

Review On Calendar Aging Mechanisms And Estimations Of Lithium-Ion Batteries In Electric Vehicles

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ABSTRACT

Lithium-ion batteries (LIBs) have changed the storage of energy in a major way, and so they are now considered an extremely important technology in electronic powered devices such as EVs (EVs), consumer electronics, and renewable energy storage systems. They have, though, under different conditions of use, had slower capacity in general due to both the increased and decreased reactions and the mechanical units that were consequently eroded in the course of their use and storage. These are cyclic aging and calendar aging. Cycle aging occurs when batteries are repeatedly charged and discharged. And calendar aging means a gradual loss of performance even when the battery is not actively being used. Both types of aging are affected by several factors such as temperature, charge level, and usage pattern.

This article focuses on how these processes affect performance and lifetime by reviewing the main mechanisms that contribute to battery aging. We also explore different models used to estimate the state of health (SOH) of batteries and estimate how long they will last. By examining empirical, semi-empirical and physics-based models, we aim to provide a clear understanding of the factors that lead to lithium-ion battery degradation and the methodologies available to accurately predict battery performance over time.

Keywords:

Lithium-ion batteries, aging mechanisms, calendar aging, capacity fade, degradation models, state of charge.

Introduction

Lithium-ion batteries have been increasingly popular in today's top of the line technology due to their large energy density, extended cycle lifespan and also high efficiency, turning them into an important type of electric storage solution intended for applications such as electrical vehicles, mobile electronic products together with utility grid-scale stored power facility. Compared to older battery technologies such as lead-acid or nickel-metal hydride (NiMH), lithium-ion batteries have a

significantly higher duty cycle. Features including faster charging, high energy output, and small size and weight have put them at the forefront of industries that require high-performance energy storage solutions [1].

Nevertheless, lithium-ion batteries have several drawbacks, and one of the items which will definitely occur is degradation over time. This degradation is visualized as the decrease in the total capacity of the battery (the amount of charge it can store) and power output, which influences how effective the energy is provided

during discharge [2]. The aging process in lithium-ion batteries is the result of a mixture of chemical, mechanical, and thermal mechanisms. As time flies, some bad chemical actions can take place in between the battery's electrodes and electrolyte, which then are transformed into by-products that abate the battery's capacity to store and deliver charge [3]. Mechanical stress that arises due to the repetition of charging and discharging can induce the electrodes to be fatigued, while high operating temperatures can accelerate the rate of their chemical degradation [4].

One of the most crucial things for EVs is the battery aging. As the battery's capacity fades, the vehicle's driving range lessens and the vehicle becomes dependent on recharges more frequently. This increased cyclability, especially when combined with fast charging practices, also speeds up the degradation process. This becomes a vicious cycle where the battery's performance diminishes over time, eventually leading to decreasing efficiency in the vehicle and possibly even the high cost of battery replacement [5]. As a result of the fact that the battery is the most expensive part of EVs, insights about degradation and their respect are important to the sustainability and economic viability of the EV market [6].

In grid storage systems, where lithium-ion batteries are used to store energy generated from renewable sources like wind and solar, battery aging can severely impact system performance. Over time, degraded batteries lose the capacity to store excess energy produced during peak generation times (e.g., sunny or windy periods). This reduced storage capacity can hinder the grid's ability to supply energy during high-demand periods, potentially increasing reliance on non-renewable backup power sources and undermining the overall efficiency of renewable energy systems [7].

To address these challenges, developing accurate models that can predict battery aging and estimate the state of health is essential. These models help forecast the remaining useful life (RUL) of batteries and inform strategies to extend their operational lifespan. By understanding the factors that contribute to battery degradation—such as charge/discharge

rates, temperature fluctuations, and depth of discharge (DoD)—researchers can optimize usage patterns and propose more effective thermal and charge management strategies [8].

To this end, this paper studies the main factors influencing the aging process of lithium-ion batteries, and also considers new methods for assessing battery degradation and predicting their condition.

