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<tr>
<td>ANN</td>
<td>Artificial Neural Network</td>
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<tr>
<td>BEV</td>
<td>Battery Electric Vehicle</td>
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<tr>
<td>BMS</td>
<td>Battery Management System</td>
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<tr>
<td>DOD</td>
<td>Depth Of Discharge</td>
</tr>
<tr>
<td>DSO</td>
<td>Distribution System Operator</td>
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<tr>
<td>ECM</td>
<td>Equivalent Circuit Model</td>
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<tr>
<td>EKF</td>
<td>Extended Kalman Filter</td>
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<tr>
<td>EOL</td>
<td>End Of Life</td>
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<tr>
<td>EV</td>
<td>Electric Vehicle</td>
</tr>
<tr>
<td>FCE</td>
<td>Full Cycle Equivalent</td>
</tr>
<tr>
<td>HEV</td>
<td>Hybrid Electric Vehicle</td>
</tr>
<tr>
<td>IC</td>
<td>Incremental Capacity</td>
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<tr>
<td>ICA</td>
<td>Incremental Capacity Analysis</td>
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<tr>
<td>LCA</td>
<td>Life Cycle Assesment</td>
</tr>
<tr>
<td>LFP</td>
<td>Lithium Iron Phospate</td>
</tr>
<tr>
<td>LIB</td>
<td>Lithium Ion Battery</td>
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<tr>
<td>LMO</td>
<td>Lithium Manganese Oxide</td>
</tr>
<tr>
<td>LNO</td>
<td>Lithium Nickel Oxide</td>
</tr>
<tr>
<td>LTO</td>
<td>Lithium Titanate Oxide</td>
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<tr>
<td>NCA</td>
<td>Nickel Cobalt Aluminium</td>
</tr>
<tr>
<td>NMC</td>
<td>Nickel Manganese Cobalt</td>
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<tr>
<td>OCV</td>
<td>Open Circuit Voltage</td>
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<tr>
<td>PHEV</td>
<td>Plug-In Electric Vehicle</td>
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<td>PV</td>
<td>Photovoltaics</td>
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<td>RT</td>
<td>Room Temperature</td>
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<tr>
<td>RUL</td>
<td>Remaining Useful Life</td>
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<tr>
<td>SEI</td>
<td>Solid Electrolyte Interface</td>
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<tr>
<td>SOC</td>
<td>State of Charge</td>
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<td>SOH</td>
<td>State of Health</td>
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<tr>
<td>SVR</td>
<td>Support Vector Regression</td>
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Glossary

C-rate  A measure of the rate at which a battery is discharged relative to the manufacturer’s rated capacity in ampere-hours. It is also related to the discharge time. For example, if the battery’s rated capacity is 40 Ah, then 1C rate is 40 A and the battery is empty after a 1-hour discharge, 2C rate is 80 A and the battery is empty after a 0.5-hour discharge, and C/4 rate is 10 A and the battery is empty after a 4-hour discharge.

Calendar life  The length of time a battery can undergo some defined operation before failing to meet its specified end-of-life criteria.

Capacity  The capacity of a battery expresses the maximum available ampere-hours when a full battery is discharged at a certain C-rate until the cut-off voltage is reached.

Cycle  A sequence of a discharge followed by a charge, or a charge followed by a discharge under specified conditions.

Cycle life  The number of cycles, each to specified discharge and charge termination criteria under a specified charge and discharge regime, that a battery can undergo before failing to meet its specified end-of-life criteria.

Cycle depth  Cycle depth (ΔDOD or ΔSOC) describes the depth of a discharge-charge cycle. Cycle depth is usually expressed in percentage.

Degradation stress factor  Degradation stress factors are all the operation practices or circumstances that accelerate the degradation in battery and thus shorten the lifetime of the cell. Also known as the state of health stress factors.

Depth of discharge  The depth of discharge (DOD) is a measure of how much charge has been discharged from a full battery. It is usually expressed in percentage. Closely related to state of charge (DOD=1-SOC). ΔDOD is used to describe the depth of discharge-charge cycles.

Discharge rate  See C-rate.

Duty cycle  The operating parameters of a battery including factors such as charge and discharge rates, depth of discharge, cycle length and rest period length.

End of life  The stage at which a battery is not anymore capable to meet its performance criteria regarding capacity or power. There are two commonly used end-of-life criteria for a battery: capacity fade of 20%, resulting in 80% of the original capacity, and power fade of 20%, resulting in 80% of the original power and 25% increase in impedance.

Internal impedance  Opposition to the flow of an alternating current at a particular frequency at a specified state of charge and temperature.
<table>
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<tr>
<td>Internal resistance</td>
<td>Opposition to direct current flow in a battery. It is the sum of the ionic and electronic resistances of a battery.</td>
</tr>
<tr>
<td>Nominal operating voltage</td>
<td>The average voltage of a battery, as specified by the manufacturer, during discharging at a specified rate and temperature.</td>
</tr>
<tr>
<td>Open-circuit voltage</td>
<td>The equilibrium voltage of a battery at a specified state of charge and temperature when there is no current flowing.</td>
</tr>
<tr>
<td>Polarization</td>
<td>The voltage deviation from the equilibrium voltage under loading, i.e., when current is flowing.</td>
</tr>
<tr>
<td>Prosumer</td>
<td>An active energy consumer, who both consumes and produces electricity.</td>
</tr>
<tr>
<td>Self discharge</td>
<td>The process by which the available capacity of a battery decreases spontaneously due to undesirable chemical side reactions or short circuits within a cell.</td>
</tr>
<tr>
<td>State of charge</td>
<td>The state of charge is a measure of how much charge is left in a battery. It is a ratio of the present charge and the full charge, and it is usually expressed in percents.</td>
</tr>
<tr>
<td>State of health</td>
<td>The state of health is a measure of ageing. It can be defined for capacity fade and power fade. Typically a battery is considered to be at its end of life when the state of health has decreased to 80%.</td>
</tr>
<tr>
<td>Thermal runaway</td>
<td>Thermal runaway occurs in Li-ion batteries when the rate of internal heat generation caused by the exothermic reactions exceeds the rate at which the heat can be expelled. Eventually, the temperature rises rapidly and the battery catches fire and burns at a very high temperature. The fire may catch nearby cells, and eventually, the whole battery may burn down.</td>
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Executive summary

One of the main objectives in the INVADE project is to connect electric vehicles and batteries to the grid in order to provide flexibility to the electricity system. This document is part of the INVADE WP6, which focuses on energy storage technologies. This deliverable includes results from Task 6.2 Battery state of health and lifetime and Task 6.4 Battery safety and lifecycle management.

A simplified state of health test procedure for batteries in INVADE pilots and methodology for more advanced SOH estimation developed in Task 6.2 are presented. Battery lifetime data from literature is collected and analysed in Task 6.2, and the results are presented in this document. These results are utilized in a battery techno-economics model developed in Task 6.3.

Task 6.4 focuses on battery safety aspects and the use of batteries after the end of life is reached in the first application, such as electric vehicles. Battery end-of-life criteria, second life batteries and their applications, and recycling of lithium-ion batteries are also addressed in this deliverable.
1 Introduction

This document is INVADE project deliverable D6.3 *Simplified state of health diagnostics tool* compiled in Tasks 6.2 and 6.4. This report presents an overview on lithium-ion battery (LIB) state of health (SOH) and lifetime, and provides methodology to assess the SOH of the batteries utilized in the INVADE pilots. Also battery end of life (EOL) criteria and second life use are addressed in this report. Background for this document is presented in deliverable D6.1 *Storage system dimensioning and design tool* [1].

The monitoring of the internal state of lithium-ion batteries is a challenging task due to complex degradation mechanisms inside the cell and limited amount of information about the internal processes available. State of health is defined to describe the present condition of the battery compared to a fresh battery, and it is typically determined by battery capacity. Monitoring the battery state of health is important, because it greatly affects the remaining useful lifetime of the cell.

