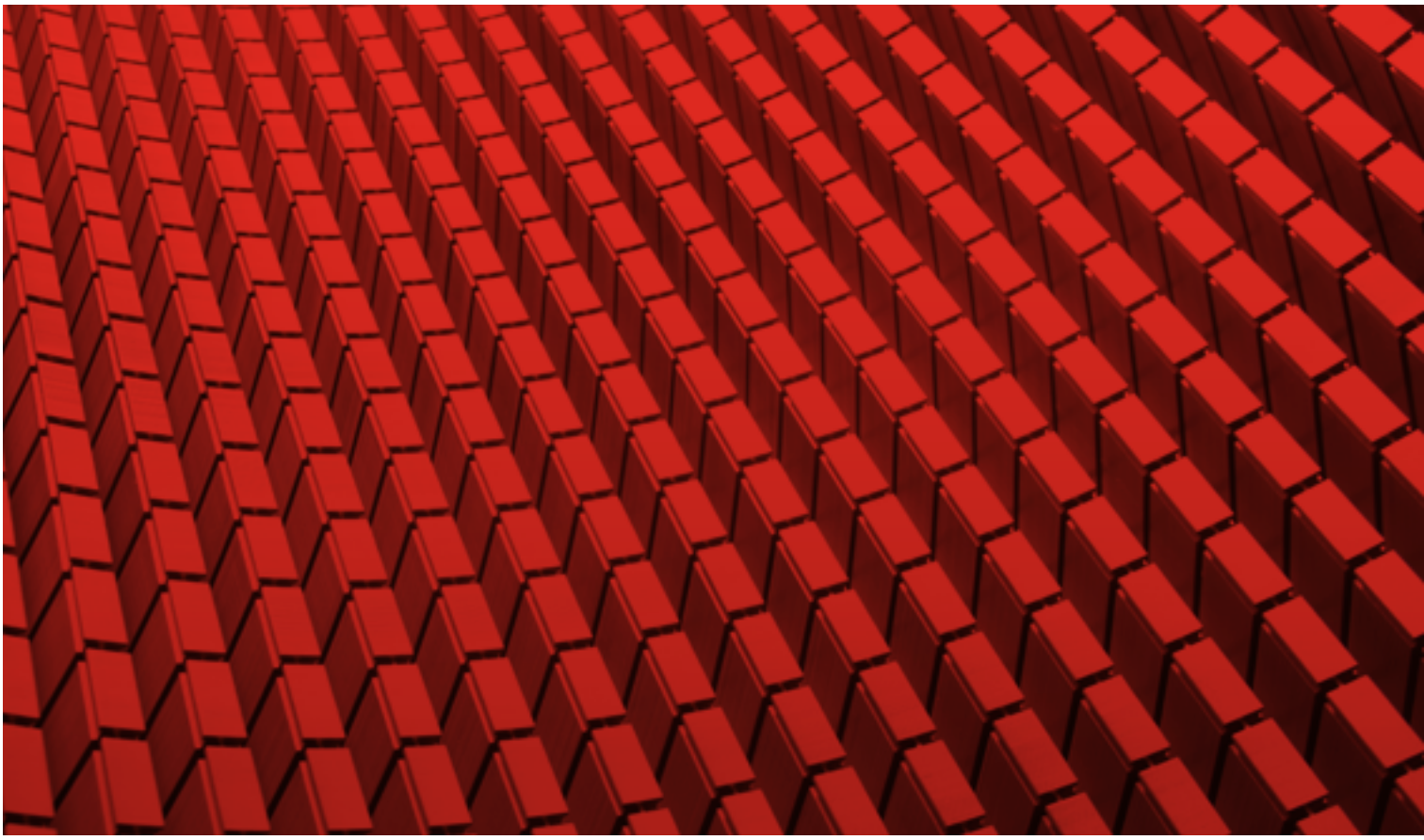
TRACTION DIVISION

Characteristics of LTO batteries

White paper



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1. Introduction

Within a very short time, lithium-ion batteries have become ubiquitous in applications from mobile devices to hybrid and full-electric cars and planes, wherever high energy density, high power, and long lifetime are required. Lithium-ion batteries can serve such diverse applications exceptionally well because they allow optimization with regard to the specific requirements of the application. For example, lithium-ion batteries for mobile devices may be optimized for high energy density at low power, whereas lithium-ion batteries in hybrid electric cars may be optimized for high power at lower energy density.

Various design choices allow us to optimize lithium-ion batteries to application requirements. Such design choices include the format of the battery cell, the internal electrode design, and the selection of electrolyte and separator. The most important design choice by far, however, is the selection of active materials, i.e., the materials that store lithium ions.

Commonly used active materials in the positive electrode of a lithium-ion cell include nickel-metal-oxides such as nickel-manganese cobalt (NMC), lithium-metal-phosphates such as lithium-iron-phosphate (LFP), and manganese based spinel type such as lithium-manganese oxide (LMO).

More important for the properties of the lithium-ion cell, however, is the choice of active material in the negative electrode. Crucial chemical processes that determine the safety, lifetime, and performance of the lithium-ion battery cell, such as solid electrolyte interface formation (Section 3.3) and lithium plating (Section 3.5) depend heavily on the choice of material for the negative electrode. Commonly used active materials include graphite (C) and lithium-titanate (LTO), and the properties of these materials are therefore analyzed in detail in this white paper.

This white paper focuses on three aspects that are especially important in commercial applications with a focus on transportation:

- Safety of the lithium-ion battery: Lithium-ion batteries store large amounts of energy, the accidental release of which can have disastrous consequences in applications that involve the transport of people or operation in harsh and badly accessible environments.
- Performance: Lithium-ion batteries in commercial applications need to provide the required power to, e.g., move heavy commercial vehicles. Even more importantly, lithium-ion batteries in commercial vehicles need to be charged in short times to maximize the availability of the vehicle.
- Total cost of ownership: Batteries in commercial applications are operated for many hours per day at high power levels. To reach a low total cost of ownership, a long useful life of the battery is required.

This white paper is structured as follows:

- Chapter 2 gives a general overview on lithium-ion cells and explains in more detail the design choices listed above.
- Chapter 3 explains in detail the different properties of lithium-ion batteries according to a specific battery design.
- Chapter 4 has a special focus on the lifetime of lithium-ion batteries.
- Chapter 5 summarizes the main points of the preceding chapters and relates them to the requirements in real applications.

2. Design of lithium-ion battery cells

2.1. Lithium-ion technology

Lithium-ion battery cells are often described as “rocking chair batteries”. This term is used to describe that lithium ions are the energy carriers that shuttle back and forth between a zone with high electrochemical energy and a zone with low electrochemical energy, thereby storing or releasing energy in the process.