Aging Mechanisms in Lithium-ion Batteries

Aging mechanisms in lithium-ion batteries - describe the growth process of fading performance over successive time periods due to physical and chemical changes taking place within the battery. Lithium-ion batteries undergo several internal changes that cumulatively affect their storage capacity, energy efficiency, and power output during repeated cycles of charge and discharge. The aging processes are complex and include numerous interdependent factors suitable for classification as cyclic and calendar aging. In this review, we delve into the detailed mechanisms and estimation techniques associated with the calendar aging of lithium-ion batteries, providing insights into their long-term performance and degradation patterns.

Calendar Aging

Calendar aging describes the degradation of lithium-ion batteries with time, typically during storage or idle conditions when the battery is not in use. Calendar aging differentiates itself from cycle aging—the aging caused by charge or discharge cycles. Calendar aging is essentially caused by environmental factors such as temperature and voltage, though in general, such aging may be negligible when comparing it to cycle aging. Continuing on its course, the battery would nevertheless undertake certain chemical processes, which, slowly but surely, erode its capability to optimally perform. Key factors contributing to calendar aging:

Voltage (State of Charge) is significant factor in calendar aging continues to be the SOC during storage. High-voltages or near-full charges speed up the aging of lithium-ion batteries. Optimal SoC levels for minimizing aging are typically between 40% and 60%.

Storing a battery fully charged (near 100% SoC) accelerates the aging process due to higher internal stress, which increases the rate of side reactions that contribute mass transfer between electrolyte and electrodes. Undoubtedly, this also presents a perilous state option at lower SoC. The reason attached is electrolyte decomposition at low SoC values, which in some cases leads to dendrite formation and thereby in turn lead to short circuits. The degradation rate is also influenced by the average voltage of the cell, as shown below:

$$\text{Degradation Rate} \propto V_{\max} \tag{1}$$

Where V_{\max} is the maximum cell voltage. Typical values for lithium-ion cells range between 3.6 V and 4.2 V, with higher voltages leading to faster aging.

In the field of derating to extend life performance, one of the metrics that might be defined. A derating factor is a ratio of the difference between the degradation rate at the derated stress and the degradation rate at the reference or maximum design stress to the degradation rate at the reference or maximum design stress at time t .

In efforts to prolong battery life, the concept of derating is often employed. Derating

involves operating a battery under conditions that are less severe than its maximum design specifications to reduce the rate of aging. The derating factor (DF) quantifies this reduction in stress, and can be calculated using Equation (2) [9].

$$DF(t) = [DR_{\text{Der}}(t) - DR_{\text{Ref}}(t)] / DR_{\text{Ref}}(t) \tag{2}$$

where DF is the derating factor, $DR(t)$ is the degradation rate at time t , $DR_{\text{Der}}(t)$ and $DR_{\text{Ref}}(t)$ are the degradation rates under the derated stress and reference stress, respectively. $DR(t)$ is defined as

$$DR(t) = [Q_{\text{Loss}}(t) - Q_{\text{Loss}}(t_0)] / (t - t_0) \tag{3}$$

where $Q_{\text{Loss}}(t)$ is the battery capacity loss at time t and $t_0 < t$. If $t_0 = 0$, then the capacity loss at the beginning is 0, $Q_{\text{Loss}}(t_0) = 0$. Then the derating factor can be updated as

$$DF(t) = [Q_{\text{Loss,Der}}(t) - Q_{\text{Loss,Ref}}(t)] / Q_{\text{Loss,Ref}}(t) \tag{4}$$

Consequently, the purpose of the derating factor in introducing the speed of battery capacity loss has to be put into practice as follows according to Equation (4), the main concern of the degradation rate is the rate of capacity loss. A low rate of capacity loss implies a long battery life. We will examine these using Lithium Iron Phosphate (LFP) and Lithium Cobalt Oxide (LCO) Li-ion battery cells.