Battery degradation is accelerated by various operation practices or circumstances. In Chapter 4, a comprehensive set of battery lifetime data from literature is collected and analysed to study the cell degradation stress factors. The main results from this analysis are exploited in the battery techno-economic model presented in D6.2 *Battery techno-economics tool* [2].

A periodic diagnostic test for determining the actual capacity of the battery is introduced in Chapter 6. The periodic diagnostic test is designed to provide reliable data about the evolution of the battery capacity. This data can be used to monitor the ageing of the battery and in the development of more advanced SOH estimation method called incremental capacity analysis (ICA). The operational principle of the ICA method is presented in Chapter 6.

The EOL criteria of LIBs in electric vehicles is 80% of the nominal capacity (80% SOH). For stationary and portable applications, the EOL criteria is 60% of the nominal capacity. In Chapter 7, the most important international standards for EOL and their relevance in current applications criteria are introduced and discussed. Chapter 8 presents the second life use of LIBs in different applications, which also includes studies on the performance, degradation and safety of second life batteries.

This document will be followed by D6.4 *Advanced state of health diagnostics tool*, which is an updated version of this document. The study on the cell degradation factors will be expanded based on the needs in Task 6.3. The periodic diagnostics test will be updated
if necessary. The results obtained from the periodic diagnostics test on pilot batteries will be used to develop the ICA method introduced in this document. Also feasibility of other SOH estimation methods will be evaluated once the amount and type of data available from pilot batteries is known.

The updated version will include more detailed description of SOH diagnostic tools for second life batteries concerning especially performance and safety. In addition, life cycle analysis (LCA) of both first life and second life LIBs will be conducted in detail. These will be reported in D6.4 Advanced state of health diagnostics tools and the LCA will be combined later with the findings of Task 3.7 Life cycle analysis and D3.4 Draft life cycle analysis and D3.5 Final life cycle analysis.
2 Lithium-ion battery ageing

2.1 Degradation processes

The degradation process in lithium-ion battery is a complex combination of electrochemical and mechanical processes, which lead to capacity decrease and power fading. Most of these processes cannot be studied independently as they occur simultaneously at similar timescales and interact with each other. Ageing processes can be divided into two groups: ageing during use and ageing during storage. In other words: ageing related to cycle life and ageing related to calendar life.

Degradation processes take place in the battery’s electrolyte, especially at the interfaces with the anode and the cathode. The ageing mechanisms strongly depend on electrodes composition. The origins of the degradation mechanisms can be either chemical or mechanical, and the mechanisms can induce for example changes in the chemical composition of the electrolyte or loss of active materials at the electrodes. The key factor in Li-ion battery ageing is the formation of the solid electrolyte interphase (SEI) layer on graphite anode. [3]

The wide range of degradation mechanism can be clustered into three degradation modes: loss of lithium inventory, loss of active anode material and loss of active cathode material [4]. A list of degradation mechanism with their causes, effects and relations to the degradation modes is presented in Figure 1.

![Figure 1: Causes and effects of different degradation mechanisms in Li-ion batteries. [4]](image)
2.2 Safety

The degradation in lithium-ion batteries does not only weaken the properties of the battery and shorten its lifetime, but at the same time increases the safety risks during operation. Therefore comprehensive monitoring of the battery condition is essential.

Lithium-ion batteries have high energy density and hence are more prone to safety issues than other battery technologies with lower energy densities [5]. The main concern related to the safety of lithium-ion batteries is the risk of a thermal runaway [6] [7]. In thermal runaway the rate of battery internal heat generation exceeds the rate at which the heat can be removed by cooling, and the excess energy is released in the form of explosive fire. Thermal runaway is started if the cell internal temperature passes a critical level. The increase of the self-heating rate for Li-ion cell in high temperatures is demonstrated in Figure 2.

![Figure 2: The increase of self-heating rate as a function of temperature (NCA cell). [7]](image)

After certain temperature the self-heating rate grows uncontrollably and the self-feeding thermal runaway process is started. For different LIB chemistries the critical internal temperature level is different. The self-heating rate as a function of temperature for cells with most commonly used cathode materials is presented in Figure 3. Lithium cobalt oxide (LCO) and lithium nickel cobalt aluminium oxide (NCA) cells have the lowest runaway temperature whereas lithium iron phosphate (LFP) is the most stable cell chemistry.
Temperature increase inside the cell can be caused by many factors such as abuse of the cell, internal or external short circuit, degradation processes or external heating. Different causes of thermal runaway in lithium-ion battery are presented in Figure 4.

Li-ion batteries have limited safe operating window regarding voltage and temperature [9]. Thus following the operating limits set by the battery manufacturer is important.

One significant safety risk in Li-ion cells is the formation of dendrites on the carbon anode. Dendrites are fiber structures consisting of tiny lithium particles. The dendrites can cause internal short circuits, which cause a sudden rise in temperature and at worst
result in thermal runaway. [5] Dendrite growth is considered as a parasitic side reaction during charging and it is more prominent in lower temperatures [10].

Thermal runaway in one cell can be devastating if it spreads out to the adjacent cells and eventually destroys the whole battery pack [9]. Even though the failure probability for a single cell was low, the probability for failure in one of the cells in a large battery pack is considerably higher. Therefore it is important to try to hinder the cell-to-cell propagation in the battery system. [11]
3 Battery monitoring

3.1 Measurable battery parameters

Battery cell is a closed system with complex electrochemical processes inside. Only couple of parameters can be directly measured to determine the state of the battery. The major measured properties are the main circuit current and voltage. In addition to these the temperature in different locations outside the battery cell can be measured. The main circuit current and voltage are the key parameters in battery diagnostics and therefore their accurate measurement is important for exact battery state estimation.

In many applications multiple battery cells are assembled into battery modules and several modules form the battery pack. All in all, the battery pack may contain hundreds or thousands of battery cells. In some applications the voltage measurement from every single cell is available for diagnostic methods, but it is common that only for example maximum cell voltage, minimum cell voltage and average cell voltage are provided. In large battery packs the surface temperature of each battery cell is rarely measured. Instead the temperature sensors can be placed among the cells to provide temperature measurements at the module level.

3.2 State of health

A key indicator in battery health diagnostics is the SOH. Battery SOH describes the present condition of the battery compared to fresh battery. The SOH is presented in percentage, being 100% for the fresh cell. Battery SOH can be determined according to many factors of which battery energy capability and power capability are the most common. These are indicated by battery capacity fade and impedance increase, respectively. In principle any battery parameter which changes significantly with age can be used for determining the battery SOH. Multiple factors can be combined with different weights to form the overall SOH estimation.

However, the SOH is commonly determined based on just battery capacity, which simplifies the estimation process. In that case the SOH can be formulated as:

$$SOH = \frac{C_{\text{actual}}}{C_{\text{rated}}}$$

where $C_{\text{actual}}$ is the maximum available capacity in current state and $C_{\text{rated}}$ is the rated capacity of the fresh cell. The SOH decreases as the degradation processes in the cell
proceed. When the SOH is determined by capacity, batteries with SOH below 80% are commonly considered unusable. However, the proper limit depends on the requirements of the application. For industrial applications a SOH (capacity) limit of 60% is used in standard IEC 62620 [12].

3.3 Battery management system

Battery management system (BMS) is a system that controls the use of the battery pack and monitors the state of the cells. The BMS consists of both hardware and software required for determining battery states. The BMS has an essential role in the efficient and safe use of batteries. It ensures that the battery is used within the safe operating area. The tasks of a BMS can be divided into three categories [1]:

- safety-related tasks such as preventing overcurrent, overcharging, and overdischarging from happening by controlling the battery current directly or indirectly
- optimization-related tasks such as cell balancing and current limiting at low and high temperatures and SOCs, which maximize the performance and lifetime of the battery
- monitoring the state of the battery exploiting the directly measurable parameters such as cell voltage, current and surface temperature, and estimating SOC, SOH and remaining useful life of the battery based on the measured parameters.

