# A Review of Lithium-Ion Battery Capacity Decline, Safety Concerns, and Potential Mitigation Strategies

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**Abstract.** The transportation sector accounts for around 25% of global energy-related carbon emissions, and their impact is expected to grow in the coming years. As a means of mitigating carbon footprint, motor industries have explored vehicle designs using renewable, zero-emission energy sources, such as lithium-ion batteries. Although widely employed, the current iteration of Lithium-ion batteries (LIB) suffers from high-capacity loss over time and safety risks. Capacity loss is attributed to active lithium-ion loss and electrolyte depletion due to forming a solid-electrolyte interphase (SEI) on the anode and internal resistance growth due to electrolyte corrosion. On the other hand, safety concerns center around the potential for thermal runaway during battery operation. The sources of these issues will be discussed in this review, along with mitigation strategies that may optimize both factors simultaneously. Possible strategies include exploring alternative anode materials, including Silicon and Tin, optimizing separator parameters, utilizing prelithiation methods, and introducing cooling systems into LIB design.

**Keywords:** Capacity; Safety; Solid Electrolyte Interphase (SEI); Thermal Runaway.

## 1. Introduction

The transportation sector accounts for around 25% of global energy-related carbon emissions, and their impact is expected to grow in the coming years [1]. As a means of mitigating carbon footprint, motor industries have explored vehicle designs using renewable, zero-emission energy sources, such as lithium-ion batteries. The prominent problems in Lithium-Ion Batteries (LIB) include capacity and safety. The battery's capacity is limited due to anode plating, electrolyte depletion, and internal resistance growth. As the battery ages, these factors significantly decrease the coulombic efficiency (CE), the charge efficiency by which electrons are transferred in batteries. The coulombic efficiency is interrelated to the capacity of the battery. Higher coulombic efficiency means fewer electrons are trapped during the charging/discharging process. Therefore, there is less capacity loss during each cycle [2]. Although not standardized, the current industry requires LIBs that fall below 70% initial capacity to be replaced, since the battery performance declines dramatically beyond this threshold [1]. Given that the average cost of battery replacement is approximately $6000**,** LIB capacity loss could impose high and continuous costs if not addressed [1]. Aside from capacity, another problematic factor is safety. The intrinsic design of the battery can cause thermal runaway and toxic gas leakage, which could further escalate into fire, explosions, and intoxication, endangering the well-being of people [3]. Many studies have been conducted to address capacity and safety issues. However, most existing studies address one of the two fields without considering the other. This approach exempts the possible trade-offs between LIB capacity and safety, which must be considered to make the design encompassing and sustainable. This paper discusses innovations overlapping capacity and safety factors in LIB design. Possible strategies, including pre-lithiation of the electrodes, adjusting separator thickness, and improving cooling systems, typically overlap between the boundaries [3], and their respective impacts need to be measured to optimize the two prominent concerns in LIB.

## 2. Lithium-Ion Battery Structure and Working Principle

As shown in Figure 1, positive lithium ions separate from the cathode, diffuse through the electrolyte, traverse past the separator nanopores, and integrate into the anode during LIB charging [4]. While the battery is charging or discharging, the electrons move in the opposite direction through the external circuit to maintain electro-neutrality between the electrodes. In other words, electrons migrate from the anode to the cathode during charging and migrate from the cathode to the anode during discharging. The electron movement between the anode and cathode generates electricity for common LIB appliances, including mobile devices and transportation [5]. For practical use, hundreds to thousands of LIB cells are typically combined, in parallel or series, into one battery pack to obtain a specific voltage and capacity [6].

