

Research Review on Safety Performance Analysis of Electrolyte and Separator Materials for Lithium-ion Batteries

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Abstract

In recent years, lithium-ion batteries have been highly favoured by the electric vehicle and new energy sectors due to their comprehensive advantages, such as long operating range and high cycle life. However, as research into increasing battery energy density has led to heightened risks and potential hazards, reports of lithium-ion battery explosions have become increasingly frequent in the news, making safety issues increasingly prominent. Dendrite crystal growth, which damages the SEI film, along with the thermal shrinkage and poor heat resistance of traditional separators, are key causes of thermal runaway. Modified composite separators and the development of novel solid-state electrolytes are regarded as effective approaches to balancing performance with commercial market demands. However, there remains a gap in systematic summaries of existing research focusing on safety rather than the increase in the battery's own capacitance. This paper provides a systematic review of research progress on the safety of various composite separators and electrolytes, analyses the composition design strategies for different thermal runaway prevention systems and their mechanisms of influence on battery performance, and highlights that striking a balance between regulating and synergistic thermal runaway prevention, suppressing dendrite growth, and further enhancing ionic conductivity and large-scale production represents the core challenge of current research. Furthermore, it is proposed that multifunctional integrated design and large-scale fabrication compatible with existing processes represent current key development directions and potential objectives for high-safety lithium-ion batteries.

Keywords

Lithium-ion Batteries; Composite Separators; Dendrite Growth; Thermal Runaway Resistance; Safety; Solid-state Electrolytes.

1. Introduction

Modern civilisation is inextricably linked to electricity. Devices that directly convert the energy generated by chemical reactions into electrical energy are known as chemical power sources, or batteries. Compared with other power sources, batteries are highly favoured for their high energy conversion efficiency, convenience, safety, ease of miniaturisation and environmental friendliness. Lithium-ion batteries are a new type of high-energy battery successfully developed in the 1990s; they are a form of lithium-ion intercalation battery that evolved from lithium secondary batteries. The continuous development and utilisation of new materials, coupled with the demand from the new energy vehicle sector for high capacity, quality and production capacity, have undoubtedly served as the cornerstone for the rapid development of lithium-ion batteries over the past two decades.

Currently, whilst lithium-ion batteries strive for high energy density, safety hazards such as thermal runaway and internal short-circuits are becoming increasingly prominent. Optimising and modifying the performance of electrolyte and separator materials has thus become a key breakthrough for enhancing battery safety and overall performance. [1]The core of thermal runaway lies in the chain reaction of exothermic side reactions and heat accumulation within the battery: when the local temperature rises, the decomposition of the SEI film and reactions between the electrolyte and electrodes occur in sequence, with the heat generation rate far exceeding the heat dissipation capacity. [2]Furthermore, existing separators are prone to thermal shrinkage or melting at high temperatures, failing to effectively isolate the positive and negative electrodes, which leads to an internal short circuit that instantly releases a massive amount of heat, triggering a fire or explosion. This problem is particularly acute in the case of dendrites: lithium dendrites grow uncontrollably during charging and discharging, readily piercing traditional separators and causing micro-short circuits. The Joule heat generated during the instant of a short circuit further exacerbates the non-uniform deposition of lithium, creating a vicious cycle of ‘dendrites – internal short circuits – localised overheating – more dendrites – dead lithium’. [3]

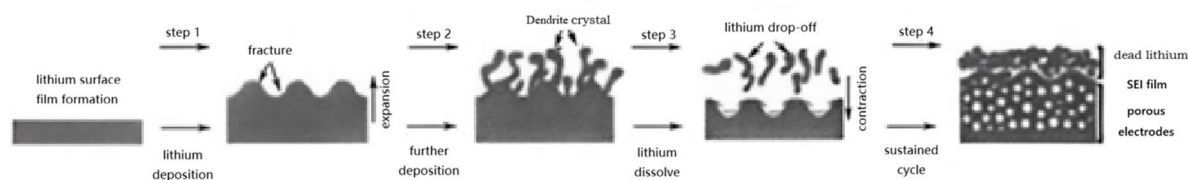


Figure 1. formation mechanism of lithium dendrites

2. Current State of Research

Early research both domestically and internationally focused on surface grafting or coating of commercial PP/PE separators, leading the way in molecular structure design, multifunctional integration and adaptation to a wide temperature range. However, most studies remained at the laboratory stage, with insufficient consideration given to large-scale manufacturing processes and economic viability. Overall, efforts should begin with cutting-edge gel polymer electrolytes, focusing on scaling up production capacity from the laboratory stage to mass production in order to establish an industrial supply chain.