Accurate estimation of the battery state is important so that the BMS can operate the battery pack as efficiently as possible and maximize the remaining useful life of the battery. The most favourable operating area within the application limits can be determined by identifying the SOH stress factors (Chapter 4) for the utilized battery.

In INVADE, the BMSs in pilot sites are provided by the battery system integrators. The commercial BMSs typically include some battery state estimation algorithms, but in addition to these the battery state estimation, especially SOH estimation, can be implemented on INVADE platform level. The battery data measured by BMS can be utilized in the development of SOH estimation methods in INVADE (Chapter 6).
4 Degradation stress factors

Degradation stress factors are all the operation practices or circumstances that accelerate the degradation in battery and thus shorten the lifetime of the cell. In the literature the degradation stress factors are sometimes referred to as the state of health stress factors. By identifying the stress factors the battery operating conditions and practices can be optimized within the application limits so that the degradation of the battery is minimized.

The degradation processes in lithium-ion batteries can be divided into two groups: degradation during cycling and degradation during storage. The degradation stress factors can be divided correspondingly: stress factors related to cycle ageing (Chapter 4.1) and stress factors related to calendar ageing (Chapter 4.2).

4.1 Cycle ageing

How the battery is operated affects greatly its degradation and remaining useful life (RUL) [5]. A list of degradation stress factors during cycling identified in the literature is presented in Table 1. Both the battery duty cycle and environmental conditions during operation affect the degradation rate. For different lithium ion battery chemistries the impact of different stress factors slightly varies, but the underlying degradation mechanisms are the same.

<table>
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<th>Stress factor</th>
<th>Effect</th>
<th>Chemistry*</th>
<th>Source</th>
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<tr>
<td>High ΔDOD</td>
<td>Capacity fade, power fade</td>
<td>LCO, LFP, NMC, NMC-LMO</td>
<td>[13] [14] [15] [16]</td>
</tr>
<tr>
<td>High average SOC</td>
<td>Capacity fade</td>
<td>LCO, NMC</td>
<td>[16] [17]</td>
</tr>
<tr>
<td>High charging current</td>
<td>Capacity fade, power fade</td>
<td>LCO, LFP, NMC</td>
<td>[5] [13] [18] [19] [20]</td>
</tr>
<tr>
<td>High discharging current</td>
<td>Capacity fade, power fade</td>
<td>LFP, NMC-LMO</td>
<td>[5] [13] [14]</td>
</tr>
<tr>
<td>High operating temperature</td>
<td>Capacity fade, power fade</td>
<td>LCO, LFP, NMC, NMC-LMO</td>
<td>[5] [13] [21] [22] [23] [14] [24]</td>
</tr>
<tr>
<td>Low operating temperature</td>
<td>Capacity fade, power fade</td>
<td>LFP, NMC, NMC-LMO</td>
<td>[5] [13] [14] [15]</td>
</tr>
</tbody>
</table>

*Chemistries with direct literature reference listed. Presumably applies for other LIB chemistries as well.

Table 1: List of degradation stress factors during cycling.
Low C-rates are employed in INVADE pilots, like in most grid-storage applications. These low C-rates cause only low heating of the battery. Moreover, the energy storage systems are typically installed in temperature-controlled facilities and thus variation in battery temperature is small. Considering the INVADE pilots, the most important cycle ageing stress factors are high cycle depth (i.e., ΔDOD) and high average SOC. For simplicity, only one degradation stress factor is included in the first version of battery techno-economics model developed in Task 6.3 [2], and it is selected to be the ΔDOD. The average SOC can be included as a second degradation stress factor in the advanced battery techno-economics model, if deemed necessary.

4.1.1 Cycle depth

The depth of the discharge–charge cycle is a significant degradation stress factor for Li-ion batteries during cycling. In this document the cycle depth is expressed with ΔDOD. Depending on the source the term DOD is used for two partially different meanings: a measure of how much charge has been discharged from a full battery or a measure describing the depth of the discharge-charge cycle. If the cycling is started from full battery, these two definitions mean the same. To avoid misunderstandings the term ΔDOD is used in this document to describe the depth of the discharge–charge cycle regardless of the initial SOC.

Example data sets from the literature studying the effect of ΔDOD to the cycle life for different Li-ion chemistries are presented in Figure 5. As can be seen from Figure 5, the increase in ΔDOD results in shorter cycle life. Even though the trend is similar in all data sets, there is variation between different chemistries and between single data sets among one chemistry.
To obtain comprehensive understanding on the effect of cycle depth on battery cycle life, data from different sources are combined in Figure 6. To be able to compare the results, the data sets are normalized based on the cycle life with 100% ΔDOD. In practice, the cycle life (Φ) at given ΔDOD is multiplied with ΔDOD to obtain the cycle life in equivalent full cycles (FCE), which is then divided by the cycle life with 100% ΔDOD (Φ|ΔDOD=100%)) to obtain the normalized cycle life:

$$\Phi_{\text{normalized}} = \frac{\Phi \times \Delta \text{DOD}}{\Phi_{\Delta \text{DOD}=100\%}}$$

As Figure 6 shows, the variation between different data sets is large. The overall trend is that the cycle life increases as the ΔDOD decreases, but there is high diversity in the magnitude of the increase and the shape of the curves. Different cycling conditions and C-rates might partially explain the diversity, but more data sets would be needed for more specific conclusions. For example in [25] Sarasketa-Zabala et al. have observed that LFP cells cycled under large and small ΔDODs both show the slowest degradation, which differs from the general understanding. The result is discussed in more detail in [25], but no clear explanation is found. This reflects the nature of the problem: a huge amount of lifetime test data with different operating practices and environmental conditions.
conditions would be needed to fully understand the effects of different stress factors on battery cycle life.

![Graph](image)

**Figure 6**: Normalized cycle life of different types of Li-ion cells as a function of ΔDOD. [26] [23] [14] [16] [27] [13] [28] [29]

An approximate model for the effect of ΔDOD to the cycle life can still be derived. The NMC-LMO data set from Wang et al. [14] is selected to be a basis for the model because it is close to average among all the data sets and a representative set as NMC-LMO and NMC batteries will be used in the pilots (Chapter 5.1). It is also close to the curves utilized by Peters et al. [29]. An exponential function is fit to the data (Figure 7). The obtained exponential model is:

\[
\Phi_{\text{normalized}} = 2.371e^{-2.438 \times \Delta DOD} + 0.7929
\]

where \(\Phi_{\text{normalized}}\) is the normalized cycle life of the battery and ΔDOD is the cycle depth. The exponential model and all the data sets are presented in Figure 8.
4.1.2 Average SOC

The average SOC during cycling has also significant effect on the cycle life of the cell. Data sets from literature studying the effect of average SOC to the cycle life for NMC and LCO cells are presented in Figure 9.
According to Figure 9, for LCO cells the lower average SOC (30%) results in longer cycle life, whereas for NMC cells the optimal average SOC would be at the middle (50%). It is good to notice that the LCO data set is obtained with 60% ΔDOD while the NMC cells are cycled with only 10% ΔDOD and this might affect the results. More comprehensive results are needed to draw further conclusions about the effect of average SOC on Li-ion battery cycle life.

4.1.3 Current

Both charging and discharging current rate affect the cycle life of Li-ion cells. Example data sets about the effects of charge and discharge rates on cycle life for different chemistries are presented in Figure 10.
Figure 10: Number of equivalent full cycles (FCE) at 80% SOH as a function of C-rate for: a) NMC cell discharge [15], b) NMC cell charge [15], c) NMC-LMO cell discharge [14], d) NMC-LMO cell charge [30], e) LFP cell discharge [13], f) LFP cell charge [13].