More specifically, during charging, lithium ions move from a zone with low electrochemical energy (positive electrode) to a zone with high electrochemical energy (negative electrode), where they are stored in a state with high energy. During discharging, the lithium ions move from the zone with high electrochemical energy (negative electrode) to the zone with low electrochemical energy (positive electrode) and release the previously stored energy.

2.2. Components of lithium-ion battery cells

The four main components of a lithium-ion battery cell are (shown in figure 2-1):

- The active material used for the negative electrode (anode), in which lithium ions are stored at high electrochemical energy.
- The active material used for the positive electrode (cathode), in which lithium ions are stored at low electrochemical energy.
- The separator, which separates the zone of high potential energy and low potential energy. The separator prevents direct contact and, thus, internal short circuit.
- The electrolyte, which is a mixture of salts and solvents. The electrolyte needs to selectively allow the lithium ions to move from positive electrode to negative electrode or vice versa. At the same time, no electrons may move in the electrolyte to prevent self-discharge.

Active materials and separator have porous structures which are soaked by electrolyte. The electrolyte thereby allows lithium ions to travel with low effort between positive and negative electrode materials and to transverse the separator.

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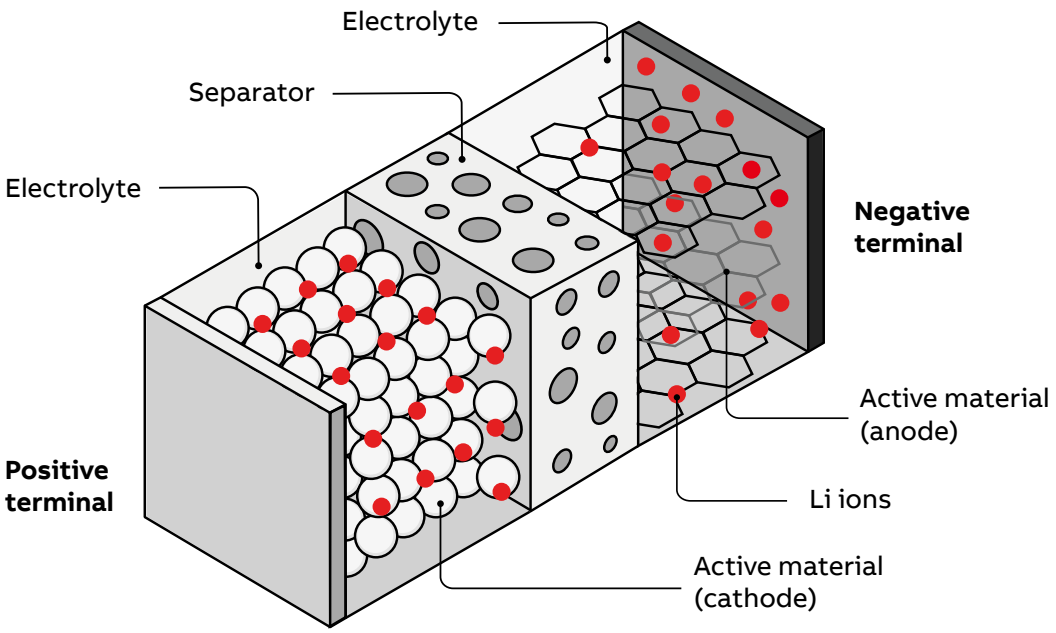
Figure 2-1: Main components of lithium-ion battery cells[1]



1 Taken from “How does a lithium-Ion battery work?” | Let’s Talk Science (letstalkscience.ca). Labels have been adapted

2.3. Classification

Lithium-ion cells can be classified according to the components used to realize them. Anode and cathode materials have the strongest impact on cell properties, so the most common of classification describes the materials in the positive and negative electrode, respectively. Figure 2-2 shows the most typical electrode materials and three examples of lithium-ion chemistries that are realized by combining suitable electrode materials:

- «NMC» lithium-ion battery cells combine an NMC (nickel-manganese-cobalt) cathode and a graphite anode.
- «LFP» lithium-ion battery cells combine an LFP (lithium-iron-phosphate) cathode and a graphite anode.
- «LTO» lithium-ion battery cells combine an NMC (nickel-manganese-cobalt) cathode and an LTO (lithium-titanate) anode. More rarely, an LMO (lithium-manganese) cathode is used for LTO cells as well.

The choice of the anode material[1] has a dominant impact on the performance of the lithium-ion cell and is discussed in detail in Chapter 3.

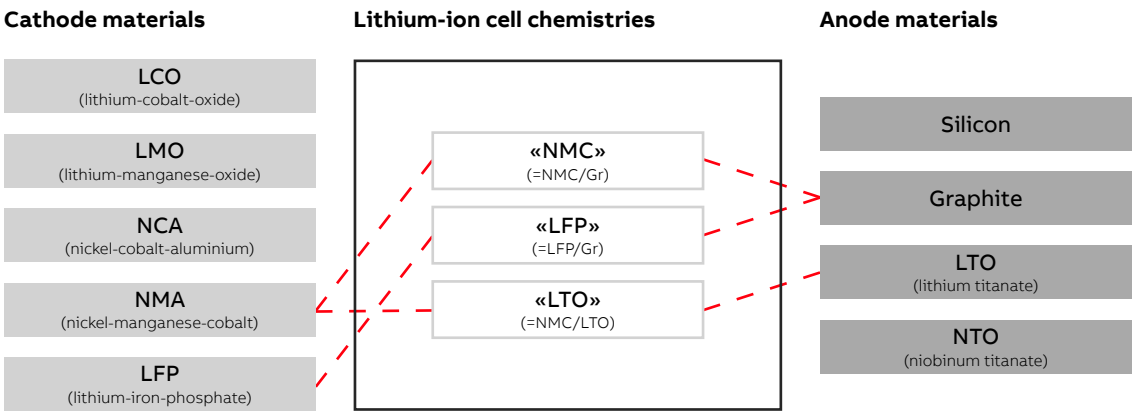


Figure 2-2: Typical electrode materials of lithium-ion cells and resulting cell chemistries

A second-level classification describes the mechanical format of the cell. The most important formats on the market are cylindrical, prismatic, and pouch cells, as shown in Figure 2-3.

Finally, lithium-ion cells can also be classified according to the materials used for separator and electrolyte. These materials are rarely specified in detail by cell manufacturers.