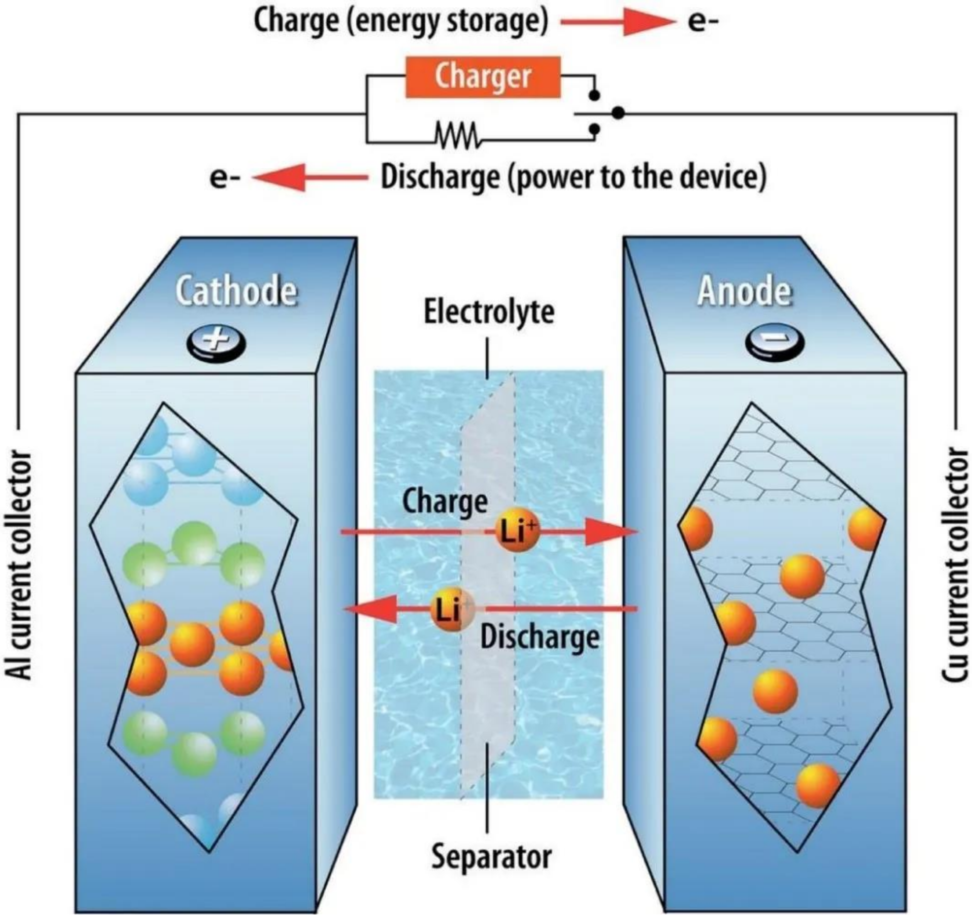

**Fig. 1** LIB Schematic [7]

## 3. Current Issues of LIB

Essential issues in LIB include capacity and safety concerns. As the battery ages, capacity loss due to Solid-Electrolyte Interface (SEI) formation and increased internal resistance become more pronounced, drastically decreasing the battery's performance. In addition, uncontrollable heat generation in LIBs could cause thermal runaway, leading to fire and safety risks.

### 3.1. Capacity

LIB Capacity highly depends on the amount of active lithium ions in the electrodes. More active lithium ions enable more Redox reactions, which in turn increase the amount of electron charge and discharge, thereby contributing to higher battery capacity. Lithium-ion loss due to Solid-Electrolyte Interface (SEI) formation and internal resistance growth are two of the most common drivers of LIB capacity reduction over time.

### 3.1.1 Active Lithium Depletion at the SEI

In lieu of intercalation, SEI Formation, often referred to as lithium plating, happens when lithium ions produce solid lithium on the anode's surface. Because the number of lithium ions moved during the electrochemical reaction defines the energy density of LIB, plating-induced loss of active lithium materials results in partial use of active materials, which reduces net capacity in subsequent battery operation [2]. As the lithium capacity of the anode becomes limiting due to aging, it will eventually be unable to accommodate all lithium during charging, leading to rate-independent lithium plating on the anode [8].