The data sets in Figure 10 are obtained with quite different ranges of C-rate, but the overall trend is that higher charge or discharge rate results in shorter cycle life. Only the data set for NMC discharge differs from that trend, but the applied discharge rates are relatively low. Anyhow, high charging rate has been found to accelerate the degradation in NMC cells, which can be seen in resistance increase [20] and capacity fade [13] [15]. Both high charge and high discharge rates have been found to decrease the cycle life of NMC-LMO cells [30] [14] and LFP cells [13]. The same trend applies for LCO cells at least regarding charging [19].
4.1.4 Temperature

The operating temperature affects greatly the cycle life of Li-ion batteries. The effect of operating temperature for different Li-ion chemistries is demonstrated in Figure 11. As can be seen from Figure 11, the cycle life of the cells decreases consistently as the temperature increases compared to the cycle life in room temperature. High operating temperature is reported to shorten the cycle life of LCO cells [21], LFP cells [13], NMC cells [23] [24] [15] and NMC-LMO cells [22] [14]. Low operating temperature also decreases the cycle life of Li-ion batteries. This accelerated degradation is observed at least for LFP cells [13], NMC cells [15] and NMC-LMO cells [14].

To obtain comprehensive understanding of the effect of temperature in cycle life, data from different sources needs to be combined. Various temperature-dependent cycle life data sets obtained from literature are presented in Figure 12. The data sets are normalized with respect to the cycle life in room temperature:

\[ \Phi_{\text{normalized}} = \frac{\Phi \times \Delta \text{DOD}}{\Phi_{\text{RT}}} \]

As can be seen from Figure 12, a large number of studies focusing on cycle life in high temperatures have been carried out, whereas studies on the cycle life in low temperatures are rare. Anyhow, both high and low temperature seem to shorten the cycle life. The most optimal operating temperature appears to be around room temperature.
Figure 11: Number of equivalent full cycles (FCE) at 80% SOH as a function of temperature for:

- a) NMC cell [31],
- b) NMC-LMO cell [14],
- c) LFP cell [32]

Figure 12: Normalized cycle life of different types of Li-ion cells as a function of temperature. Normalization is done with respect to the cycle life in room temperature (22-26°C). [23] [31] [24] [14] [13] [32] [33]
4.2 Calendar ageing

A list of major degradation stress factors during storage is presented in Table 2. The degradation rate during storage is mainly affected by storage temperature and the SOC level of the cell. These stress factors are discussed in more detail in Chapters 4.2.1 and 4.2.2.

<table>
<thead>
<tr>
<th>Stress factor</th>
<th>Effect</th>
<th>Chemistry*</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>High storage temperature</td>
<td>Capacity fade</td>
<td>LFP, NMC, NMC-LMO</td>
<td>[34] [31] [35]</td>
</tr>
<tr>
<td>High storage SOC</td>
<td>Capacity fade</td>
<td>LFP, NMC, NMC-LMO</td>
<td>[36] [17] [35]</td>
</tr>
</tbody>
</table>

*Chemistries with direct literature reference listed. Presumably applies for other LIB chemistries as well.

Table 2: List of degradation stress factors during storage.

4.2.1 Storage temperature

Storage temperature is a key factor considering the calendar ageing of lithium ion batteries [3]. The effect of storage temperature on calendar life for different Li-ion chemistries is presented in Figure 13. As Figure 13 shows, there is a clear decreasing trend in the calendar life of the cells as temperature increases. This trend has been detected at least for LFP cells [34] [18] [27], NMC cells [23] [26] and NMC-LMO cells [35]. However, it is good to notice that all these studies focus on calendar life at temperatures close to room temperature or higher, and the effect of low storage temperature is less studied.
Figure 13: Calculated storage time before reaching 80% SOH in different storage temperatures. 
a) NMC cell [31], b) NMC-LMO cell [35], c) LFP cell [34]

### 4.2.2 Storage SOC

Another important factor in calendar ageing is the battery SOC during storage. The calendar life as a function of storage SOC for different Li-ion battery chemistries is presented in Figure 14. For observed NMC, NMC-LMO and LFP cells the higher storage SOC seems to result in shorter calendar life. However, there is still significant variation in the magnitude of this phenomenon between the chemistries. To properly compare the chemistries, the results need to be normalized. In Figure 15 various data sets from literature are presented in normalized form:

\[
\Phi_{\text{normalized}} = \frac{\Phi \times \Delta \text{DOD}}{\Phi_{\text{SOC}=100\%}}
\]

According to Figure 15, the storage SOC affects the calendar life of NMC cells the most, whereas the effect on LFP cells is relatively much smaller. The effect of high storage SOC on cell degradation should be taken into account especially in backup applications, where batteries are stored in high SOC for long periods of time.
Figure 14: Calculated storage time before reaching 80% SOH for different storage SOC values. a) NMC cell [17], b) NMC-LMO cell [35], c) LFP cell [36]

Figure 15: Normalized calendar life of different types of Li-ion cells as a function of storage SOC. [17] [23] [37] [35] [18] [36] [34]
5 State of health estimation methods

Battery SOH estimation is a challenging task due to complex degradation processes inside the cell and the limited amount of information available. A large number of SOH estimation methods is presented in the literature [38] [39] [40]. However, many of the methods are still under research and not yet implemented in real applications. Various SOH estimation methods have some constraints or requirements related to the monitored battery system, exploited battery chemistry or available measurement data, which limits the utilization of the methods in a specific application such as the INVADE platform.

5.1 Characteristics of the INVADE pilot sites

Battery systems in the INVADE pilot sites are different and they are used in various applications. Brief descriptions of the pilots are given in [41]. The offered flexibility services are provided in [42], and some preliminary information about the storage requirements are gathered in [1]. Basic information from each pilot site is presented in Table 3.

<table>
<thead>
<tr>
<th>Pilot</th>
<th>Application</th>
<th>Total capacity</th>
<th>Battery type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulgaria*</td>
<td>Centralized battery for hotel and restaurant, connected to PVs.</td>
<td>200 kWh</td>
<td>NMC</td>
</tr>
<tr>
<td>Norway</td>
<td>30 residential batteries connected to PVs, EV chargers and smart heating systems.</td>
<td>30 x 4.2 kWh (residential)</td>
<td>LMO+NMC (Nissan 2nd life)</td>
</tr>
<tr>
<td>The Netherlands*</td>
<td>Centralized battery next to an office building. Local balancing: solar panels, windmills, EV charging.</td>
<td>138 kWh</td>
<td>NMC</td>
</tr>
<tr>
<td>Spain*</td>
<td>Backup battery storage system connected to the grid. Secures electricity supply for critical buildings. Can also be used to balance production and consumption in the area.</td>
<td>200 kWh (100 kWh for backup, 100 kWh for balancing)</td>
<td>LMO+NMC (Nissan 2nd life)</td>
</tr>
</tbody>
</table>

* Preliminary information, definitive battery system selection and purchase process not yet concluded.

*Table 3: Applications and characteristics of the batteries used in the INVADE pilot sites.*

The application where the battery system is implemented greatly affects the typical duty cycles of the battery. In all pilots the batteries are used to provide flexibility services: at grid level for distribution system operator (Spain) or at prosumer level (Bulgaria, Norway,
The Netherlands) [42]. At distribution system operator (DSO) level the batteries can be used to reduce peak loads or increase the load during peak generation of solar or wind power. For most grid-storage applications only low C-rates are employed. At prosumer level the battery can be used for time-of-use optimization, reducing the maximum load of the prosumer or self-balancing. Also in these applications the utilized C-rates will be fairly low.

In the Spanish pilot also backup services are provided. For the battery, it means that at the maximum 50-100% SOC can be used for flexibility services in a normal situation as half of the battery storage capacity is reserved for backup. In other pilots the available SOC range in daily use is not limited by the applications.

The pilot batteries are controlled with 15-min time resolution. This means that the minimum duration for discharging or charging phase will be 15 minutes and no faster cycling will be executed.