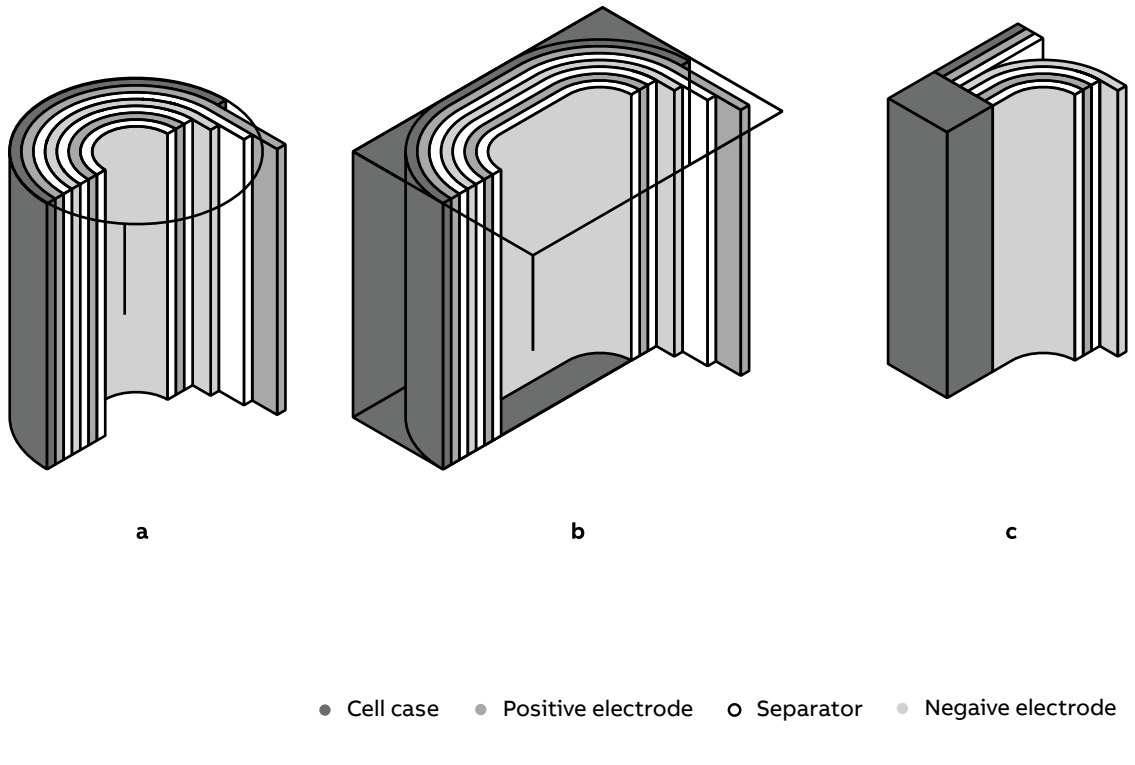


Figure 2-3: Common lithium-ion cell formats, (a) cylindrical cell, (b) prismatic cell, (c) pouch cell[2]

1 Of the four anode materials shown in Figure 2-2, only graphite and LTO are used on an industrial scale. Niobium titanate anode materials have very similar properties to LTO anode materials but can provide for higher energy density and are currently in research state. Silicon anode materials allow extremely high energy density but show very poor lifetime and are not used in industrial scales (see Section 3.7)

2 Bilgin et al., "Making the Case for Electrified Transportation", IEEE Transactions on Transportation Electrification, vol. 1, 2015

3. Properties of lithium-ion cells

3.1. Voltage

Lithium-ion cells are realized with a region with high electrochemical energy for the lithium ions (the anode) and a region with low electrochemical energy for the lithium ions (the cathode), see Section 2.1. The potential difference is manifested as a voltage at the terminals of the cell.

The potentials of the electrode materials are not constant. They typically depend on the amount of lithium ions stored in it (the degree of lithiation). Figure 3-1 shows the electrode potentials of typical electrode materials used in lithium-ion cells.

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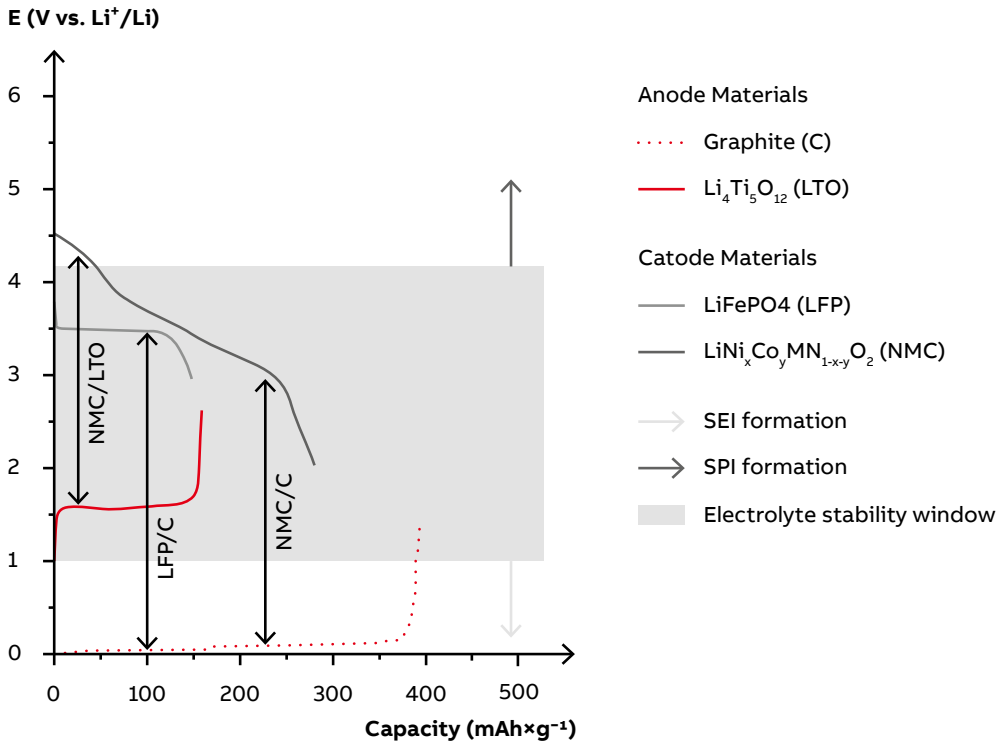
Figure 3-1: Electrode potentials of typical electrode materials and the stability window of the electrolyte[1]



In the example of an NMC/Graphite cell, the cathode (NMC) potential varies between 2 V and 4.5 V as a function of state-of-charge, whereas graphite (C) has a relatively stable potential of 0–250 mV versus Li/Li+. The mean difference between cathode and anode is approximately 3.7 V, which is a measure for the energy which can be stored in a single lithium ion.

In the example of an NMC/LTO cell, LTO has a higher potential of approximately 1.5 V versus Li/Li+, which lowers to mean difference between cathode and anode to approximately 2.3 V.

Example
3.7 V is a typical nominal voltage of an NMC/C lithium-ion cell, whereas 2.3 V is a typical nominal voltage of an NMC/LTO lithium-ion cell.

Each lithium ion in an NMC/C cell can therefore store more energy than a lithium ion in an NMC/LTO cell. Consequently, the energy density of a typical NMC/C cell is higher than the energy density of an NMC/LTO cell. At the same time, the lower voltage of graphite introduces additional challenges, which are discussed in the following sections.

