



Progress and prospect on failure mechanisms of solid-state lithium batteries

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HIGHLIGHTS

- Summarize fading behaviors of solid state batteries during working and storing.
- Review fading mechanisms of solid state batteries from the aspect of interfaces.
- Discuss methodologies in studying fading mechanisms of solid state batteries.
- Put forward prospects for high performance and high safety solid state batteries.

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ABSTRACT

By replacing traditional liquid organic electrolyte with solid-state electrolyte, the solid-state lithium batteries powerfully come back to the energy storage field due to their eminent safety and energy density. In recent years, a variety of solid-state lithium batteries based on excellent solid-state electrolytes are developed. However, the performance degradation of solid-state lithium batteries during cycling and storing is still a serious challenge for practical application. Therefore, this review summarizes the research progress of solid-state lithium batteries from the perspectives of failure phenomena and failure mechanisms. Additionally, the development of methodologies on studying the failure mechanisms of solid-state lithium batteries is also reviewed. Moreover, some perspectives on the remaining questions for understanding the failure behaviors and achieving long cycle life, high safety and high energy density solid-state lithium batteries are presented. This review will help researchers to recognize the status of solid-state lithium batteries objectively and attract much more research interest in conquering the failure issues of solid-state lithium batteries.

1. Introduction

Lithium-ion battery (LIB) is an important sustainable technology for the future energy storage and transportation. In 1991, the firstly commercialized LIBs consisting of LiCoO₂ cathode, carbon anode, and organic liquid electrolyte renovated the portable electronics [1]. After 27 years' unremitting efforts in scientific research and technical innovation, thinner, lighter, smarter and longer standby time electronics has been progressively realized by gradually raising the energy density of LIBs. Moreover, due to the advantages of high energy density, high operating voltage, long cycle life, low self-discharge rate, no memory effect and eco-friendly, the application fields of LIBs have been expanded from commercial electronics to electric vehicles (EVs) and grid-energy storage systems [2–6]. Especially, the development of EVs has been determined as the national strategy to cope with the energy crisis, environment pollution and climate change. However, the endurance mileage, life and cost of recent EVs cannot compare with the

performance of internal combustion vehicles [7,8]. This is because the state-of-the-art LIBs are in shortage of energy density, power density, durability, and cost [9,10]. More importantly, the burning of EVs powered by LIBs has brought safety concerns.

It is worth noting that the limited energy density and potential safety risks of LIBs are strongly associated with the organic liquid electrolyte, which cannot match with high energy density cathode materials and tends to volatilize, flame and explode [11]. Replacing the organic liquid electrolyte by non-flammable, safe and highly reliable solid-state electrolyte is expected to enhance the energy density, cycling life and safety of LIBs [12,13]. Solid-state electrolytes used in batteries mainly contain lithium-ion-conductive solid polymers, ceramics, and organic-inorganic composites. The high energy density lithium metal anode is also revived by solid-state electrolytes, which have high mechanical strength to suppress the lithium dendrites growth [14,15]. Therefore, solid-state lithium-ion battery (SSLIB) or solid-state lithium metal battery (SSLMB) with prominent energy density and safety are

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viewed as main power battery candidates for EVs in the near future [8,16,17].

Nevertheless, the large-scale commercialization of high energy density and safety solid-state lithium batteries (SSBs) is still challenged by several issues. Firstly, power density is quite important for quick charging of SSBs [18]. In recent decades, plenty of efforts and progress have also been made to improve the kinetic performance of SSBs, such as exploring novel electrolyte materials with high ionic conductivity at room temperature, studying interfacial ion-transport mechanisms and modifying the interface properties [19–25]. Secondly, long term stability is essential for application of SSBs. The cycle and calendar life (lifetime of batteries in terms of number of discharge/charge cycles and time after production) of batteries should be superior to the life of EVs. For example, both the China's new technology roadmap and United States Advanced Battery Consortium (USABC) have made a battery life goal of 10 years by 2020, which means 1000 cycles life under 80% depth of discharge [9,10]. Though SSBs are expected to have long cycle life and excellent calendar life because of the limited side reactions between electrodes and solid electrolytes, the laboratory prototype SSBs with such long cycle life have been rarely reported. Therefore, the capacity fading is the main issue limiting the commercialization of SSBs. Thirdly, short circuit caused by Li dendrites is a potential safety hazard in SSLMB [26–35]. Safety seems to be a less critical issue in SSBs than conventional LIBs, but its harm is still fatal. Thus, the safety failure of SSBs also hinders the widespread application of SSBs, especially SSLMBs.