5.2 Review on potential state of health estimation methods

The most relevant SOH estimation methods for the INVADE platform are listed in Table 4. Also the advantages and the limitations of the methods are shortly presented. These methods are introduced in more detail in the following subchapters.

The most simple and straightforward method is to exploit coulomb counting for battery capacity estimation and thus for SOH estimation. An enhanced coulomb counting method is presented in Chapter 5.2.1. More complex but a common approach is to utilize various battery models for battery state estimation. A potential choice from the model based methods is Kalman filtering, which is often combined with an equivalent circuit model (ECM) of the battery. Kalman filtering is presented in more detail in Chapter 5.2.2.

The limitation with the model based methods is that the observed battery system has to be known in detail to determine the model parameters. No details about the battery are needed if a machine learning based method, neural networks, is used. Instead, a large amount of data is needed for training the neural network. The neural networks method is presented in Chapter 5.2.3. An alternative approach is presented in Chapter 5.2.4, where a method called incremental capacity analysis is described. In incremental capacity analysis battery charging or discharging data is analysed exploiting differential methods.
## 5.2.1 Enhanced coulomb counting

Coulomb counting, also known as Ampere-hour method, is the most common battery SOC estimation method [43]. The operation principle is straightforward: if the initial capacity stored in the battery is known, the capacity at each time point can be determined by measuring the amount of energy charged to and discharged from the battery. The transferred charge cannot be directly measured, but it is determined integrating the charge/discharge current over time. The SOC estimation in coulomb counting method can be represented as:

\[
SOC(t) = SOC(t_0) - \frac{1}{C_{\text{rated}}} \int_{t_0}^{t} \eta I(t) \, dt
\]

where \( t_0 \) is the initial time, \( C_{\text{rated}} \) the rated capacity of the battery, \( \eta \) the operating efficiency, and \( I(t) \) the current which is positive at discharge and negative at charge. The operating efficiency \( \eta \) is not constant, but it is affected for example by the charge/discharge current, temperature, and the SOH of the battery [43].

The disadvantage of the coulomb counting method in SOC estimation is the accumulation of error in the long term. A small error in the current measurement will eventually result in significant error in the integrated value. To avoid the problem, an enhanced coulomb counting method with dynamic re-calibration of the SOC estimation has been developed in [44]. In addition to the re-calibrated SOC estimate, the method generates an updated SOH estimate in the process.

The battery voltage and current are monitored in the enhanced coulomb counting method. A safe operating range regarding the cell voltage is specified by the battery

![Table 4: Advantages and limitations of potential SOH estimation methods.](image-url)

<table>
<thead>
<tr>
<th>Method</th>
<th>Advantages</th>
<th>Limitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enhanced coulomb counting</td>
<td>Simple implementation</td>
<td>Fully charged and fully exhausted states need to be reached occasionally</td>
</tr>
<tr>
<td></td>
<td>Low computational cost</td>
<td></td>
</tr>
<tr>
<td>Kalman filtering</td>
<td>Adaptive to inaccurate or noisy input values</td>
<td>ECM parameters needed</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Relatively high computational cost</td>
</tr>
<tr>
<td>Neural networks</td>
<td>Universality: no details about the battery needed</td>
<td>Extensive training data needed</td>
</tr>
<tr>
<td></td>
<td></td>
<td>High computational cost</td>
</tr>
<tr>
<td>Incremental capacity analysis</td>
<td>Only partial (dis)charging data during operation needed</td>
<td>Low C-rate constant current (dis)charging data needed</td>
</tr>
<tr>
<td></td>
<td>Relatively low computational cost</td>
<td>IC curve shape varies with cell chemistry</td>
</tr>
</tbody>
</table>
manufacturer. At maximum voltage, $V_{\text{max}}$, the battery is considered to be full and at minimum voltage, $V_{\text{min}}$, the battery is considered to be exhausted. The amount of charge charged to or discharged from the battery is calculated using the coulomb counting method:

$$Q = \int_{t_1}^{t_2} \eta I(t) \, dt$$

If the maximum voltage is reached, the SOC value is re-calibrated to 100%. Respectively, if the minimum voltage is reached, the SOC is set to 0%. [44]

The capacity charged or discharged between consecutive fully charged and fully exhausted states is determined with the coulomb counting method. The obtained actual capacity of the battery ($C_{\text{actual}}$ as denoted in Chapter 3.2) can be used for updating the SOH estimation. In other words, the SOH is updated every time the cell reaches fully charged state after fully exhausted state or vice versa. The battery operation between the states can include both charging and discharging. [44]

The enhanced coulomb counting method is computationally simple and can therefore be easily exploited in online SOC and SOH estimation. The biggest disadvantage is that fully charged and fully exhausted states need to be reached occasionally to re-calibrate the SOC estimate and update the SOH estimate. The accuracy of the method depends on the accuracy of the utilized operating efficiency $\eta$. Also the frequency of the re-calibration affects the estimation accuracy.

**Feasibility in INVADE**

Requires reaching fully charged and fully discharged battery states occasionally, which makes the method unsuitable for online SOH estimation considering the typical duty cycles in INVADE pilots. However, the coulomb counting method can be exploited for capacity estimation in the periodic diagnostic tests (Chapter 6.1).
5.2.2 Kalman filtering

Kalman filter is a mathematical tool that uses measurements containing noise and other inaccuracies to produce estimates of variables describing the state of the system. The strength of the Kalman filter is that it can combine multiple inaccurate and noisy measurements to generate an estimation, which is usually more precise than none of the individual measurements.

In battery SOH estimation Kalman filter is typically used in combination with the ECM of the cell. An example of an ECM is presented in Figure 16. To utilize the Kalman filter, the examined system needs to be represented in state space form. The state-space representation is a mathematical model of the system as a set of input, output and state variables related to each other by first-order differential equations. The state-space representation can be derived from the equivalent circuit model of the cell. Exploiting the Kalman filter, the internal parameters of the cell are estimated and the estimates corrected based on the measurements. [45]

![Diagram of battery equivalent circuit model](image)

*Figure 16: An equivalent circuit model of the battery. [45]*

The recursive Kalman filter algorithm consists of two steps: prediction step and correction step. The operation principle of the Kalman filter algorithm is presented in Figure 17. In the prediction step, also known as time update step, the state in the next time step is projected from the current state based on the system model. In the correction step, also known as measurement update step, the measurements and the state estimate generated in the prediction step are combined exploiting the Kalman gain, and the overall state estimate is formed. The Kalman gain is a matrix that indicates how much the measurements and the model estimate are weighted when generating the overall state estimate.
Normally, the use of Kalman filters is limited to linear systems. However, for non-linear systems, such as batteries, an extended Kalman filter (EKF) has been developed. EKF and its variations are widely used for battery internal parameter estimation as well as SOC and SOH estimation. [40] [45]

Kalman filtering can be used in SOH estimation of multiple cell battery systems. There are both offline and online implementations of the Kalman filtering method. However, Kalman filtering requires performing several matrix operations, which increases the computational complexity. The SOH estimation error with Kalman filtering is typically below 3%. [40] Kalman filter is often used in combination with another diagnostic method for SOH estimation.

**Feasibility in INVADE**

Requires specific parameters from the utilized battery system to build the battery model. That is not very practical in INVADE where the battery setup in each pilot is different, and thus requires different kind of battery model.
5.2.3 Neural networks

Neural networks, or more precisely artificial neural networks (ANN), are computational models based on the structure and functions of biological neural networks. Artificial neural networks represent an effective technique to approximate the input/output behavior of complex and nonlinear systems such as batteries [45]. ANNs consist of basic units called neurons, which are organized in layers: an input layer, one or more hidden layers and an output layer (Figure 18). Weighted interconnections between the neurons at different layers form the network. One special characteristic of neural networks is their ability to learn during a training phase without former knowledge about the observed system [45]. In the supervised training process the weights between the neurons are adjusted through learning algorithms so that a desired output is obtained with given input. The more complex the observed system is, the more training data is needed [46].