1 Zhao et al., "Film-forming electrolyte additives for rechargeable lithium-ion batteries: progress and outlook", Journal of Materials Chemistry A, vol. 7, 2019

3.2. Stability of the electrolyte

The potentials of the electrode materials not only determine the energy densities of the battery cell, but they also impact the other components used in the battery cell. Graphite has a very low electrochemical potential (only 150 mV higher than lithium itself, which is a very reactive material) and is similarly reactive. As can be seen in Figure 3-1, the potential of graphite is so low that it is even outside of the stability window of state-of-the-art electrolytes.

The high reactivity of graphite therefore leads to a comparably low lifetime by driving chemical side reactions, which consume lithium and make electrodes unusable. Specifically, the high reactivity of the graphite causes the gradual decomposition of electrolyte. Decomposition products form a layer on the anode, the solid electrolyte interface (SEI) – see Section 3.3 – which lowers the capacity of the cell and increases its internal resistance.

By comparison, the higher electrochemical potential of LTO penalizes the energy density of the cell, but it also means that both electrodes are now operated within the stability window of the electrolyte, which makes the cell much more stable.

3.3. Solid electrolyte interface formation

The instability of the electrolyte in contact with the graphite anode manifests itself in the decomposition of the electrolyte, as shown in Figure 3-2. In a cell with graphite anode, this decomposition does not occur in an uncontrolled way. Instead, the decomposition products form a passivating layer on the anode to protect the electrolyte from continued decomposition. The passivation layer is called the solid electrolyte interface (SEI).

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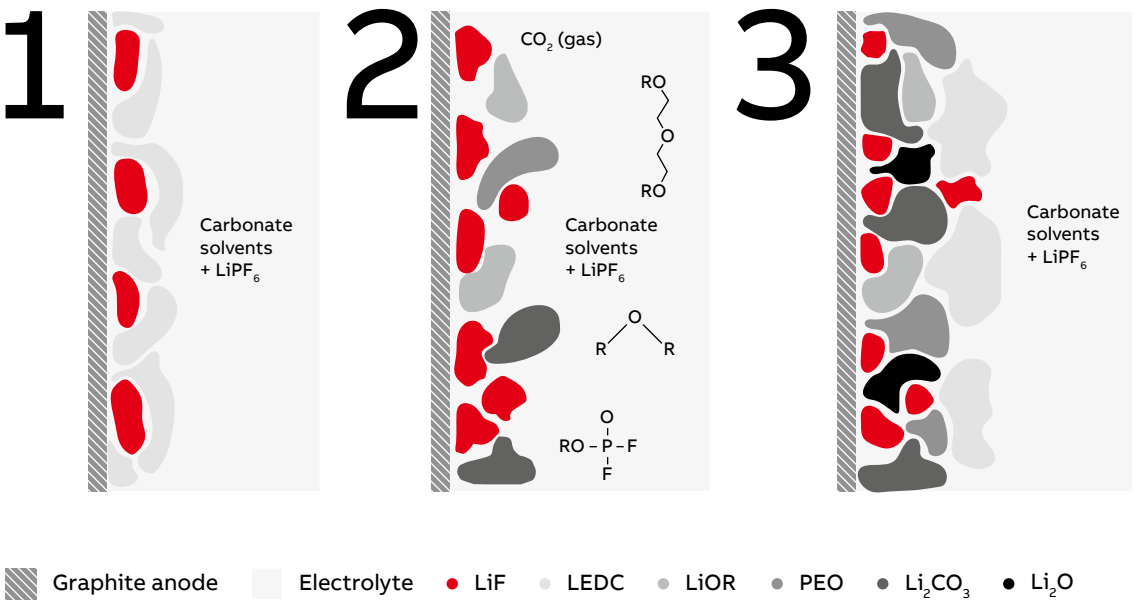
Figure 3-2: Formation of the solid electrolyte interface[1]



1. Initial reduction generates LEDC and LiF

2. Upon aging, the instability of LEDC reults in decomposition and dissolution, increasing SEI porosity

3. Continued cycling results in EC reduction, thickening of the SEI and more stable inorganic species near the surface

While protecting the electrolyte from decomposition, the formation of the solid electrolyte interface layer results in disadvantages.

- The solid electrolyte interface is thermodynamically unstable and can go into thermal runaway at comparatively low temperatures. Decomposition and thermal runaway of the solid electrolyte interface are often the starting point of thermal runaway of the lithium-ion cell (see Section 3.8).
- The solid electrolyte interface is an additional layer for lithium-ions to transfer during charge and discharge processes and therefore increases the internal resistance of the lithium-ion cell.

1 Mukai, "Zero-Strain Insertion Materials for All-Solid-State Li-Ion Batteries", Solid Electrolytes for Advanced Applications, 2019

Most importantly, the solid electrolyte interface is mechanically not perfectly stable. Due to volume expansion or contraction of the graphite anode (see Section 3.7), the solid electrolyte cracks and crumbles and needs to be re-formed, as shown in Figure 3-3. As can be seen from Figure 3-3, the formation of the solid electrolyte interface consumes lithium and solvent. The consumed lithium and solvent are no longer available to store energy or allow the transfer of lithium ions. The continuous formation of solid electrolyte interface therefore lowers the capacity and increases the internal resistance. As such, the continuous formation of solid electrolyte interface drives the aging of lithium-ion cells with graphite anode.

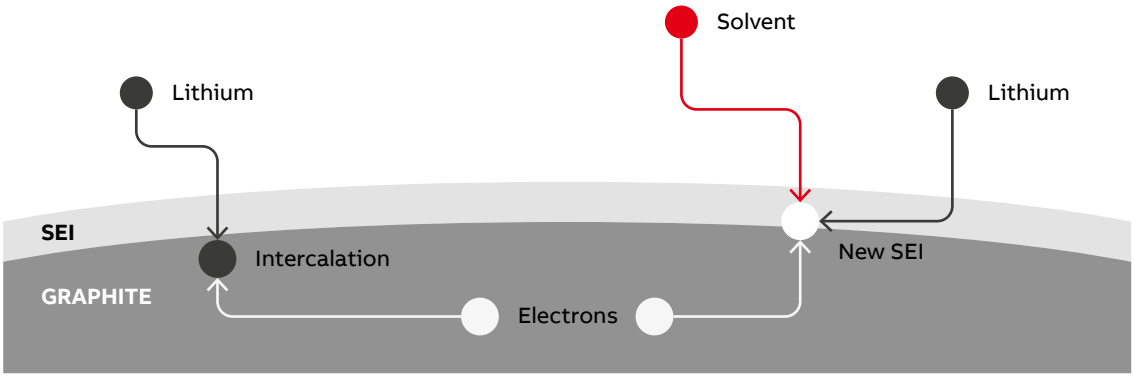


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Figure 3-3: Continuous formation of solid electrolyte interface[1]

3.4. Absence of SEI formation in LTO cells

Lithium-ion cells with LTO anode do not form a solid electrolyte interface, as the anode potential is always within the stability window of the electrolyte (Figure 3-1).