3. Research Methods and Objectives

This review examines several research approaches centred on improving lithium-ion batteries by addressing the hazards posed by dendrite formation and thermal runaway, and discusses how gel polymer electrolytes (GPEs) can fundamentally resolve the issue of dendrite formation. [4]This review focuses on enhancing the safety and wide-temperature-range adaptability of lithium metal batteries through the optimisation of conventional electrolytes, the modification of separators, and the design of polymer electrolytes that integrate both approaches. It systematically summarises the key breakthroughs achieved by the aforementioned separator materials in suppressing lithium dendrites, realizing thermal shutdown functions, and achieving high ionic conductivity.

4. Main Research Approaches for Enhancing Safety

4.1 Conventional Methods - Suppressing Thermal Runaway and Dendrite Formation by Adjusting Electrolyte Composition or Separator Materials

4.1.1 Fluorinated carbonate solvents

School of Safety Science and Engineering, Fluorinated ethylene carbonate (FEC), di-(2,2,2-trifluoroethyl) carbonate (TFEC) and methyl trifluoroethyl carbonate (FEMC) were selected as fluorinated solvents; electrolytes were prepared by combining these with 1 mol/L LiPF₆ lithium salt. which demonstrated a significant improvement in safety performance under various

environmental conditions, particularly in batteries using an electrolyte containing FEC and TFEC in a mass ratio of 2:3; these exhibited the highest capacity retention rate after 100 cycles, reaching 92.69%; This type of electrolyte features low interfacial resistance and high conductivity, which facilitates the insertion and extraction of Li^+ at the electrode–electrolyte interface. Furthermore, it promotes the formation of a stable, smooth and relatively thin cathode electrolyte interface (CEI) layer within the battery, thereby enhancing interfacial stability, suppressing the formation of dendrites at the interface. Furthermore, all the aforementioned electrolytes are non-flammable and demonstrate superior safety performance under electrical, mechanical and thermal abuse conditions: in the needle-puncture test, the battery voltage exhibited a rapid rise followed by stabilisation and a subsequent decline, with no instances of thermal runaway occurring. This demonstrates that the impact of the needle-puncture condition—the most common trigger for thermal runaway in everyday life—on these batteries is minimal, thereby enhancing their safety under mechanical abuse conditions. With regard to the thermal runaway trigger temperature, fluorinated carbonate batteries exhibit lower thermal runaway severity and lower trigger temperatures. This may be because their stable CEI layer reinforces the electrode interface, preventing the release of oxides caused by phase transitions in the cathode, thereby reducing the severity and likelihood of thermal runaway. This approach yields superior results compared to traditional improvement methods involving the addition of materials, whilst the reinforcement of the CEI layer may also help to mitigate the issue of lithium-ion dendrite formation. [5–6]

4.1.2 Polyimide-based Functionalised Porous Membranes

In response to the two shortcomings of existing commercial polyolefin separators—insufficient thermal stability and poor interfacial contact with electrode materials, which result in suboptimal performance of the electrode materials—and given that polyimide (a polymer material with imide rings in its main chain, i.e. PI) possesses high thermal stability, excellent mechanical properties and chemical stability, as well as the high mass density of existing commercial flat copper foil current collectors, Guangdong University of Technology Through structural design and process optimisation, lightweight Cu/PI composite current collectors with a porous structure and PI separator materials containing carboxyl functional groups were prepared using an electrospinning process. This method demonstrates significant advantages in separator preparation. It is characterised by its simplicity of operation and broad applicability, and is suitable for large-scale production, sufficiently meeting the current high-volume market demand [7]. Furthermore, owing to the good thermal stability of PI and the high-temperature meltability of PVDF-HFP, the separator exhibits high thermal stability and a thermal runaway temperature of nearly 150 °C [8]. Overall, this method enhances the thermal stability of the separator by improving its stability.

4.1.3 Polyethylene Composite Separators

Current research into separators is primarily focused on composite modification techniques based on traditional, inexpensive and widely used polyethylene (PE) separators, incorporating ceramic oxide nanomaterials with high melting points and high electrolyte affinity, such as silicon dioxide (SiO_2) and aluminium oxide (Al_2O_3). [9–10] This has led to the development of three new types of separator materials: inorganic particle-coated composite separators, inorganic particle-grafted composite separators and inorganic particle-filled composite separators. Compared with traditional PE separators, composite PE separators exhibit significant improvements in thermal stability, electrolyte wettability and ionic conductivity. These enhancements, alongside improved stability and slightly increased mechanical strength, serve to enhance the safety of lithium-ion batteries and reduce the likelihood of thermal runaway.