Chemistry	Nominal Capacity (Ah)	Charge Cut-Off Voltage (V)	Charge Cut-Off Voltage (V)
Graphite-LFP	3.0	3.6	2.0
Graphite-LCO	1.5	4.2	2.75

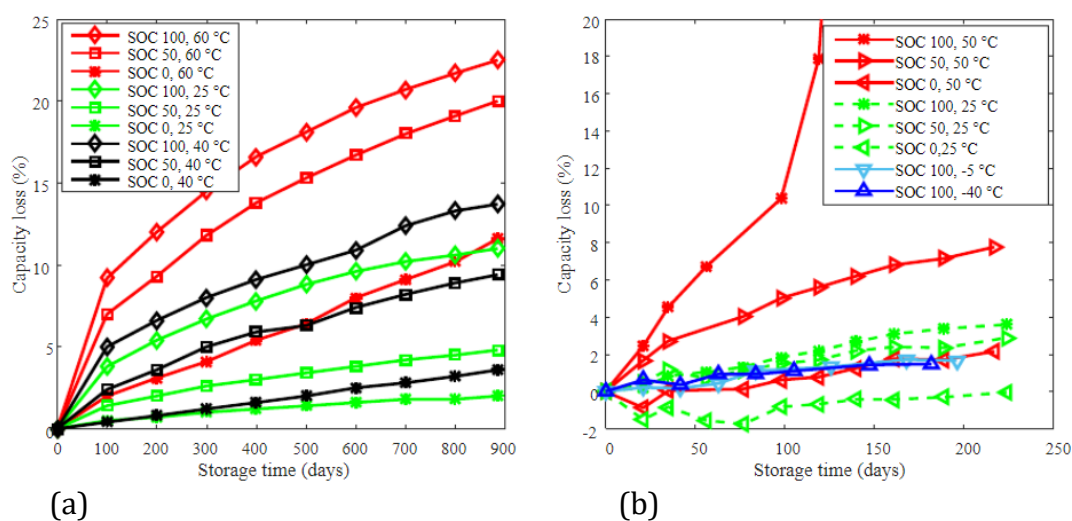


Figure 1. Relevance of SOC to battery capacity.

Figure 1. Calendar life the data of the test. (a) LiFePO₄ (LFP) batteries were tested under three temperatures (60°C, 40°C, and 25°C) and three SOCs (SOCs) (100%, 50%, and 0%) with an experiment duration of over 885 days. “SOC 100, 60°C” refers to LFP batteries were stored at SOC 100% and 60°C. The detailed testing information can be referred to [10] (b) “SOC 100, 50°C” refers to LCO batteries were stored at SOC 100% and 50°C in a temperature chamber. All batteries underwent capacity testing and impedance measurement every 3 weeks. Capacity and impedance characterization: Batteries were constant charged at a rate of C/2 to 4.2 V, then constant voltage charged until the current fell below C/100 rate. The batteries were discharged at a rate of C/2 to 2.75 V to measure the deliverable maximum capacity. Then, the batteries were fully charged using the same constant current constant voltage (CCCV) profile followed an impedance measurement [9].

Temperature: One of the most critical factors influencing calendar aging is temperature. High temperatures accelerate unwanted side reactions within the battery, leading to the decomposition of the electrolyte and the growth of the Solid Electrolyte Interphase (SEI) layer on the anode. These processes consume active lithium ions, reducing the available capacity of the battery. For every 10°C increase in temperature, the rate of degradation typically doubles, following an Arrhenius relationship. Therefore, batteries stored at higher temperatures experience more rapid capacity loss. The Arrhenius equation can be used to describe how temperature influences the degradation rate:

$$k(T) = k_0 \cdot e^{-\frac{E_a}{RT}} \tag{5}$$

where $k(T)$ is the rate of degradation, k_0 is the pre-exponential factor, E_a is the activation energy (in Joules), R is the gas constant ($8.314 \text{ J/mol}\cdot\text{K}$), T is the absolute temperature (in Kelvin) [2].

When it comes to lithium-ion batteries, temperature increases of 10°C usually duplicate the degradation rate. As a result, storing a battery at 40°C instead of 20°C can accelerate the loss of capacity. This effect is especially noticeable at high temperatures, where electrolyte decomposition and other parasitic reactions take place more rapidly.

Figure 2a shows the changes in the capacity of LFP cells and LCO cells at different temperatures after 885 and 140 days, respectively. The reference temperatures for LFP cells and LCO cells are 60°C and 50°C, respectively, then the curves of derating of temperature are plotted in Figure 2b.