![Figure 18: Architecture of an artificial neuron and a multi-layered neural network. [46]](image)

A big advantage of ANNs for SOH estimation is its universality: it is not necessary to take into consideration all the details of the battery to obtain an accurate SOH estimate. However, the disadvantages of the neural network method are the high computational cost and need for large amount of training data to obtain an accurately working network. Neural networks can be used for online SOH estimation, but first the network needs to be trained offline with comprehensive training data. The SOH estimation error with ANN methods is usually at the maximum 2%. [39] [40]

Feasibility in INVADE

Requires large amount of training data before usage in SOH estimation, which limits the use of the method at least in the early stages of the INVADE pilots. However, later on if large amount of battery data is collected, neural networks is a potential tool for SOH estimation.
5.2.4 Incremental capacity analysis

Incremental capacity analysis is a method used for online SOH estimation [47] [48]. In ICA, the battery charged capacity is differentiated with respect to the battery terminal voltage to obtain the incremental capacity (IC) curve. An example of the IC curve for a LFP cell after different amount of cycles is presented in Figure 19. In the IC curve, there are characteristic peaks depending on the battery chemistry. A clear change in the shape of the peaks can be detected as the cell ages [49]. The changes in the IC curve reflect the changes in the behaviour and electrochemical properties of the cell. Therefore the cell degradation can be monitored by observing the evolution of the IC curve.

![Figure 19: Incremental capacity curves of a LFP cell after different amount of cycles.](image)

ICA can be implemented for both charging and discharging data. Voltage and capacity data during constant current charging/discharging is needed for forming the IC curve. The major constraint related to the required data is that the C-rate of the charging or discharging needs to be around C/2 or lower to obtain proper IC curves. The lower the C-rate, the better the results. An example of incremental capacity curves obtained from discharging and charging of a NMC cell with C/5 is presented in Figure 20.
Figure 20: The evolution of incremental capacity curves obtained from discharging and charging of a NMC cell. [50]

Depending on the battery chemistry, the characteristic peaks are at different voltage levels. Observing the transformation of the characteristic peaks is enough to monitor the degradation of the cell. Therefore only partial charging or discharging data is needed to cover the voltage levels where the characteristic peaks are formed.

Feasibility in INVADE

A potential SOH estimation method for batteries in the INVADE pilots. Requires partial charging or discharging data with low C-rate. If proper charging or discharging phases are included in typical duty cycles of the batteries, the method can be implemented without any interruption to normal operation.
6 State of health estimation in INVADE

6.1 Necessary battery parameters from pilots

Typically the BMSs in battery systems collect a variety of data during the operation of the battery: direct measurement data from the battery and possibly some battery state estimations obtained with BMS’s own algorithms. Many of these parameters can be utilized in the development of SOH estimation methods for the batteries in INVADE pilots.

A list of required battery parameters as well as useful parameters from the BMSs in INVADE pilots is presented in Table 5. Most important parameters for SOH estimation are battery current and voltage, which are needed in high frequency (e.g. 1 Hz) to be able to implement advanced SOH estimation methods such as the ICA method. Another important parameter is battery temperature, which is a central cell degradation stress factor. The other parameters in the list are examples of useful parameters for the development of SOH estimation methods. The actual available parameters from pilot batteries are not yet known as the definitive battery system selection for pilots has not yet been concluded.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Importance</th>
</tr>
</thead>
<tbody>
<tr>
<td>battery current</td>
<td>required</td>
</tr>
<tr>
<td>battery voltage</td>
<td>required</td>
</tr>
<tr>
<td>battery temperature</td>
<td>required</td>
</tr>
<tr>
<td>battery power</td>
<td>if available</td>
</tr>
<tr>
<td>cell voltages</td>
<td>if available</td>
</tr>
<tr>
<td>minimum cell voltage</td>
<td>if available</td>
</tr>
<tr>
<td>maximum cell voltage</td>
<td>if available</td>
</tr>
<tr>
<td>SOC (BMS)</td>
<td>if available</td>
</tr>
<tr>
<td>SOH (BMS)</td>
<td>if available</td>
</tr>
<tr>
<td>ampere-hour count</td>
<td>if available</td>
</tr>
<tr>
<td>cycle count</td>
<td>if available</td>
</tr>
<tr>
<td>battery kWh charged</td>
<td>if available</td>
</tr>
<tr>
<td>battery kWh discharged</td>
<td>if available</td>
</tr>
</tbody>
</table>

Table 5: List of necessary and useful battery parameters form pilots for SOH estimation.
6.2 Periodic diagnostic test

To develop more advanced SOH estimation methods for the batteries in INVADE pilot sites, reliable reference data about the condition of the battery is needed. As the SOH is determined based on battery capacity, reliable data about the evolution of the battery capacity is required.

The capacity data can be obtained with a periodic diagnostic test, where the battery is discharged from full state to empty state and the discharged charge is calculated with the coulomb counting method (Chapter 5.2.1). The amount of charge discharged from the battery reveals the total available battery capacity. The periodic diagnostic test is designed for two purposes:

1. To produce a reliable estimate of the total available battery capacity. This estimate can be used directly for monitoring the degradation of the battery, and as a reference in the development of other SOH estimation methods.

2. To provide proper charging data for the development of the incremental capacity analysis method (Chapter 5.2.4). At the later stage, the goal is to replace this with charging data from normal use.

The proposed periodic diagnostic test is defined below:

**Three steps diagnostic run (9-10 hours)**

1. Charging to full state
   - Present SOC → 100% SOC
   - Charge current 1C or lower
   - 60 min rest time after charging
2. Discharging from full state to empty
   - 100% SOC → 0% SOC
   - Constant C/3 discharge current
   - 60 min rest time after discharging
3. Charging from empty to full state
   - 0% SOC → 100% SOC
   - Constant C/3 charge current
   - 60 min rest time after charging

The proposed diagnostic test simply consists of consecutive full discharge and full charge phases with low C-rate. If C/3 charge and discharge rate is used, the total
duration of the diagnostic test would be 9-10 hours, depending on the initial SOC. By increasing the utilized C-rate, the duration of the test could be slightly decreased, but the data would be less suitable for the incremental capacity analysis method.

To obtain proper data about the degradation of the battery, the diagnostic test needs to be carried out regularly. The suitable testing frequency depends on the duty cycle of the battery. If the battery is operated with high ΔDOD, high C-rate or frequent charge-discharge cycles, the degradation will be faster. Diagnostic test approximately every 200 full charge–discharge cycles would be sufficient to monitor the cell degradation precisely. However, the number of cycles is not the only factor in cell degradation as the degradation occurs also during idle time. For batteries with dense cycling the suitable test frequency would be every 2–3 weeks and for batteries with lesser usage every 4–6 weeks, depending on the constraints set by the application.

6.3 Degradation monitoring with incremental capacity analysis

ICA can be used for monitoring the battery SOH as presented in Chapter 5.2.4. The ICA method requires voltage and charge capacity data from low current charging, which can be obtained from the C/3 charging step in the periodic diagnostic test (Chapter 6.1). The IC curve can be produced with three steps:

**Producing the IC curve**

1. Voltage and charge capacity data from diagnostic test
2. Fitting a smooth curve to the voltage - capacity data
3. Numerical derivation of the obtained curve

In principle, the charge capacity \( Q \) is just differentiated with respect to the battery terminal voltage \( V \) to obtain the IC curve \( dQ/dV \). However, if the differentiation is implemented directly on the measurement data, the obtained IC curve typically contains a large amount of noise. Therefore various curve fitting methods are used to fit a smooth curve to the voltage - capacity data. In [47], a method called support vector regression (SVR) is found to be accurate and robust method for curve fitting.