Looking very closely at Figure 3-1, one can see that at very high cell voltages (>2.6 V), the cathode potential is also outside of the stability window of the electrolyte. If this is the case, a similar process takes place at the cathode, forming the solid permeable interface (SPI). However, as the potential is still very close to the stability window and as operation at very high cell voltage is only intermittent, this process is much slower than the solid electrolyte interface formation at the graphite anode.

The absence of solid electrolyte interface formation is one of the major reasons for the superior lifetime and safety of LTO lithium-ion cells (see Chapter 4).

3.5. Lithium plating

This section explains how lithium is stored in the active materials of the positive and negative electrodes during charge and discharge processes. The potential difference between metallic lithium and the active materials ensures that this intercalation process takes place.

However, in the case of a graphite anode, the potential difference between graphite and bare lithium is only 150 mV. At high charge currents or when the movement of lithium ions is additionally restricted, e.g., at low temperatures, it can become energetically favorable for lithium ions to accumulate as metallic lithium at the surface of the anode rather than intercalate into it. The solid electrolyte interface is an additional obstacle for lithium ions to transfer before being intercalated into the graphite anode and therefore further exacerbates this problem, Figure 3-4.

Example
It is a well-known recommendation for batteries in end-user applications, such as cell phones or electric bikes, not to charge such devices at low temperatures. This recommendation is directly related to the risk of lithium plating, internal short circuit and resulting fire.

When bare lithium accumulates at the surface of the anode, it forms lithium dendrites. Dendrites are sharp, needle-like structures which can pierce the separator (see Figure 3-4). When the separator is damaged, the cell is short-circuited internally. In case of a cell-internal short circuit, all the energy of the cell is released into the cell itself, producing heat and potentially thermal runaway.

1 Heiskanen et al., “Generation and Evolution of the Solid Electrolyte Interphase of Lithium-Ion Batteries”, Joule, Volume 3, Issue 10, 2019

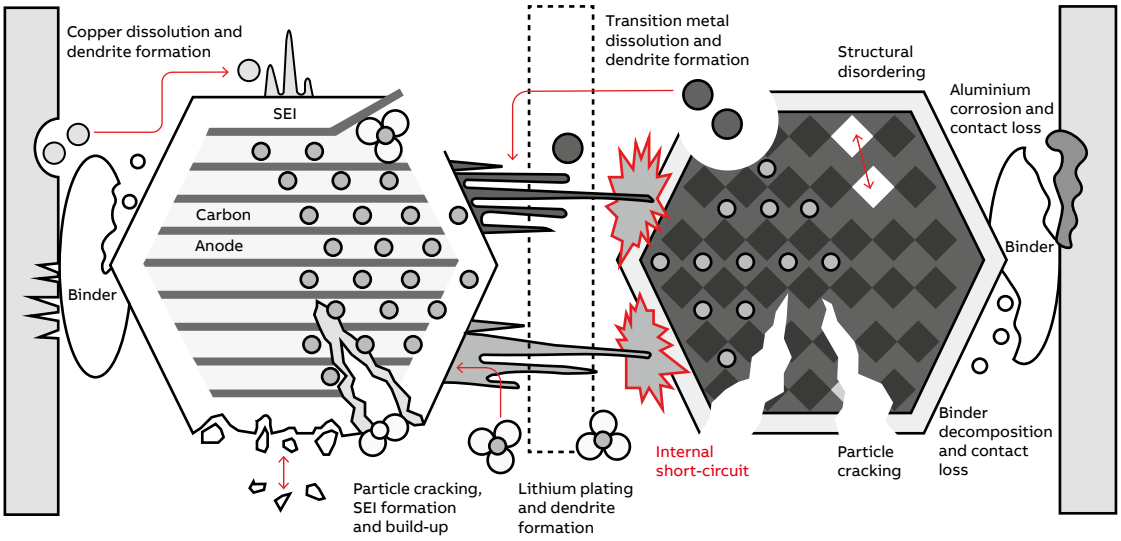

Figure 3-4: Lithium plating and cell-internal short circuit[1]

3.6. Absence of lithium plating in LTO cells

If an LTO cell is operated within its allowed voltage window, lithium plating does not occur in the lithium-ion battery cell with LTO anode. Specifically, the potential difference between bare lithium and LTO is more than ten times higher than the potential difference between bare lithium and graphite. Additionally, the absence of the solid electrolyte allows much easier intercalation into the anode material itself. Consequently, a lithium-ion cell with LTO anode can be safely charged at high power, even at very low, sub-zero temperatures.

3.7. Volumetric expansion and strain

When lithium ions are intercalated or deintercalated from the electrode materials, such materials may undergo volume changes. Volume changes impose mechanical stress and can reduce the performance of the cell due to mechanical degradation. Aging mechanisms which are driven by mechanical stress are described in more detail in Section 4.3. Figure 3-5 shows the volume expansion of different electrode materials used in lithium-ion cells.