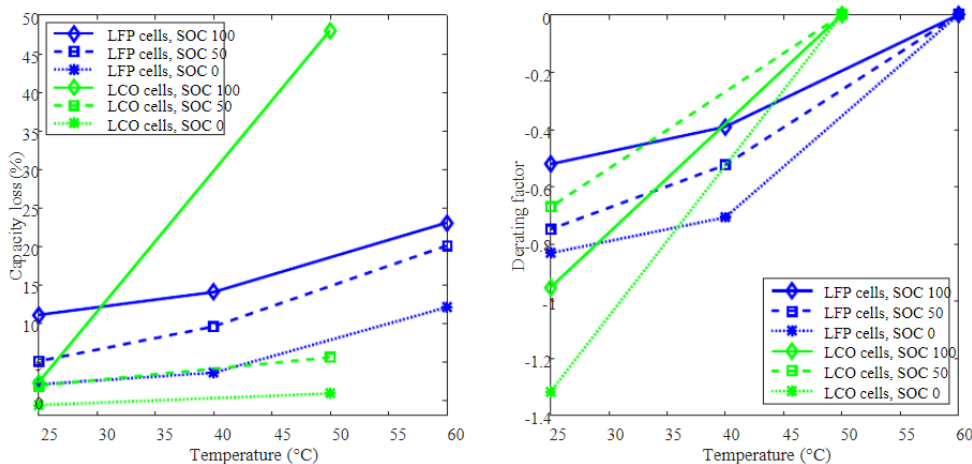


Figure 2. Relevance of temperature to battery capacity.

Derating of temperature for calendar life. (a) Capacity loss vs temperature (b) Derating factor vs. temperature. The derating factors of temperature were calculated using Equation (4).

In the following figure demonstrated Calendar aging behavior of NCA/Gr-SiO_x21700 cells.

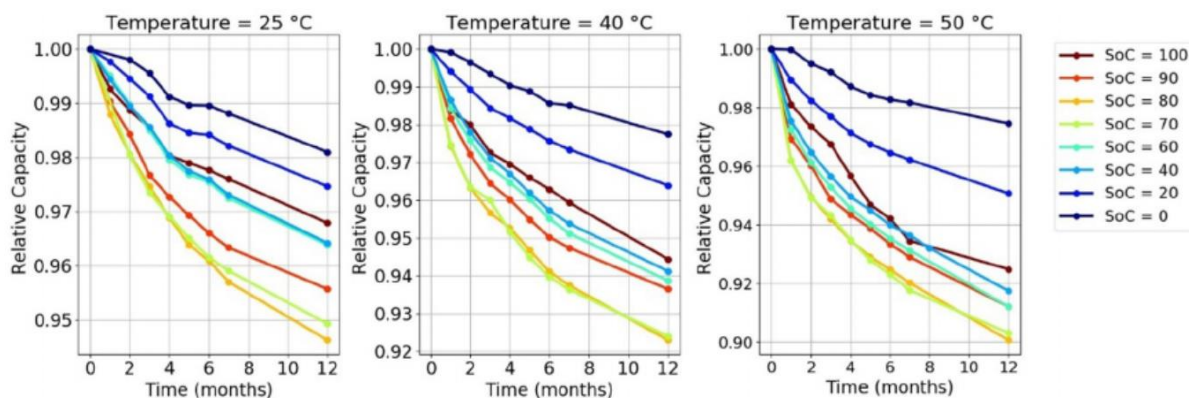


Figure 3. Calendar aging behaviour of NCA cells.

Relative capacity as a function of time for all SoCs tested, at three different temperatures. The greatest capacity-fade is observed when cells are at 70 and 80 % SoC, at all temperatures. Capacity values are $I=1.5$ A and $V<2.5$ V.