An example of the SVR fit with data from an LFP cell is presented in Figure 21. An IC curve obtained directly from the data and an IC curve obtained from the smooth SVR fit curve are compared in Figure 22. Even though the example measurement data presented in Figure 21 is already very smooth, the IC curve obtained directly from it is
rather noisy (Figure 22). The noise level in the IC curve increases dramatically if more noisy measurement data is utilized without any curve fitting method.

![LFP cell](image)

*Figure 21: A smooth SVR fit curve on C/3 charging data of an LFP cell.*

![LFP cell](image)

*Figure 22: IC curves obtained from C/3 charging data of an LFP cell. An IC curve obtained directly from data and an IC curve obtained from smooth SVR fit curve are compared to demonstrate the effect of noise.*

As can be seen in Figure 22, there are clear characteristic peaks in the IC curve. The characteristic peaks in the IC curves for different lithium ion battery chemistries are
presented in Figure 23. The shape of these peaks changes as the cell ages. Typically the peaks become lower and the area of the peaks decreases.

The height and area of the characteristic peaks at different steps of battery ageing can be compared to corresponding battery capacity results from the periodic diagnostic test to study the correlation between them. In [47] a clear correlation between the height of the characteristic peak and cell capacity for LFP cells is detected. If a clear correlation between the height or area of the characteristic peaks and the cell capacity is found, it can be used to estimate the capacity of the cell during operation based on the incremental capacity curve.
7 Battery end of life criteria

When the battery can no longer meet its performance requirements, it has reached its EOL, and has to be removed from the application. Being able to properly define this point is important, because it affects both the system performance and safety. If the battery has degraded too much, it may not be able to respond to the requirements set by the application where the battery is used. Also the risk of a critical failure happening in the battery pack will increase when the battery degrades. In this section, the EOL criteria found from literature and standards are presented, and possible improvements for determining the EOL are discussed. The main focus is on the electric vehicle batteries, as in the INVADE project, one goal is to use second hand electric vehicle (EV) batteries for second life. However, also other battery applications are mentioned briefly.

For EV batteries a typical definition for EOL is when 70-80% of the original energy capacity is remaining [51] [52]. This originates to a standard established by the US Advanced Battery Consortium (USABC) in 1996. According to this standard the EOL has been reached when either the net delivered capacity of a cell, module or battery is less than 80% of its rated capacity or the peak power capability is less than 80% of the rated power at 80% DOD [53].

Another standard for electric road vehicle batteries is IEC 62660. There a cycle life test is specified for both battery electric vehicles (BEV) and hybrid electric vehicles (HEV). The battery is cycled with a given charge and discharge cycle. For BEVs, capacity, dynamic discharge capacity and power are measured before the cycling, and after each step has been completed. The dynamic discharge capacity is defined by the time integrated value of charge and discharge current. During the test a fully charged battery is discharged repeatedly by a dynamic discharge profile specified in the standard. The power is measured at 50% SOC. The test will be terminated if the test sequence is repeated 6 times, or if any of capacity, dynamic discharge capacity or power is decreased to less than 80% of its initial value. For HEVs only capacity and power are measured. Also the test cycle is different for HEVs than BEVs. The test is terminated when any of the performance indicators has decreased to less than 80% of its initial value or if the test sequence has been repeated 6 times, or if the cell temperature exceeds a chosen upper limit [54]. For HEVs the test is terminated when either capacity or power has decreased to 80% of the initial value or if the test is repeated for 6 months. [54]

For industrial applications including stationary storages, the EOL is defined by the IEC 62620 standard. The EOL has been reached when the remaining capacity of the battery
has dropped to 60% of the rated capacity [55]. A similar definition is given in the IEC 61960 standard for portable applications [56]. According to [55], the battery should last at least 500 cycles before EOL. The definitions for EOL that were found from standards are listed also in Table 6.

However regarding EVs, in most cases in literature, the effect of the power limit is neglected, and only the capacity is considered. This is usually sufficient, because the EV batteries provide maximum charging and discharging power limits that are much higher than the power limits of a traction motor. Therefore it is the motor that limits the acceleration and regenerative braking rather than the battery [51].

<table>
<thead>
<tr>
<th>Standard</th>
<th>Application</th>
<th>End-of-life criteria</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>USABC Electric vehicle battery test procedures manual, revision 2</strong></td>
<td>Electric vehicles</td>
<td>When either (1) the remaining capacity is less than 80% of the rated capacity or (2) the peak power capability is less than 80% of the rated power at 80% DOD</td>
</tr>
<tr>
<td>IEC 62660</td>
<td>Electric road vehicles including battery electric vehicles (BEV) and hybrid electric vehicles (HEV)</td>
<td>If capacity, dynamic discharge capacity or power has decreased to less than 80% of the initial value (BEV) If capacity or power has decreased to less than 80% of the initial value (HEV)</td>
</tr>
<tr>
<td>IEC 62620</td>
<td>Stationary applications: telecom, uninterruptible power supplies (UPS), electrical energy storage system, utility switching, emergency power and similar applications Motive applications: fork-lift truck, golf cart, AGV, railway, marine</td>
<td>When the remaining capacity is less than 60% of the rated capacity</td>
</tr>
<tr>
<td>IEC 61960</td>
<td>Portable applications</td>
<td>When remaining capacity is less than 60% of the rated capacity</td>
</tr>
</tbody>
</table>

Table 6: Lithium ion battery end-of-life definitions found from standards.
It has not been very well explained where the 80% EOL criteria comes from. The ability of the battery to meet the daily travel needs of the driver is influenced by both energy capacity and peak power ability of the battery. Especially in BEVs it is important that the battery capacity does not decrease too much, because that would reduce the range of the EV. The power affects the vehicle performance such as acceleration, gradeability and regenerative braking ability [51].

However, some papers have stated that the EOL could be set lower 80%. The SOC is typically determined as a remaining charge relative to the rated capacity. However, this method does not take into account the capacity fade which happens when the battery degrades. According to [57] the modern BMS systems can take into account this, and thus such control strategies can be implemented that prevent the battery using extreme SOCs which would accelerate the degradation. Therefore [57] suggests that when thinking of the EOL, at least in plug-in hybrid vehicles (PHEVs) the replacement of the battery would be sensible only when a significant improvement in efficiency or performance could be achieved. For example, if the savings of fuel costs would be greater than the replacement cost of the battery. In BEVs, however, the battery performance directly affects the performance of a vehicle, and therefore the EOL criteria have to be different.

In [51] the ability of the battery to meet the driving needs of the US drivers has been investigated using a simulation tool, which is provided input data from a survey of 24-h vehicle usage of US drivers. Unexpected trips that the driver may need to do have been taken into account as a worst case scenario. Battery SOC profiles are computed for each travel itinerary in the database at each level of energy capacity fade down to 30% remaining storage capacity. The vehicle is assumed to meet the driver’s needs if the battery does not run out of charge. The results show that the batteries are able to meet the driver’s needs well below the 70–80% limit [51]. Also a degraded battery can continue to meet the daily travel needs of drivers who have only shorter range trips, which means that the degraded EV batteries could be used in cars rated for short range travel [51].
8 Second life use of batteries

The development of second life applications of used LIBs is still a very new market because most of the EV batteries are still in the early phase of their life [58] [59]. It will still take years before these batteries reach their EOL and are ready for either recycling or being prepared for second life applications. Due to the lack of supply of batteries for refurbishment the cost is still high. The major technical challenge is to combine different battery modules and chemistries into second life applications because each battery pack is unique with its own history. The ability to determine the SOH and remaining capacity of used battery modules is still the key challenge. One could say that there are two main objectives for the future:

1) Reduce the remanufacturing costs of second life batteries; and

2) Establishment of a battery management system to store all data at individual battery cell level in EVs.