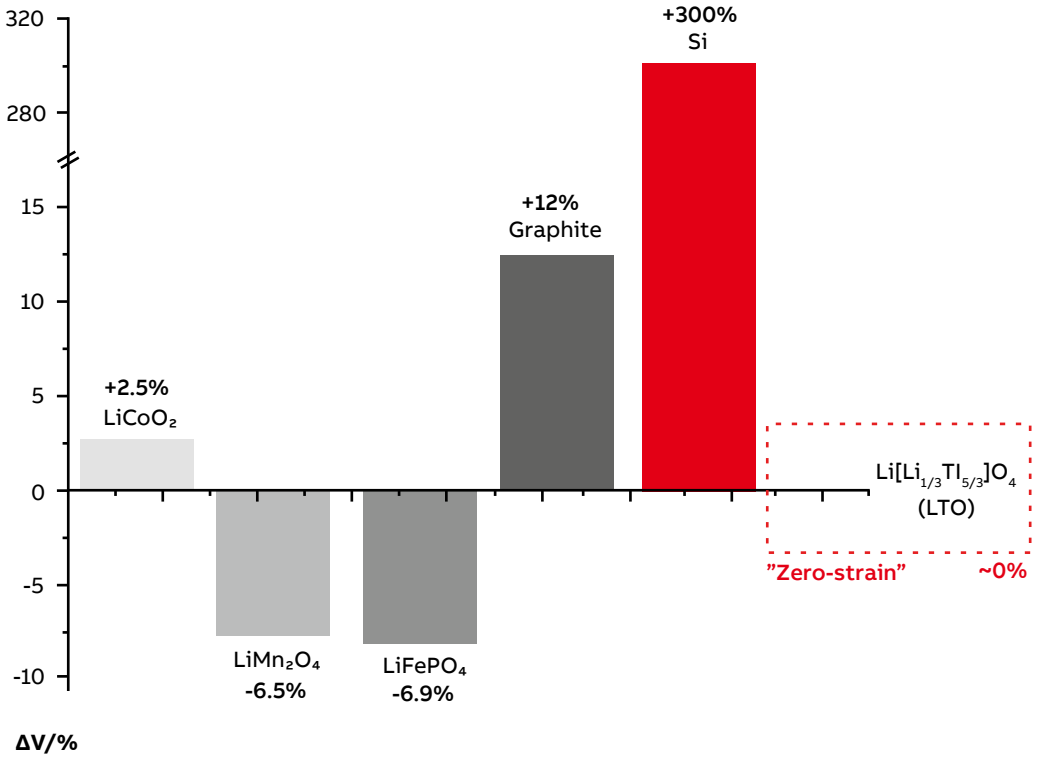

Figure 3-5: Volume expansion of electrode materials[2]

1 Birkl et al., "Degradation diagnostics for lithium ion cells", Journal of Power Sources, vol. 341, 2017

2 Pinson et al., "Theory of SEI Formation in Rechargeable Batteries: Capacity Fade, Accelerated Aging and Lifetime Prediction", 1210.3672.pdf (arxiv.org)

The volume of lithiated graphite can vary by up to 12% between the fully lithiated state (corresponding to a fully charged cell) and the fully delithiated state (corresponding to a fully discharged cell). Such volume changes do not only damage the anode particles themselves; more importantly, they lead to continuous cracking and re-formation of the solid electrolyte interface (see Section 3.3). The continuous re-formation of the solid electrolyte consumes lithium and slowly reduces the capacity of the lithium-ion cell.

Example
Silicon anodes can promise exceptional energy densities for lithium-ion cells, however, the mechanical stress produced by volume changes of up to 300% currently allows for only a very small number of cycles, and no economical lifetimes can be reached with the current state of technology.

LTO, on the other hand, shows virtually no volume change during intercalation or deintercalation of lithium ions. It is commonly referred to as a zero-strain material. Together with the absence of the solid-electrolyte interface, the absence of volume changes contributes to the exceptional lifetime of LTO lithium-ion cells.

3.8. Thermal runaway

Thermal runaway is the most critical safety related process in lithium-ion cells. Thermal runaway denotes a process in which the lithium-ion cell is decomposing exothermally and releasing heat at ever-accelerating rate. Such a process cannot be stopped and eventually leads to venting, fire, or explosion of the battery cell.

Thermal runaway may be triggered in a variety of ways, such as external overheating, overcharging, or due to cell internal short (circuit, see Section 3.5). The process of thermal runaway starts when the heat produced in such events elevates the temperature of the cell above the maximum temperature that can be sustained by one or several materials inside the lithium-ion cell. In a chain of events, the heat released by the decomposition of such materials will further heat the cell until the maximum temperature of more and more materials inside the cell is exceeded.

Figure 3-6 shows the typical chain of events during a thermal runaway. Thermal runaway typically starts at the solid electrolyte interface, which is a thermodynamically unstable material (see Section 3.3). The heat released during decomposition of the solid electrolyte interface causes thermal runaway of the (graphite) anode and accelerated reactions at the interface between electrolyte and graphite anode.

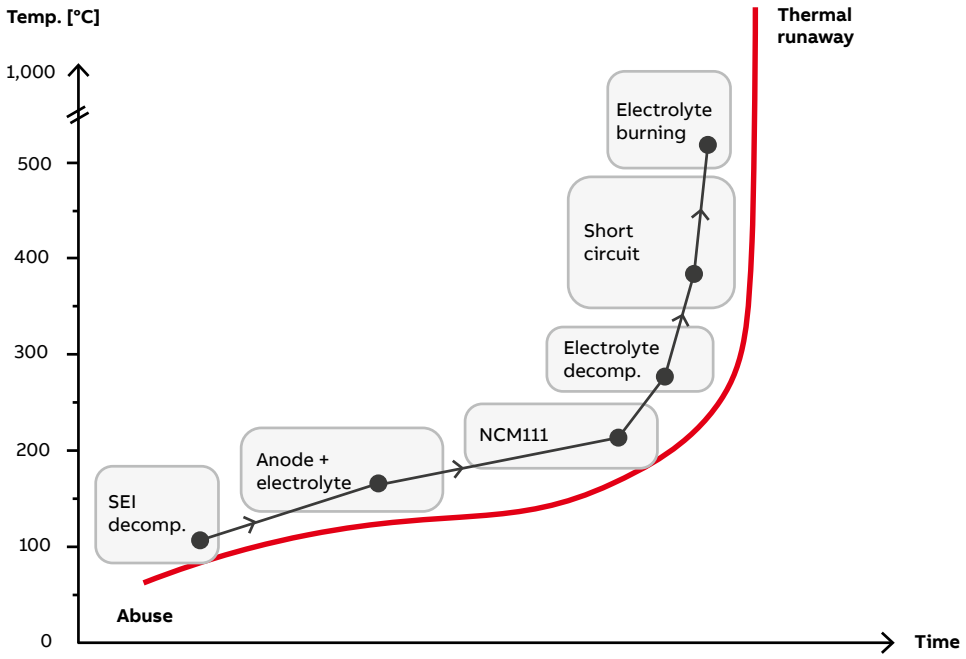

Figure 3-6: Chain of events during thermal runaway[1]

Lithium-ion cells with LTO anode do not exhibit a solid-electrolyte interface (see Section 3.4). Compared to cells with graphite anode, the component with lowest onset temperature for thermal runaway has thus been eliminated. Consequently, lithium-ion cells with LTO anode are stable until much higher temperatures and therefore less prone to thermal runaway than battery cells with graphite anode.

1 Feng et al., "Thermal runaway mechanism of lithium ion battery for electric vehicles: A review", Energy Storage Materials, vol. 10, 2018

4. Aging

4.1. Overview

Chapter 3 has already described processes that change the composition of a lithium-ion cell, such as decomposition of electrolyte (Section 3.2) and solid-electrolyte formation while consuming lithium (Section 3.3). The loss of electrolyte reduces the capability of the lithium-ion cell to transfer lithium ions between the positive and negative electrode, i.e., it increases the internal resistance of the lithium-ion cell. Similarly, the lithium which is consumed to form the solid electrolyte interface can no longer store energy, i.e., the capacity of the cell is reduced.