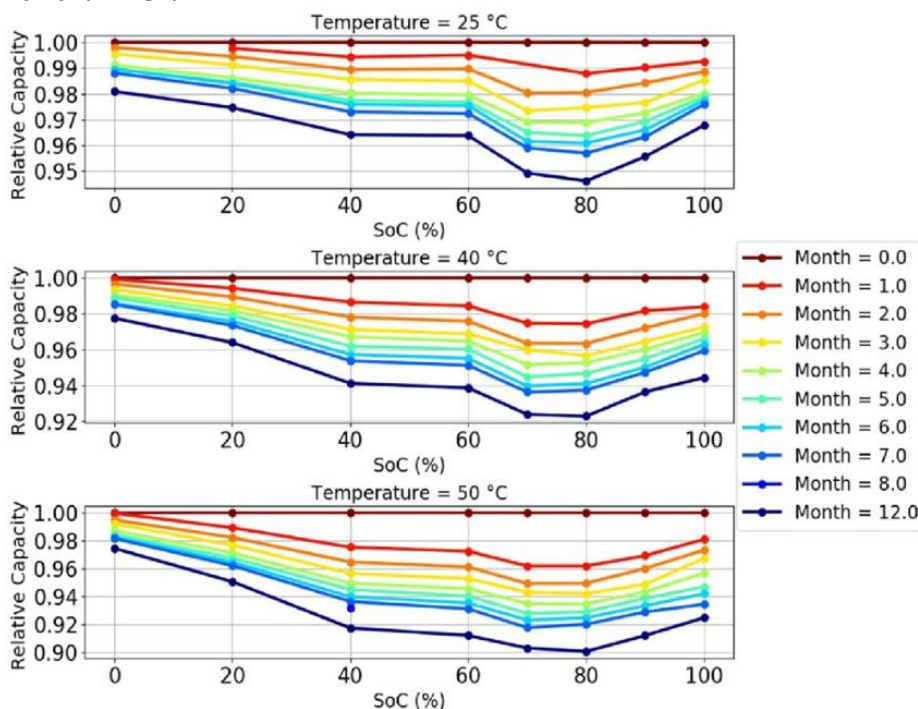


Figure 4. Time evolution of the relative capacity as function of the storage SoC, for cells stored at 25 °C, 40 °C and 50 °C.

In Figure 3, the battery capacity is analyzed over a time at various SOC's and temperatures. The degradation is observed to be more rapid at higher SOC's until 70 %. At exceedance of 70 Up to 80 °C, the trend is akin to the one at 70 With Loss of capacity demanding further investigation after the period of 12 months. Curiously, cells that are kept at battery 90 % and battery full capacity 100 % are found to deteriorate the least.

Figure 4 depicts 'spoon profile', which also indicates a much better capacity retention for 100 % SOC stored cells compared to the 70

and 90 % SOC stored cells, with the least performance attained at SOC's of 70 % and 80 %. One year later, this capacity was reduced to approximately 94%, 92%, and 90%, at 25°C, 40°C, and 50°C, respectively. The most substantial fade occurs within the first three months, especially for SoC values exceeding 60%, after which the fading markedly decreases. At 25 °C, the capacity fade went down from 1.2%/month to 0.2% by the end of the year; at 40 °C, it went down from 2.5% to 0.3%, and at 50 °C, from 3.8% to 0.4%. This observation is consistent with models that

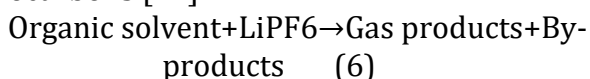
ascribe the early slumping to the growth of the solid-electrolyte interphase. Loss of lithium ions at the anode is a major contributor, though loss of active material is also hypothesized. The literature shows less variation of trends that look like 'spoons' with respect to different cell chemistries and different studies [11].

In a surprising twist, the increase in internal resistance (R_{int}) does not follow any of the trends of capacity fade. The only clear increase in R_{int} is seen after three months, particularly at 20% SoC or above, where a 30% increase in R_{int} was noted at 100% SoC and 50°C.

Electrolyte Decomposition:

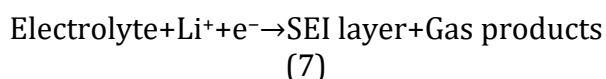
Decomposition of the electrolyte is associated with either extreme temperatures or excessive voltages. These processes of decomposition result in gas evolution and higher internal resistance which reduces the effectiveness of the battery. Its constituents can start forming as low as 30 °C to rapid degradation above 40 °C.

The electrolyte in lithium-ion batteries is usually composed of organic solvents (e.g., ethylene carbonate) and lithium salts (for instance, lithium hexafluorophosphate, LiPF_6). Under high temperatures, the components within the electrolyte – including organic solvents and lithium salts, decompose, and bore gases like CO_2 , CO and methane (CH_4) and other hydrocarbons [12].



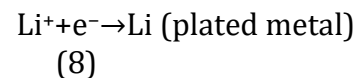
These reactions are especially common at temperatures higher than 30°C, and massive decomposition occurs above 40°C.

