure of molecular materials in which the cations are disorderly arranged rather than traditional layered. This enables more efficient migration of lithium ions in the 3D transition metal skeleton to achieve higher specific capacity. In fact, since the Ceder team of MIT in the United States first announced the DRX cathode material $\text{Li}_{1.211}\text{Mo}_{0.467}\text{Cr}_{0.3}\text{O}_2$ in 2014, the world has set off a boom in DRX cathode material research. Naoaki Yabuuchi et al. found that the performance of lithium batteries using $\text{Li}_{1.3}\text{Nb}_{0.3}\text{Mn}_{0.4}\text{O}_2$ as the positive electrode was significantly better than that using LiVO_2 as the positive electrode, and the specific capacity of first discharge of this material was 300mAh/g at 60°C and 1.5-4.8V. The specific capacity can be stabilized at 230mAh/g after repeated use [7]. As a new type of cathode material, DRX material no longer needs expensive metals such as cobalt and nickel to maintain the structure, which makes the manufacturing cost of DRX much lower than that of traditional cathode materials, and is important for the development of cheap batteries in the future.

Based on the energy storage principle, the cathode materials of lithium cell can be divided into intercalation type, transformation type and alloy type. Carbon-based materials are a very common intercalated new negative materials, such as graphite, soft carbon, hard carbon and so on. Among them, graphite because of its high safety and kinetic properties, make it become the main cathode material of most batteries today. As shown in Figure 1, the volume fluctuation of the battery with graphite as the negative electrode material is less than 10% during charging and discharging, and it also has the characteristics of good circulation, low price and green environmental protection. However, graphite with a theoretical negative capacity of 372mA/g cannot make high-energy density batteries [8].

From the study of solid-state batteries to today, researchers usually divide solid electrolytes into three categories: Composite solid electrolyte [9], Organic polymer solid electrolyte [10] and inorganic ceramic solid electrolytes [11]. At present common sulfide solid electrolyte is a kind of inorganic ceramic electrolyte.

At the end of last century, Kato et al. discovered a sulfide $\text{Li}_{9.54}\text{Si}_{1.74}\text{P}_{1.44}\text{S}_{11.7}\text{Cl}_{0.3}$, which has a three-dimensional Li^+ transport path. But it is prone to react with water in the air to produce toxic hydrogen sulfide gas, which makes its normal transport and production extremely difficult [12].

2.3.2 Advantages of solid state lithium-ion batteries

In the development of lithium ion battery, solid state lithium ion battery has attracted much attention. It mainly contains solid electrolyte, positive and negative electrode material three parts. Solid batteries have many advantages over conventional batteries because of solid electrolyte replaces the original electrolyte.

Solid-state batteries use non-combustible, heat-stable solid electrolytes, reducing the possibility of fire caused by the battery. Secondly, due to the use of solid electrolyte, poor liquidity, should not leak, greatly reduce the safety risks. Meanwhile, the solid electrolyte can restrain the growth and puncture of lithium dendrites. This allows lithium-ion batteries to use high capacity positive/negative electrode materials, making the energy density of the battery possible to improve [13].

In the assembly of the battery, liquid battery because the electrolyte is liquid, there is fluidity, which makes the electrolyte need to be packaged first, and then assembled in series; And solid-state batteries for each component is solid, without considering its liquidity, packaging can be first series again. This greatly reduced the use of packaging materials [14].

2.3.3 Challenges faced by solid state lithium-ion batteries

For the past few year, the solid state batteries due to its HED and safety performance and widely by the researchers studies, solid-state batteries still faces challenges. It is mainly the challenge of the interface between the positive and negative electrodes and the electrolyte. Solid state batteries require a stable electrode and electrolyte interface due to the change of electrode material and electrolyte volume. However, the electrolyte of the common solid-state batteries nowadays cannot fully wet the surface of the anode electrode material, leading to insufficient flatness of the anode-electrolyte interface.

In addition, the commercialization of solid-state batteries also faces difficulties such as high manufacturing cost, difficulty in recycling and inability to scale up production.

Take sulphide solid-state batteries for example. Softpack battery is one of the criteria to judge whether the battery is practical. The liquid electrolyte of the liquid soft pack battery and the electrode material are in liquid-solid contact, which can perfectly penetrate into the active substance of the electrode. And solid soft package battery electrolyte and electrode materials is solid-solid contact, and the electrode active material full contact, seriously affected the transmission rate of Li^+ . Meanwhile, the cost of producing sulfide solid electrolytes is extremely high. At present, the cost of large-scale preparation of raw material Li_2S for the production of sulfide solid electrolyte is \$1000 per kg, while the cost of laboratory production is \$10,000 per kg. Moreover, the production of sulfide solid electrolyte requires high equipment and energy consumption [15].

In order to promote the solid-state lithium-ion battery from laboratory to life, it is necessary to carry out in-depth research mainly around the two directions of "interface optimization" and "cost control".

In view of the problem of wettability in lithium inorganic solid electrolyte and cathode, through "interface coating" or "composite electrolyte" strategy to solve - for example, in the construction of the lithium cathode surface LiF artificial membrane solid electrolyte interface (SEI), or a development "inorganic ceramic particles and polymer composite electrolytes (e.g., LLZO/PEO), The polymer phase is used to perfect the interfacial contact while retaining the high conductivity of the inorganic phase. In order to reduce the cost, it is possible to find low-cost alternative materials (such as Al replacing La and Zr in LLZO), or develop new solid electrolytes without rare metals (such as sulfide $\text{Li}_7\text{P}_3\text{S}_{11}$). Process level: Optimize the large-scale preparation technology of solid-state batteries (such as the use of flow method to prepare ultra-thin solid electrolyte film and laser welding to achieve interface compaction) to

reduce production energy consumption and cost -- the goal is to reduce the cost of solid electrolyte from the current \$500 /kg to less than \$100 /kg, so that the cost of CRIEPI is the same as that of liquid batteries. At the same time, a whole-life cycle management system can be established for solid-state batteries, and combined physical-chemical recovery technology can be developed (such as low-temperature roasting to separate solid electrolytes and electrode materials), so as to realize the recycling of metal resources such as lithium, cobalt and nickel, and promote the green and sustainable development of solid-state battery industry.

3 Conclusion

Driven by the new energy revolution and the goal of "dual carbon", the demand for high security and high energy density energy storage devices in new energy automobile, large-scale energy storage and other fields is increasingly urgent. As a new generation of energy storage technology, solid-state lithium-ion battery replaces liquid electrolyte by solid electrolyte, solves the safety hazard of traditional batteries from the root, and has the advantages of HED and simplified assembly process, which is the core solution to meet the future energy storage demand. In this paper, the development of Li-ion and solid batteries is summarized, and the advantages and challenges of solid-state battery are elaborated. Now the challenge for the solid-state batteries is mainly "interface optimization" and "cost control" two direction. As for the development direction of solid state battery, there are mainly the following aspects.

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