Increase of internal resistance and loss of capacity are generally referred to as “aging” of the lithium-ion cell. When the lithium-ion cell ages, its performance reduces. When the performance has reduced to a level at which the lithium-ion cell can no longer meet the demands of the application, the lithium-ion cell is said to have reached its “end-of-life”. A lithium-ion cell at its end-of-life needs to be disposed of and replaced, at high cost, to continue meeting the demands of the

Cause and effect of degradation mechanisms and associated degradation modes

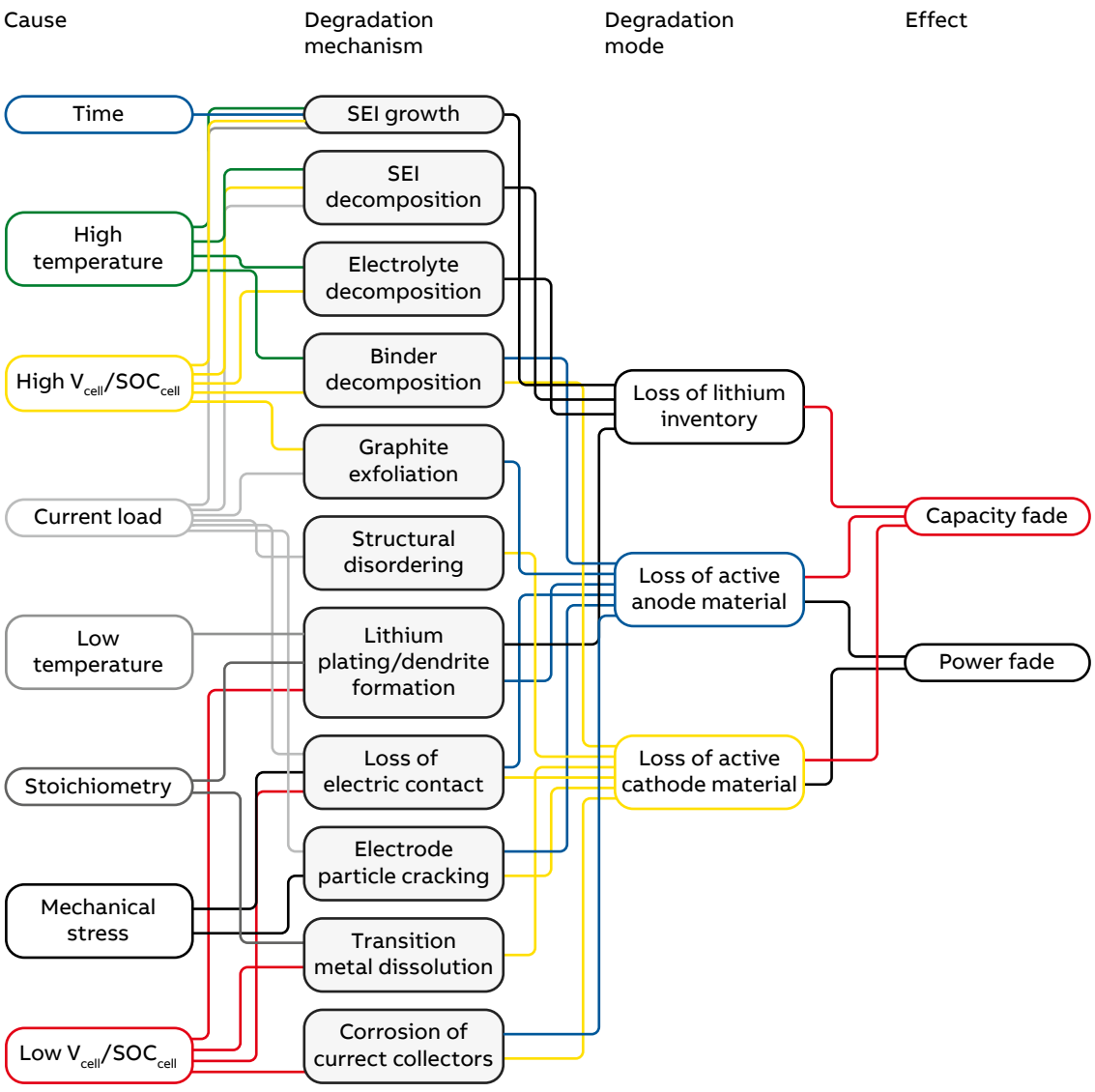

Figure 4-1: Degradation mechanisms in lithium ion cells, application conditions in which they are triggered, and impact on the performance of the lithium-ion cell[1]

1 Birkl et al., “Degradation diagnostics for lithium ion cells”, Journal of Power Sources, vol. 341, 2017

application. A long useful lifetime therefore allows a low total cost of ownership by maximizing the lifetime of the initial investment and saving on cost and effort for replacements at short intervals.

This chapter provides an overview on aging mechanisms in lithium-ion battery cells. Figure 4-1 shows the dominant degradation mechanisms in lithium-ion cells, how such degradation mechanisms are triggered and their effect on the lithium-ion cell. Based on Figure 4-1, the following sections further explain degradation mechanisms in lithium-ion cells, compare the lifetime of lithium-ion cells based on graphite and LTO anodes, and explain the superior lifetime of lithium-ion cells with LTO anode.

Lithium-ion cells age both during storage and during operation. Aging during storage is referred to as calendar aging, whereas aging during operation is referred to as cycle aging. Typically, more degradation mechanisms are triggered during cycling than during storage, and aging during operation is faster than aging during storage at similar conditions.

4.2. Degradation due to SEI formation

4.2.1. Overview

The first group of degradation mechanisms is related to solid electrolyte interface formation. This group comprises SEI growth, SEI decomposition, electrolyte decomposition, and lithium plating and dendrite formation. All degradation mechanisms in this group can be traced to a common cause: the instability of the electrolyte at the potential of the electrode (see Sections 3.2 and 3.3).

In application, such aging mechanisms are accelerated at high voltages and high temperatures.

4.2.2. Impact on lithium-ion cells with graphite and LTO anode respectively

In a cell with graphite anode, degradation due to SEI formation, decomposition, and re-formation are the dominant aging mechanism. These degradation mechanisms can lead to calendar aging, but they are especially severe when coupled with mechanical stress. In such a case, the solid electrolyte interface cracks and constantly needs to be re-formed (see Section 3.7).

A lithium-ion cell with LTO anode exhibits interface formation on a much smaller scale only (see Section 3.4). The absence of the solid electrolyte interface is the primary reason for the exceptional lifetime of cells with LTO anode compared to cells with graphite anode.

4.3. Degradation due to mechanical stress

4.3.1. Overview

The second group of degradation mechanisms is related to mechanical stress. This group comprises exfoliation of graphite, loss of electric contact, and electrode particle cracking. Mechanical stress can be exerted from externally, but it can also be produced by processes within the cell itself. Lithium-ion cells in prismatic and cylindrical format (Section 2.3) are well protected from external stress whereas lithium-ion cells in pouch format need to be protected more carefully.