During the charging and discharging process, various factors cause lithium loss, such as SEI formation, volumetric Change, and lithium loss at defect sites. SEI (Solid Electrolyte Interphase) formation, another expression for lithium anode plating, occurs because the electrolytes are thermodynamically unstable on negatively polarized anode surfaces. As a result, interfacial reactions between the anode and electrolyte and electrolyte decomposition occur irreversibly, leading to SEI formation that consumes active Lithium ions. Volumetric Change is another process that causes active Lithium ion loss in LIB. Lithium ions may migrate deeper into the anode or toward the anode surface during the initial charge/discharge cycle. During this lithiation stage, the anode material could expand. Silicon-based anodes, for instance, are highly prone to volume expansion during lithiation and could grow to approximately 280% [9]. As the anode expands, the lithium ions that diffuse more profoundly into the anode become trapped, thereby decreasing the active Lithium ions available [10]. Defect sites are an additional factor causing lithium depletion; in the case of silicon anodes for example, there are a lot of flaws at the interfaces and grain boundaries [11]. Strong atomic bonds with lithium ions are likely to develop at these defect sites, which would lower the amount of lithium ions in the LIB [10].

### 3.1.2 Electrolyte Depletion at the SEI

Higher working temperatures cause more noticeable side reactions inside the LIB, which speeds up the drying of the electrolyte [12]. Furthermore, the gas's solubility is diminished. As a result, additional gas molecules are trapped within the cell, which may hinder ion flow and reduce CE and battery capacity [13]. Lee et al. [14] measured the correlation between electrolyte depletion and discharge capacity loss. At 50% electrolyte depletion, the battery discharge capacity decreases by 5% at a discharge rate of 0.05 C. When 50% electrolyte depletion is coupled with 30% lithium loss, the discharge capacity reduces by 46%. Lai et al. [15] punctured fresh cylindrical 18,650 cells and adjusted the electrolyte volume. They found significant capacity reduction in the battery cell when the electrolyte level dropped below 70%, and the battery cell stopped operating below 40% electrolyte level.

### 3.1.3 Internal Resistance Growth

Internal resistance growth is another factor that decreases LIB capacity. As LIB ages, the internal resistance, quantifying the amount of opposition to the current flow in and out of a battery, rises [16]. Higher internal resistance creates more overpotential, which reduces battery capacity. The resistance growth rate multiplied by the applied current determines the magnitude of the overpotential growth [8]. The growth rate in internal resistance is attributed to external temperatures and side reactions. In their research, Chen et al. [3] found that when external temperature decreases, the internal resistance of LIB increases, thereby contributing to lower capacity. Thus, a higher operating temperature could improve the overall capacity of LIB. In another case study, side reaction products within LIB were studied and found to also correlate with internal resistance growth. Since oxide-based electrode materials function well above the electrolyte's potential range, internal resistance growth was more noticeable for these materials.

## 3.2. Safety

The primary safety hazard associated with LIB is the risk of thermal runaway. Without a way to mitigate heat generation, the battery could overheat, causing fires and explosions and endangering well-being.