At higher voltages, in particular above 4.2 V, the electrolyte starts experiencing side reactions that cause formation of SEI (Solid Electrolyte Interphase) and gas evolution:



As an example of Lithium Plating, it occurs when lithium metal deposits on the anode surface instead of intercalating into the graphite structure. This often happens at high SoC (SOC) and low temperatures. Lithium plating leads to irreversible capacity loss and can cause safety issues such as short circuits. At

low temperatures or high charging rates, lithium ions (Li^+) are unable to intercalate into the anode quickly enough. Instead, lithium metal deposits on the surface, forming lithium dendrites.



That process is common especially often in charging the battery at a high SoC, because the graphite anode saturates and ceases to take up more lithium. It occurs by dissolution of a metallic lithium at excess current in the electrolyte and results irreversibly and reduces battery capacity, since the plated lithium becomes unavailable for intercalation.

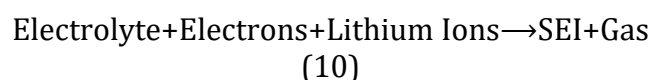
The Nernst equation can help understand the thermodynamic conditions leading to lithium plating by determining the potential at which Li metal deposition occurs:

$$E_{cell} = E^\circ - \frac{RT}{nF} \ln \left(\frac{\text{Li}^+}{\text{Li}} \right) \quad (9)$$

where, E° is the standard electrode potential, R is the gas constant, T is temperature, n is the number of electrons transferred, F is the Faraday constant, Li^+ and Li represent the concentrations of lithium ions and lithium metal, respectively.

These reactions slowly pull lithium from the cathode into the SEI, removing some of it from circulation and thus lowering overall battery performance.

Electrolyte decomposition also depends on the operating voltage of the battery. At high voltages (typically >4.2 V for lithium-ion batteries), side reactions involving the electrolyte become more likely: at lower voltages (~3.0–4.2 V), decomposition is minimal and above 4.2 V, decomposition rates increase, forming solid electrolyte interphase (SEI) layers and gas. The reaction can be described as:



Gas formation and SEI growth contribute to a rise in internal resistance, which is modeled as:

$$R_{int} = R_0 + \frac{1}{\sigma_{SEI}} \cdot L_{SEI} \quad (11)$$

where, R_0 is initial resistance, σ_{SEI} is SEI layer conductivity and L_{SEI} : SEI layer thickness [13].

One of the following reactions that affect the properties of Li-ion batteries (Nickel Manganese Cobalt Oxide, Nickel Cobalt Aluminum Oxide) is cathode degradation and

which can experience stage transitions at high SoC and temperature for extended storage time. This eventually turns out in cathode such that the active Lithium available at intercalation process decreases and it causes structural changes into the cathode.

Parameter	Effect Due to Aging	Influencing Factor
Capacity Fade	Reduction in the battery's ability to store charge. Typically, batteries lose 5-10% of capacity per year depending on temperature and SoC [14]	High temperature, high SoC
Power Fade	Reduction in the ability to deliver energy quickly, particularly in high-demand situations. Increased internal resistance limits the power output [15]	SEI growth, electrolyte degradation
Increased Resistance	The internal resistance of the battery increases due to SEI layer growth and the breakdown of electrolyte. Higher resistance causes additional heat generation during use [16]	Temperature, storage voltage

Table 1. Effects of Calendar Aging on various parameters.

It can be seen in the following table that examined the aging behavior of Nickel Manganese Cobalt (NMC) lithium-ion batteries stored at varying temperatures and SoC levels [17]. The study demonstrated that temperature and SoC have a compounding effect on capacity fade. Table 1 shows the capacity loss of NMC batteries after one year of storage under different conditions.

Storage Condition	Capacity Loss After 1 Year (%)
25°C, 50% SoC	2%
25°C, 100% SoC	5%
40°C, 50% SoC	4%
40°C, 100% SoC	10%

Table 1. Capacity Loss of NMC Lithium-ion Batteries After One Year of Storage

The data clearly indicates that both high temperature and high SoC accelerate the aging process. Batteries stored at 40°C and 100% SoC experienced up to 10% capacity fade, which is five times higher than those stored at 25°C and 50% SoC. The interaction of high temperatures and high voltage stresses the electrodes and leads to more rapid degradation of both the electrolyte and the SEI layer, as observed in various post-mortem studies [18].