(1) Inorganic nanoparticle-coated polyethylene separators are the most widely used type of polyethylene separator. Unlike other types of separators, they offer advantages in terms of physical properties, cost and chemical stability. However, their surface is non-polar, resulting in poor wettability; they have a low melting point (130–138 °C) and are prone to shrinkage at high temperatures, meaning they exhibit poor thermal runaway behaviour. Currently, the development of

composite separators primarily involves introducing multifunctional, heat-resistant inorganic materials directly onto the surface of the PE separator; applying a surface coating to the PE separator using a composite ceramic solution or inorganic ceramic materials; or coating the separator with a composite gel polymer. Ceramic materials can enhance overall strength through their inherently excellent mechanical strength and heat resistance. Consequently, polyethylene composite separators, which use polyethylene separators as the substrate, possess the aforementioned advantages and hold a competitive edge in large-scale application and production, offering broad prospects for application; whilst their coating layer enhances the separator's wettability and electrolyte absorption capacity, it may increase the resistance to lithium-ion diffusion. Mun et al. ([11]) primarily employed chemically modified SiO_2 to impregnate PE separators; these exhibit thermal stability at high temperatures of 120 °C and a long cycle life, but the challenge lies in maintaining the separator's thickness and pore structure. Lü et al. ([12]) developed a sponge-like nanoporous silica-coated separator; compared with the previous approach, this nanoporous SiO_2 further improved the thermal stability of the PE separator and enhanced its wetting properties with the electrolyte. This reduces the likelihood of entering a vicious cycle of temperature rise–thermal runaway–temperature rise–thermal runaway under high-temperature conditions.

The properties and advantages of Al_2O_3 are similar to those of SiO_2 ; both play an important role in ceramic materials. Cheng et al. ([13]) Al_2O_3 and polyvinylpyrrolidone (PVP) ceramic slurry were applied as a single-sided coating to the surface of a PE separator to prepare an Al_2O_3 /PVP-PE composite separator. This separator demonstrated excellent heat resistance and dimensional stability even at high temperatures approaching 180 °C. With regard to the preparation of complex networks, Ahn et al. [14] prepared reactive Al_2O_3 particles using a water-soluble binder; after coating the PE separator with these particles, they were thermally cross-linked with a gel precursor to form a three-dimensional network, thereby further reducing the likelihood of thermal runaway and dendrite formation.

(2) Ceramic-coated PE separators with grafted inorganic nanoparticles are effective at suppressing internal short circuits in batteries through physical barrier action, thereby reducing the losses generated by the battery itself during cycling. However, an increase in coating thickness and the blockage of the porous structure can lead to a significant decrease in the energy density and power density of lithium-ion batteries; issues such as uneven distribution of ceramic particles and weak bonding strength of the polymer binder are not advantageous for large-scale commercial production. In contrast, surface grafting modification techniques-carried out under appropriate irradiation conditions-enhance adhesion and heat resistance whilst minimising changes to the polymer matrix composition. These techniques include physical irradiation methods such as electron beam grafting, plasma treatment and γ -ray irradiation. The free radicals generated during irradiation initiate polymer chain cross-linking reactions, thereby improving the physicochemical properties of the separator. For example, Jiang et al. [15] treated a PE separator with a high-energy electron beam (EB) to generate free radicals that initiated a grafting reaction with vinyltris(2-methoxyethoxy)silane (VTMEOS). Whilst there were no significant changes to its porous structure or thickness, the separator exhibited improved electrolyte wettability, ionic conductivity and thermal stability. Zhu et al. ([16]) first grafted vinyltrimethoxysilane (VTMS) onto the surface of the PE separator, and then subjected it to a co-hydrolysis condensation reaction with sodium silicate under acidic conditions.

(3) Compared with traditional inorganic particle coating and grafting methods, the direct filling of PE separators with inorganic nanoparticles not only eliminates cumbersome steps but also improves the separator's thermal shrinkage rate and puncture strength. These enhancements in shear and tensile strength offer advantages in laboratory-scale production testing and refinement, thereby reducing experimental costs, as demonstrated by Zhang Yumei et al. [17] Blends of heat-resistant ceramic boehmite and UHMWPE-filled separators can be applied to batteries with more flexible structural configurations, enabling further improvements upon current designs.