### 3.2.1 Thermal Runaway

Thermal runaway is classified as a set of exothermic chain reactions that result in an uncontrollable increase in LIB temperatures. They are detrimental to safety and well-being, as thermal runaway could escalate into destruction of the battery, fires, and explosions. When the rate of heat generation surpasses the rate of dissipation, thermal runaway takes place. Heat will build up and raise the temperature, leading to severe exothermic side reactions that produce more heat. [17]. This continuous cycle of heat generation and chain reactions is the principle behind thermal runaway. Degradation and regrowth of the solid electrolyte interphase are caused by the accumulation of heat emitted during battery operation or external heat when the temperature is below 120 degrees Celsius. At 130 to 150 degrees Celsius, the electrode separator will begin to shrink, which could cause an internal short-circuit between the anode and cathode. When the short circuit takes place, the cell temperature will rise sharply up to 200 degrees Celsius, triggering the next stage of thermal runaway. Oxygen compounds will start to be released by the cathode, causing a serious redox reaction of the batteries' flammable components. The redox process continuously elevates the cell temperature until failure or fire [17]. Thermal runaway is typically attributed to three causes: local thermal abuse, electrical abuse, and mechanical abuse. Local thermal abuse happens when the heat generated during normal LIB operations cannot dissipate quickly [3]. Electrical abuse, including overcharging or over-discharging, occurs during electrolyte decomposition - which is caused by battery aging. Rapid heat buildup within the battery could result from electrolyte breakdown at the cathode interface caused by an external short circuit or high state of charge. When external factors cause localized damage to the LIB, mechanical abuse occurs. Due to the high energy density of LIB, physical collisions could release a significant amount of heat within the battery, propagating thermal runaway. In extreme circumstances, the separator could also be torn or pierced, allowing small amounts of air to penetrate the cell and initiating vigorous lithium-oxygen exothermic reactions that release more heat [3].

## 4. Trade-Offs Optimizatiton Strategies

LIB capacity loss and safety risks impose limitations on battery utility. While the two issues differ, present-day improvement strategies cannot improve one factor without worsening the other. In other words, trade-offs are often unavoidable when introducing innovations in LIB. In this review, trade-off factors, including anode material selection, separator thickness, pre-lithiation, and cooling systems, will be discussed, and potential innovations within their field that may optimize capacity and safety will be introduced.

### 4.1. Improvement of Anode Materials

The anode is the negative electrode, where oxidation reactions occur in the LIB. The electrons migrate away from the anode during charging and towards the anode during discharging. Anode material selection must be optimized to improve safety and capacity, as the selected material will directly affect the battery's CE and performance. Carbon, for example, is commercially used due to its high conductivity and abundance [10]. However, its downside is that it lacks specific and rate capacity and introduces safety risks, such as dendrite formation [10]. Innovations that increase electrode capacity will raise the battery's overall performance and effectiveness in terms of longevity and current density. Aside from capacity, safety factors must also be considered to optimize the overall battery function. Silicon, for instance, is an exceptional anode material that provides high specific capacity and low safety risk [10]. Its significant volume variation, which happens during the discharge and charging process, is a disadvantage, though. According to Li et al. [18], a significant volume growth of almost 310% was obtained by inserting the richest phase of the Li-Si anode ($Li_{22}Si_5$) at 415 degrees Celsius. High volume fluctuation can cause a variety of issues, such as particle breaking and erratic and unstable electrical contact [18]. To address volume expansion, researchers have attempted to modify the nanostructure of Si by making the material porous. For instance, introducing pores into the Si-based materials allows them to adjust the volume expansion during the lithiation process. preventing the pulverization and particle cracking process [19]. The increased specific surface area of porous Si anode materials also facilitates the increased transportation of Lithium ions, improving the coulombic efficiency of Si-based anodes. Aside from Silicon, Tin is also a promising anode material with excellent specific capacity and safety [2]. The drawbacks of tin-based anodes, however, are that mechanical stress resulting from significant volume variations during the material mixing processes causes their capacity to deteriorate dramatically as they age. Yamauchi et al. [20] created a GSPO (Glassy SnO-$P_2O_5$) approach to address capacity deterioration, which efficiently controls volume variations during cycling. The solution changes into a nanocomposite comprised of metallic particles in an unstructured lithium phosphate substrate during the initial charge. The capacity loss is gradually reduced by the new electrochemical method, which now includes consecutive reversible lithiation of tin anodes. Although further research and innovations on Silicon and Tin anodes are required before wide-scale application, the anode materials offer significant upside in capacity and safety.