Recently, although the scientific issues and development of SSBs have been reviewed previously, the comprehensive overview of failure analysis on SSBs has been presented seldom. Most reviews about SSBs mainly focus on the solid-state electrolytes with high ionic conductivity, electrochemical stability and mechanical properties, the compatibility between solid-state electrolyte and electrodes, the structure of the composite electrodes and the interface modification strategies [36–40]. For example, Kim et al. have comprehensively reviewed the important advance of electrolyte materials and battery design in SSBs and concluded that one of the major reasons for unsatisfactory output power density and cycle life of SSBs was the fast degradation of lithium-contained cathode materials resulting from the microstructural change or dendrite formation [41]. Kerman et al. have highlighted the kinetic limitations of the solid-state interface and elucidated the technological challenges in processing solid-state cells [42]. Though there are many works about fading analysis of LIBs, the differences in physical state, structure, and composition between liquid and solid electrolytes make the research experience in conventional LIBs cannot be fully and straightforwardly applied in SSBs [43–52]. However, the detailed fading analysis of SSBs has not been summarized systematically and comprehensively. Recently, Kerman et al. have summarized two of the key high level failure modes including impedance growth and electrical shorting related to interfaces in SSBs [42]. But, the fading mechanisms and corresponding research methods for SSBs has not been reviewed yet. Recognizing the fading behavior and understanding the fading mechanism with the advanced research methods will help researchers develop SSBs with excellent comprehensive performances. Therefore, we present the first review on the fading analysis of SSBs from the aspects of calendar life, cycle life, internal resistance augmentation, short circuit and thermal runaway. In this work, the fading behaviors of SSBs are firstly introduced. Then, the fading mechanisms and analysis methods are summarized. Finally, we outlook the studies on SSBs and give some suggestions to future research.

2. Failure behaviors of solid-state lithium batteries

Failure behaviors determine the reliability, safety and life of cells, and therefore directly influence their application in energy storage devices. Correctly detecting and identifying the failure behaviors of SSBs will help researchers to solve the failure problems. After

investigating recent research works about SSBs, the failure behaviors are classified as the calendar failure, cycle failure, internal resistance augmentation, short circuit, and thermal runaway.

2.1. Calendar failure

Calendar failure is the capacity loss during battery storage [43]. Battery calendar failure is usually studied by monitoring the self discharge rate, capacity fade and resistance augmentation under the variables of storage time, temperature, and state of charge (SOC). In LIBs, high temperature tends to facilitate the secondary reactions and lithium loss, and high SOC promotes the precedent chemical reactions by exacerbating the potential disequilibrium on the electrode/electrolyte interface. Thus, high temperature and high SOC usually aggravate the calendar failure. Furthermore, the calendar failure is not linear with time, suggesting the strong interaction of failure behavior with time [43]. However, there are only several reports about the calendar failure of SSBs. These works investigated the resistance augmentation after storage tens or hundreds of hours by measuring the electrochemical impedance spectroscopy (EIS) of Li/solid-state electrolyte (including $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$, $\text{Li}_{10}\text{SiP}_2\text{S}_{12}$, PEO–PMMA–LiTFSI, PEO–PMMA–LiTFSI– Al_2O_3 , $\text{Li}_7\text{P}_3\text{S}_{11}$, glass ceramic–PEG–silane–LiTFSI)/Li cells at 20 °C or room temperature [53–56]. The resistance increase of symmetric lithium-lithium cells during storage varies with different electrolytes. After 24 h storage, the higher passivation layer resistance and charge transfer resistance for the cell with $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ grow nearly six and five times as much as that of cell with $\text{Li}_{10}\text{SiP}_2\text{S}_{12}$, respectively [53]. Furthermore, both cells show nearly linear increase of resistance during storage, indicating the steady-state uniaxial growth in the electrolyte decomposition layer formed at the electrolyte/Li interface. In sharp contrast, the interfacial resistance increases initially with the storage time and then attends to be stable in PEO–PMMA–LiTFSI or $\text{Li}_7\text{P}_3\text{S}_{11}$ based symmetric cells. The former is attributed to the interface passivation layer and the continuous morphology adjustment of polymer electrolyte membrane over time, while the latter is ascribed to the completely finished interphase formation reaction [54,55].

Nevertheless, the capacity fade of SSBs during storage has not been reported. And, the calendar failure at different temperatures and SOC has also not been investigated. More importantly, there is no work about the calendar failure of full cells containing cathode and non-Li metal anode. As SSB is a quite potential energy storage technology for EVs and grid-energy storage systems, calendar life is one of the key parameters to assess the durability of SSBs. To find out the reason for resistance increase during storage and solve the calendar failure problems will promote the application of SSBs. Therefore, much attention has to be paid on the calendar failure of SSBs under different temperatures and SOC. Technically, the calendar failure of SSBs has to be measured under the work environment or reasonable acceleration conditions.

2.2. Cycle failure

Cycle failure of SSBs happens during charge or discharge. It usually manifests as the capacity loss during cycling. Capacity retention rate (the N cycle discharge capacity/the first cycle discharge capacity $\times 100\%$) and coulombic efficiency η ($\eta = \text{discharge capacity}/\text{charge capacity} \times 100\%$) during cycling are two important parameters to evaluate the cycle failure. Cycle failure crucially depends on the charge-discharge voltage, current density, utilization and work temperature of SSBs. As summarized in Table 1, increasing voltage and current density tends to accelerate capacity fading, while high temperature helps capacity retention during long term cycling. However, except the LiFePO_4 -based batteries, most SSBs have unsatisfactory cycle life (less than 300 cycles). Strictly, the cycle failure of these SSBs might be much severe under the laboratory conditions.

The electrode materials in SSBs usually offer a much lower initial

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