4.1.4 Cellulose-based Separators

Cellulose, as the most abundant natural polymer on Earth, exhibits enormous application potential due to its unique physicochemical properties [20]. As a linear polymer composed of D-dehydropyran-type glucose rings linked linearly via β -1,4-glycosidic bonds, forms a linear polymer compound. Hydrogen bonding between hydroxyl groups—both intermolecularly and intramolecularly—creates highly ordered crystalline regions, endowing cellulose separators with excellent high-temperature dimensional stability; they can maintain structural integrity even at 200°C, effectively preventing the risk of thermal runaway in batteries under extreme conditions. In the field of lithium-ion batteries, research into cellulose-based separators is now well-established, having progressed from simple papermaking-based preparation methods, through the precise control of microscopic pore structures, to the introduction of functional materials to achieve multifunctionality. In current research, nanocellulose has effectively addressed the issue of insufficient mechanical strength in cellulose derivatives. Nanocellulose is primarily classified into three categories: cellulose nanofibres (CNF), cellulose nanocrystals (CNC) and bacterial cellulose (BC). In the study by Gonçalves et al., CNF enhances mechanical strength through a dense network of hydrogen bonds acting as a substrate, whilst CNC primarily relies on high stiffness to improve performance and therefore typically exhibits extreme brittleness, as confirmed by this research. From a safety perspective alone, the physical entanglement and abundant hydrogen bonds within BC make it resistant to penetration by crystal branches, resulting in good tensile and puncture resistance; however, this also makes it difficult to disperse uniformly in water and to form a uniform, porous structure, thereby limiting the electrochemical performance of BC membranes.

Among the numerous modification strategies, TEMPO pretreatment is widely employed. The carboxylate anions introduced at the C6 position effectively weaken the hydrogen bonding forces between cellulose fibres through electrostatic repulsion, thereby significantly improving the membrane's porosity and, consequently, its tensile strength.

Although cellulose separators, as heat-resistant separator materials, do not possess a particular advantage in preventing thermal runaway at high temperatures, their excellent tensile, compressive and puncture resistance enable them to retain good physicochemical properties even under the aforementioned harsh conditions. As their large-scale production is gradually being incorporated into planning, they are set to capture a certain market share in today's commercial sector.

4.2 Using a Solid-state Electrolyte as the Separator to Replace the Liquid Electrolyte and Mitigate the Risk of Thermal Runaway Caused by Puncture

4.2.1 Solid-State Electrolytes

Solid-state electrolytes (SSEs) offer significant advantages in terms of safety due to their excellent thermal stability, which helps suppress phenomena such as lithium dendrite formation; however, in inorganic solid-state electrolyte systems, the quality of interfacial contact remains a key bottleneck affecting overall battery performance. Consequently, a study [18] prepared a series of structurally tunable HNBR samples using heterogeneous catalytic hydrogenation technology. Building on this, the study focused on the HN41-36W-97% polymer—characterised by high nitrile content, high molecular weight and high degree of hydrogenation—and further incorporated the high-ionic-conductivity lithium-lanthanum-zirconium-tantalum oxide (LLZTO) filler and polyethylene oxide (PEO) plasticiser. The resulting composite electrolyte exhibited optimal performance, demonstrating excellent room-temperature ionic conductivity and a high lithium-ion mobility. Consequently, this solid-state electrolyte material possesses outstanding dendrite suppression capability, interfacial stability and cycling stability, and offers a technical pathway for the development of high-performance and high-safety solid-state lithium batteries, presenting promising prospects.

4.2.2 Gel Polymer Electrolytes

Gel polymer electrolytes (GPEs), as a new type of electrolyte, possess higher ionic conductivity at room temperature and a larger contact area with the anode and cathode materials. They can mitigate

the thermal runaway issues associated with organic electrolytes and further address problems inherent to solid-state electrolytes, such as poor interfacial contact. In terms of volume, solid-state polymer electrolytes occupy less space between the electrodes and account for a smaller proportion of the battery's mass than liquid electrolytes. Consequently, they offer higher energy density and are suitable for use in new energy vehicle batteries. In the event of extensive damage to the vehicle, their smaller volume and surface area reduce the likelihood of damage to the battery itself, as well as the risks and pollution associated with electrolyte leakage. To meet the growing demand for energy storage devices with long cycle life and high energy density, lithium-ion batteries based on a novel gel polymer electrolyte system—combining ultra-long cycle life, high energy density and high safety—can significantly enhance the application value of lithium-ion batteries. [19]