## 4.2. Improvement of Separator

The separator is sandwiched between the positive and negative electrodes to prevent physical contact while permitting ionic conduction through the electrolyte. The separator's thickness and porosity profoundly impact capacity and safety. Thinner separators enable reduced ionic resistance and greater diffusion abilities, allowing for higher LIB capacity. However, thinner separators are more susceptible to degradation. They could be easily pierced by lithium dendrite growth and exhibit higher shrinkage rates at elevated temperatures [21]. In other words, they are more prone to safety risks. Thicker separators have the exact opposite traits as thin separators. They show greater mechanical strength but poor ionic transport kinetics [22]. A trade-off exists between safety and capacity when choosing separator thickness. Thicker separators are safer but exhibit poor capacity, while thinner separators have higher capacity but are more susceptible to safety risks. The optimal thickness of separators in academic research is usually restricted between 20 to 25 micrometres [23]. Another element influencing battery safety and capacity is the porosity of the separator. The percentage of holes in a material that aids in moving lithium ions between the electrodes is known as porosity. The capacity of the battery is increased via more porous structures, which allow for more ionic transport. Higher porosity, however, also increases the separator's vulnerability to short circuits because of the development of lithium dendrites [24]. As a result, there is a trade-off between separator porosity and safety when it comes to thickness. To maximize safety and capacity, a porosity of about 40% is advised for commercial LIBs [24]. Recently, ceramic coating has been studied and tested as a potential additive to separators that may improve safety while retaining capacity. In their research, Zhang et al. [25] co-coated the separators with boehmite ceramics and LATP ($Li_{1.5}Al_{0.5}Ti_{1.5}(PO_4)_3$) solid-state electrolytes and studied the battery's performance. In order to create a thicker SEI-filled LATP layer that reduces separator shrinkage at high temperatures, the LATP combines with the electrolytes and lithium during lithiation. The battery maintained a capacity of 87.99% after 500 cycles, but its thermal runaway breakdown criterion rose from 146.2 to 162 degrees Celsius in the thermal abuse testing [25]. While ceramic coating is a promising strategy for improving safety, its main drawback is a slight decrease in ionic conductivity and capacity [25] . However, considering that only boehmite ceramics were examined, additional testing with other ceramic materials and coating methods could return more optimized results for safety and capacity retention.

## 4.3. Pre-lithiation Modification

In order to compensate for and restore lithium loss during cycling, pre-lithiation is the technique of pre-impregnating additional lithium ions into the anode [26]. Triggering chemical pre-lithiation requires adjusting reagent molecules' electrochemical potential to below the anodes' potential [2]. Existing pre-lithiation approaches include electrochemical and direct contact methods [26]. The electrochemical method applies a galvanostatic current to the electrolytic cell, initiating chemical reactions that transfer lithium ions from a metal source to the working electrode. In research labs, the technique is frequently employed. However, because of metallic lithium's combustibility, it raises safety issues such as thermal runaway [26]. Another pre-lithiation strategy is the direct contact method. When the lithium metal and anode interact, they form an electric field that drives electrons from the lithium to the anode. The main downside of the direct contact method is that lithiation will likely be inhomogenous because their area of contact is not uniform [26]. Therefore, there will still be a degree of active lithium loss in each cycle, which decreases capacity over time. While traditional pre-lithiation methods cannot encapsulate both capacity and safety, alternative pre-lithiation methods have been trialled and show potential in optimizing the two factors. Jang et al., for instance, developed a LAC (Lithium-arene complex) to enhance the electrochemical reversibility of Silicone-based anodes. Since the LAC has a lower redox potential than Silicon, submerging the silicon-based anode within the LAC enables lithium ion transfer to the anode [2]. The process pre-forms an SEI on the anode, which increases the initial CE as there will be reduced parasitic reactions consuming active lithium ions due to the presence of the pre-formed SEI. The LAC prelithiation method shows promise in optimizing safety and capacity. The formation temperature is typically under 50 degrees Celsius, well below the thermal runaway range, and the method enables homogenous lithiation, minimizing capacity loss over time. The main drawback is that the technique requires a relatively long immersion time to complete the prelithiation process. Jang et al. found that five minutes of immersion increases the CE to 91%, and it takes thirty minutes of immersion to increase the CE pass to 100% [2]. Therefore, more research and innovation in the area are required before a LAC lithiation method could become commercially viable.