The solid electrolyte interphase (SEI) layers are of great importance for the efficiency of the battery. However, their increase thickness during storage causes an increase in internal resistance and a decrease in capacity. Conducted studies in which Graphite/LiCoO₂ cells were long-term stored under several voltages, and it was observed that the more the voltage the faster the growth of the SEI layer, which caused more capacity loss [2].

Storage Voltage	Temperature (°C)	SEI Layer Thickness (nm)	Capacity Loss (%)
4.2V	25°C	35	5.1
4.2V	40°C	50	7.8
3.7V	25°C	12	1.8

3.7V 40°C 18 3.5

Table 2. SEI Layer Growth as a Function of Voltage and Temperature

It was found that when stored at a high voltage of 4.2V and a temperature of 40 °C, a solid electrolyte interface (SEI) layer of 50 nm thickness was formed, which is almost twice than that which forms at 3.7V and a temperature of 25 °C. The increase in thickness of the SEI layer is directly proportional to the increase capacity loss which lays out the necessity of managing voltage and temperature during prolonged storage.

Calendar aging is one of the aspects which differ due to the type of cathode material as well. Compared performance of calendar aging of LiFePO4 (LFP) against NMC chemistries. At higher temperature levels, LFP which is quite stable in terms of temperature showed significantly less capacity fade than NMC [6].

Cathode Material	Storage Condition	Capacity Loss After 1 Year (%)
NMC	40°C, 50% SoC	4%
LFP	40°C, 50% SoC	1%
NMC	40°C, 100% SoC	10%
LFP	40°C, 100% SoC	3%

Table 3. Capacity Fade of Different Cathode Materials at Elevated Temperatures

Analysis indicates that batteries LFP are more durable with respect to high temperature thermal degradation than NMC batteries as after one year of storage the temperature storage loss capacity was 1-3% variation for LFP while 4 -10% variation for nmc batteries. This is due to the stable interaction between the cathode and electrolyte in LFP cells which prevents excessive electrolyte degradation and SEI buildup.

In summary, calendar aging is strongly influenced by storage temperature, SoC, and electrolyte composition. A condensed overview of the main results of the studies on the issues provided in the table is presented in the table 4 [2,18,6].

Parameter	Condition	Capacity Fade
Temperature	40°C, 100% SoC	10%
Temperature	25°C, 50% SoC	2%
Additives	40°C, 100% SoC	5%
Cathode Material	NMC, 40°C, 100% SoC	10%
Cathode Material	LFP, 40°C, 100% SoC	3%

Table 4. Calendar Aging Effects from Various Studies

The results presented in the table schematically illustrate how external factors and selection of materials affect the calendar aging of lithium-ion batteries. If these parameters are adequately managed, the lifetime expectancy of batteries will be considerably improved, especially in use cases such as electric cars and grid storage.

Conclusion

This review examines the long-term performance and storage conditions of lithium-ion batteries, particularly how the mechanisms of calendar aging are affected. Calendar aging, which is temperature and SOC related, is responsible for certain effects like capacity fade, raised internal resistance, both of which occur over time even when batteries are not

operational. The heat promoted growth of the Solid Electrolyte Interphase layer at the extreme charge states of the battery makes it wear out faster. The review incorporates the in detail technical reports which helps to photographically illustrate how the health of the battery is affected by different storage conditions. These reports as well as predictive ones with empirical and semi-empirical as well as physics-based models make it possible to estimate the State of Health and the Remaining Useful Life of the systems. Such knowledge is important in coming up with operating policies which include thermal management and optimal SOC levels so as to mitigate calendar ageing. In EVs, better management of the SOC of the battery system will diminish the impact of

storage on battery deterioration, resulting in longer range and fewer battery replacements. With grid energy storage applications, the correct storage environment allows for higher retained capacity which is essential for reliable use of intermittent renewable energy sources.

In conclusion, this piece of work uses available theoretical models alongside technical results to enhance the understanding of calendar aging and give solutions to prolonging the life of batteries thus supporting green energy.

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