Stress produced inside the cell itself is generally more relevant. The main reason for mechanical stress is volume changes of active materials during charge and discharge cycles (see Section 3.7). However, pressure build-up due to electrolyte decomposition and production of gas is another reason.

In operation, mechanical stress is mainly caused by temperature cycles, high current rates, and cycles with high cycle depth.

4.3.2. Impact on lithium-ion cells with graphite and LTO anode respectively

In a cell with graphite anode, mechanical stress due to expansion and contraction of anode particles (Section 3.7) and the resulting cracking of anodes particles, cracking of SEI, and re-formation of SEI is the main reason for aging (see Section 4.2.2).

A lithium-ion cell with LTO anode exploits the zero-strain property of LTO (Section 3.7), and aging due to mechanical stress is much slower. The absence of mechanical stress is another main reason for the exceptional lifetime of cells with LTO anode.

4.4. Degradation due to chemical processes

4.4.1. Overview

The third and last group of degradation mechanisms is related to chemical degradation of the components within the lithium-ion cell. This group comprises binder decomposition, structural disordering, corrosion of current collectors, and transition metal dissolution.

Binder decomposition refers to degradation of the binder material used to keep contact between electrode particles and the current collectors. Binder decomposition therefore eventually leads to loss of capacity. Structural disordering and transition metal dissolution refer to changes of the crystal structure of the cathode materials and, thus, the capability to store lithium ions. Transition metals, such as manganese, can also be dissolved in the electrolyte and migrate to the anode, thereby lowering the capacity and disturbing the solid electrolyte interface. Finally, corrosion of the current collector degrades the electrical contact between the current collector and the electrode materials, and increase the internal resistance.

Degradation due to chemical processes is strongly driven by extreme, i.e., both very high and very low, values of state-of-charge. Chemical processes are accelerated at high temperatures, which consequently accelerates the aging process.

4.4.2. Impact on lithium-ion cells with graphite and LTO anode respectively

Chemical degradation occurs primarily at the cathode. As such, the aging mechanisms described in this section apply to LTO cells as well as to cells with graphite anode. While there are differences between different cathode chemistries, aging processes at the cathode generally have a smaller impact compared to aging processes at the anode.

4.5. Summary

Figure 4-2 shows the resulting lifetime of selected lithium-ion cells at typical operating conditions (80% depth of discharge) versus specific energy. The graph first shows that even cells of the same chemistry, such as NMC, can be optimized with respect to specific energy or with respect to lifetime, however, both targets at once cannot be met. The “technology curve” clearly shows how lifetime reduces with increased specific energy. Secondly, it can be seen that LTO is very clearly far above the technology curve, even when extrapolated to the respectice specific energy.

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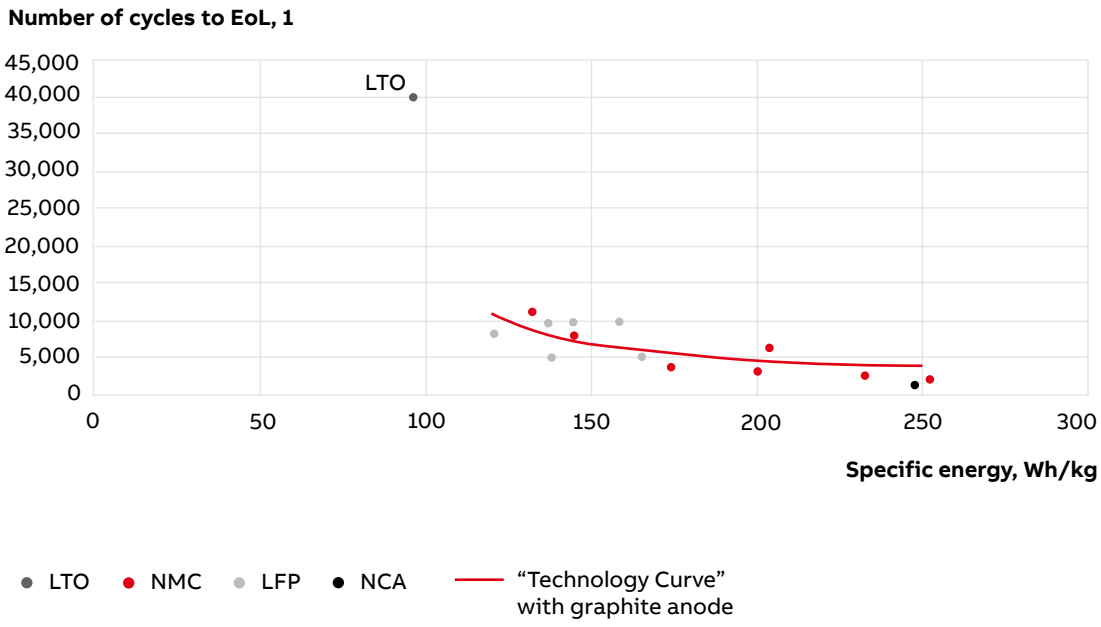
Figure 4-2: Lifetime of lithium-ion cells with LTO and graphite anode



5. Summary

This white paper has explained in detail the properties of lithium-ion cells, and the available design choices which affect energy density, cost, lifetime, and safety. The comparison has focused on properties with special importance for energy storage systems in commercial applications, namely, total cost of ownership, safety, and performance. It has been shown that the LTO cell chemistry performs exceptionally well with regards to these three requirements. In this last chapter, the most important characteristics are revisited:

5.1. Total cost of ownership

Due to the higher specific energy of lithium ions stored in a graphite anode (Section 3.1) battery cells with graphite anode have a higher specific energy. The higher specific energy also means that less material input is needed to realize a defined capacity, which also reduces the specific cost of battery cells with graphite anode.

At the same time, battery cells with LTO anode have higher performance and higher intrinsic lifetime (see Chapter 4). The same application requirements can typically be met with less installed capacity. Additionally, the energy storage system with LTO cells will have superior lifetime due to slow cycle aging and the virtual absence of calendar aging.

5.2. Safety

The safety of cells with graphite anode suffers from solid electrolyte interface formation (see Figure 3-2) and lithium plating (see Figure 3-4). The solid electrolyte is the trigger point for thermal runaway in cells with graphite anode.

By comparison, lithium-ion cells with LTO anode exhibit neither solid electrolyte interface formation nor lithium plating. The onset temperature for thermal runaway is much higher and the battery cell is both intrinsically safer and more robust to propagation.

5.3. Performance

The performance of cells with graphite anode, specifically the charge performance, is limited by lithium plating (see Section 3.5). At high charge rates, or at low temperatures, plated lithium can produce internal short circuit and destroy the battery cell.

By contrast, lithium-ion cells with LTO anode do not exhibit lithium plating (see Section 3.6). Consequently, lithium-ion cells with LTO anode can be charged at high rate even at very low, sub-zero temperatures. LTO batteries can typically be recharged in less than 10 minutes.

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