### 4.4. Improvement of Cooling System

Due to the high-speed cycling of batteries, their heat content increases rapidly, which could induce safety risks such as thermal runaway . It is also important to note that low temperatures negatively influence the battery's capacity. Under 0 degrees Celsius, dendrite growth becomes more pronounced, therefore, consuming more active lithium ions and dramatically decreasing the capacity over time [27]. Feng et al. [28] also estimated that every 5% increase in temperature difference causes a 2% decrease in battery capacity. Therefore, the best strategy for ensuring optimal safety and capacity is to keep the battery at a homogenous temperature between a specific interval, namely 15 to 35 degrees Celsius, according to Wu et al. [27]. A cooling system would help maintain the temperature between this temperature range, improving the overall safety and capacity of the battery. Air cooling is most inexpensive and straightforward. Its drawbacks, however, are in its poor cooling rate and inability to maintain a consistent temperature distribution [29]. Aside from air cooling, direct contact and indirect contact cooling are alternative approaches. Direct contact cooling entails immersing the battery in a coolant and transferring the battery heat to its external environment [27]. Unlike air cooling, the direct contact approach enables homogeneous temperature distribution. The main drawbacks of the method are the possibility of short-circuiting despite high thermal performance and potential leakage issues [30]. Indirect contact cooling, on the other hand, uses heat transfer components, such as tubes and plates filled with cold fluids, spatially arranged around the battery, to transfer heat onto its external environment without directly contacting the fluids [27]. The indirect contact method has weaker heat transfer performance compared to direct contact method. However, it is much easier to incorporate into LIB and requires less power for fluid pumping [30]. ICLC also enables homogeneous temperature distribution throughout the battery, thereby retaining strong battery performance. Mist cooling is another interesting method in addition to commercial cooling systems. Following heat absorption from the battery, the water particles will entirely evaporate at high operating temperatures. As a result, it eliminates the possibility of oxidation from surface wetness or short circuits. The approach also offers a large cooling capacity, effectively controlling battery temperature. The shortcoming of mist cooling is that it is only effective in hot environments. Under colder conditions, mist particles may be retained on the battery, which could induce corrosion and potential circuit failure. A possible area of research, thus, could be to trial non-reactive elements with lower boiling points as potential mist sources. Optimizing safety and capacity in LIB requires the battery temperature to be maintained between 15 to 35 degrees Celsius. Commercially available approaches such as DLCL and ICLC and alternative mist cooling systems have a high potential for achieving this objective.

## 5. Conclusion

Capacity degradation was mainly attributed to SEI formation and increased internal resistance over time. These processes cause active lithium loss in the battery, which disrupts the overall capacity. Like capacity, thermal runaway is another common issue compromising safety, as uncontrolled heat generation within the battery may cause fire, explosions, and toxic gas leakage. Looking deeper into these issues, trade-off factors, including anode material, separator thickness, pre-lithiation, and cooling systems, were discussed extensively, along with potential innovations in each field. Silicon and Tin were explored as potential anode materials that could optimize safety and capacity. However, the high volume expansion must be mitigated before wide commercial use. The separator thickness was a trade-off between safety and capacity, and ceramic coating application shows promise in optimizing this subject. Different pre-lithiation strategies were discussed along with their strengths and drawbacks, and a new Lithium-arene complex approach that may optimize both safety and capacity was introduced. Lastly, the trade-offs between safety and capacity at different operating temperatures were considered, and potential cooling systems, including indirect cooling, direct cooling, and mist cooling, were studied. While the current review focuses primarily on performance, future work into the sustainability and environmental impact of LIB should also be integrated into battery design, along with capacity and safety, to optimize the full life cycle of the battery.

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