(1) 1,3-Dioxolane-based gel polymer electrolytes

The School of Chemical Engineering at Hefei University of Technology has focused on in situ polymerisation strategies to develop polymer electrolytes with long-term cycling stability, designing poly-DOL-PETEA-based gel polymer electrolytes (DP-GPEs) and poly-DOL-VC-PETEA-based double-network gel polymer electrolytes (DVP-GPEs). In laboratory experiments, the prepared gel polymer electrolytes demonstrated significant advantages in terms of ion transport kinetics, cycling stability and feasibility for industrial application, providing a viable strategy for the industrialisation of highly safe, long-life solid-state lithium-metal batteries. Using aluminium trifluoromethanesulphonate as an initiator, 1,3-dioxolane (DOL) and pentaerythritol tetraacrylate (PETEA) via an in situ copolymerisation at room temperature using aluminium trifluoromethanesulphonate as an initiator. The resulting poly-DOL-PETEA-based gel polymer electrolytes (DP-GPEs), which possess a three-dimensional cross-linked network, also serve to suppress the growth of lithium dendrites. This significantly enhances the discharge specific capacity of lithium ions following insertion–extraction cycles, demonstrating excellent cycling stability at various charge–discharge rates.

Furthermore, to accommodate ternary cathode materials with high energy density, a cross-linked network was constructed by introducing vinyl carbonate (VC) III and PETEA onto the DP-GPEs framework; DVP-GPEs were prepared through the synergistic action of DOL cationic ring-opening polymerisation and VC/PETEA radical thermal polymerisation. The lithium-ion transport pathways were optimised and the degree of cross-linking in the polymer network was increased; the resulting DVP-GPEs combine high ionic conductivity ($0.676 \text{ mS} \cdot \text{cm}^{-1}$), a high lithium-ion mobility (0.825) and a low activation energy ($E_a = 0.116 \text{ eV}$), whilst maintaining excellent cycling stability alongside high mechanical strength (tensile strain $> 60 \text{ MPa}$).

It can thus be concluded that these improvements to gel-type electrolytes, which address the current issues of thermal runaway and dendrite formation in chemical batteries, provide crucial direction and guidance for future small-scale research and production. They also refine the theoretical foundation of previous research on solid-state electrolytes, and their advantages in terms of cycling performance and stability offer significant economic development prospects.

(2) Gel electrolytes incorporating tetrahydrofuran groups

Also in the field of gel electrolytes, the Guangxi Key Laboratory of Non-ferrous Metals and Special Materials Processing, and the School of Resources, Environment and Materials at Guangxi University, have employed methods to effectively immobilise anions in order to increase the lithium ion (Li^+) migration number and stabilise the lithium metal anode in GPEs [20]. By incorporating tetrahydrofuran (THF) groups into acrylate monomers, ion–solvent coordination was regulated at the molecular level whilst preserving the fluidity of the film-forming additive, fluorinated ethylene carbonate (FEC), thereby maintaining efficient Li^+ transport and uniform interfacial chemistry. Furthermore, owing to the establishment of a balanced ion transport network and a stable electrode–electrolyte interface, the battery retains a high capacity retention rate even after prolonged cycling, effectively mitigating the formation of dendrites during lithium-ion transport.

5. Conclusion

This paper focuses on the core issue of high safety performance in lithium-ion battery electrolytes and separators, systematically reviewing research and progress towards commercial-scale application in this field. It provides an overview of existing traditional approaches-such as adjusting electrolyte composition or separator materials-as well as cutting-edge research methods that utilise solid-state electrolytes as separators to replace liquid electrolytes. These include composite separators, polyethylene separators coated with inorganic nanoparticles, and inorganic nanoparticle-grafted polyethylene separators, ceramic-coated polyethylene separators and polyethylene separators directly filled with inorganic nanoparticles-as strategies to address thermal runaway and suppress dendrite formation; as well as the application and refinement of solid-state electrolytes to prevent dendrite formation and the use of gel polymer electrolytes to regulate heat resistance. It is evident that both traditional modification methods and cutting-edge ‘hard-soft hybrid’ approaches, which hold great promise, are sparing no effort to improve the cycling performance of batteries; however, they all still suffer, to a greater or lesser extent, from common issues that render them unsuitable for large-scale commercial production. Based on the above analysis, incorporating the mass production of gel polymer and highly conductive ternary copolymer separators into research plans has become a key direction in the development of high-safety lithium-ion batteries.

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