The EOL criteria for LIBs of EVs is 80% of the initial capacity as described in the previous chapter. However, the used batteries from EVs still retain a considerable amount of capacity that could be utilized in other applications, mostly stationary applications, because for stationary applications the EOL criteria is 60% of the initial capacity. At the EOL the battery pack will be removed from the vehicle and is available for secondary use or recycling. Many life cycle assessments (LCAs) and economic studies have calculated environmental and economic benefits if LIBs are reused after the first life in the car instead of direct recycling [60] [61] [62]. However, in addition to environmental advantages, the use of second life batteries has to be a viable business case in order to become a significant factor in energy markets. This might need initial government support and new inter-industry partnerships as studied by Jiao and Evans [63].

8.1 Potential use cases

Four main use cases for second life batteries [59] introduced in this chapter are:

- Residential applications
- Energy utility
- Commercial and industrial applications
- Off grid applications
Residential applications: A study by Madlener and Kirmas [61] developed a model of a household with an integrated photo voltaic (PV) storage system and evaluated its economic viability. They concluded that households can profit from using second life batteries with PV under certain conditions even without financial incentives. However, positive effects on the electricity grid are achievable only with grid-optimized operating strategies. This means that in order to relieve the electricity grid the battery has to be charged at the time when the power generation peak is highest, which require precise forecasts. An economic study by Heymans et al. [64] evaluated the use of second life batteries for energy storage and peak shaving for residential users in Canada. This use case shifts in power demand from peak times to off-peak times and thus reduces strains on the electric grid. They found out that the approach has marginal economic feasibility without government intervention and moderate economic feasibility with intervention. Thus, in this case, there is a need for the government to offer incentives for adopting second life battery technology.

Energy utility: Batteries being a multifunctional technology they can be used for power, transmission and distribution. Batteries can secure capacity and supply, and protect the grid from stress events by balancing consumption and generation during the day. In addition to performance characteristics, battery pack systems can be built up fast almost at any location and they are scalable. Renewable energies such as solar and wind are intermittent and batteries can increase the system performance, thus growing their
feasibility for energy markets significantly. For example, in the case of a PV power station a sudden decreases in generation due to clouds can be countered with a battery pack system, hence batteries enable renewable energies to be used on demand. Thus, the use of second life batteries for renewable energy applications is quite probable. [59] [65] [66] [67]

**Off grid applications:** Building electrical mini grids to rural areas offers a potential use case for second life batteries [59] [67] [68] [69] [70]. In developing countries and rural areas, second life batteries—combined with PV and possibly other renewable energy sources—can become the storage hubs for community-scale grids. This could also offer affordable and reliable energy for people living in regions where grid connection is underserved. Australia is a good example where renewable off grid power applications could be the most cost-efficient way to provide electricity for communities that are not even in remote areas but far enough from the main grid [71]. In addition, telecom towers, remote industry and other decentralize infrastructure could discard or at least minimise the use of diesel generators for continuous energy demands. However, there are still challenges due to high cost of second life LIBs as well as reaching the rural areas and convincing customers to switch to greener solutions.

**Commercial and industrial applications:** Conventional power plants and industry benefit from batteries due to their versatility. Batteries can reduce grid and peak demand charges, and provide back-up power. In industry, power sensitive production equipment can be protected against frequency hops and voltage pikes thus increasing production efficiency and reliability. By using batteries, commercial players can also participate in the ancillary services market, thus creating new revenue sources. In this case, predictive analytics is needed in order to anticipate and control real time energy demand. [59]

### 8.2 Performance, degradation and safety

Battery history and SOH are considerable determinants in the life cycle performance of second life battery packs. Hence, the capability of a battery to be used in a second life application depends on the degradation of the battery during the first life use. This is illustrated in Figure 25 where modelled battery range highly depends on the phase 1 and phase 2 use profiles (slow vs. fast).
Figure 25. Modeled range of battery degradation profiles, each composed of an initial Phase 1 degradation, followed by a Phase inflection point and terminal Phase 2 degradation. Each of these parameters is defined by a base case value, as well as by values describing faster or slower degradation. [3]

The study by Waldmann et al. [72] showed that the safety of aged cells (commercial 3.25-Ah 18650-type cells with graphite anodes and NCA cathodes) is not simply a function of the SOH. They noticed that safety strongly relates to the main ageing mechanism and to the history of operating parameters during the lifetime of the cell. The ageing history is mostly affected by the operating parameters, that is stress factors, such as charging C-rate, temperature, and SOC range, which have a large influence on the main ageing mechanism. For example, the main ageing mechanisms for cycling at a rate of C/2 at 0 °C is homogeneous Li plating. When cycled at C/2 at 45 °C SEI growth and adhesion loss of the anode active material are the main ageing mechanisms. Therefore, it is a priority to collect data from the first life use in an EV and understand the degradation mechanisms of the battery components in order to determine its potential for re-use. In addition, an adequate choice of the battery chemistry combined with optimal working condition would be beneficial in order to improve the battery lifetime, minimize the risks for the end users, and therefore reduce the environmental cost. For example, a LCA of second life batteries from EVs by Casals et al. [73] studied two cases: an energy arbitrage and an island second life system. In the case of an energy arbitrage (100% ΔDOD), the energy storage suffers a nominal capacity drop from 80% to 60% within only 8 years. In the case of an island second life battery system, the lifetime reaches 20 years due to more favourable conditions of use, that is, the battery will rarely be fully charged.
or discharged (small $\Delta$DOD). This difference in lifetime is mainly due to the cycling conditions and more particularly the $\Delta$DOD of the charge/discharge cycles.

Figure 26. Scheme of the transition from first life to second life testing. [74]

Technical capabilities of second life batteries was studied by Marinez-Laserna et al. [74] as illustrated in Figure 26. First life batteries with different ageing profiles were mixed into homogeneous and heterogeneous stacks or kept at cell-level. They took two different second life applications: a residential demand response management service and a power smoothing application for a grid-scale PV plant. SOH of the second life batteries was evaluated and second life ageing performance was analysed. One finding was that the reused cells which did not reach the “ageing knee” during their first life of use performed very well. In contrast, the reused cells which did reach the “ageing knee” during their first life use exhibited a very fast degradation. This again proves that the history from the first life use plays an important role for the suitability of the second life batteries. If the EV batteries have gone through fast first life battery ageing, reusing them in highly demanding second life applications does not seem feasible. It should be still further studied if:

1) Cells after a less demanding first life use are capable of showing good performance in demanding second life; and

2) Batteries aged with severe first life conditions are able to provide a stable second life performance in less demanding applications.
8.3 Recycling lithium-ion batteries

When the EV battery reaches its EOL, it has to be decided what to do with the used battery. The possible choices are recycling, reuse (second life) or remanufacturing the battery. So far, recycling of LIBs is focused on reclaiming the metals, not organic materials [52]. The main economic driver is the recovery of cobalt, as it is the most rare and expensive material. However, recycling has several downsides. Recycling of LIBs is more difficult than for other battery types for several reasons [75]. The technology is still evolving, and there are several different chemistries in use. Li-ion batteries utilize wide variety of materials in a form of powder coated onto metal foil which makes the separation difficult. One LIB pack most likely has 100 or more cells which also makes the separation complicated. The EV batteries can have various sizes and shapes, which makes it difficult to remove the batteries from vehicles. Because around 80% of the capacity is remaining when the battery is removed from the vehicle, the bulk materials in the battery are active. Recycling of such batteries will reduce active bulks in the batteries to material constituents and leads to total loss of remaining 80% of available capacity. Moreover, EV batteries require high-grade materials for their components, which means that both purity and microstructure of the recycled materials must be proven to be suitable before reuse in EV batteries [52]. One issue is also that there are no regulations regarding recycling Li-ion batteries [75]. Therefore new processes, reagents and operations to recover substantial pure materials at low cost are required before recycling of LIBs is feasible [52], [75].

Recycling processes have been described in [76] and [77]. These include pyrometallurgical recycling (smelting), intermediate recycling process and direct recycling. In pyrometallurgical recycling, the battery modules are fed to a high temperature shaft furnace along with a slag forming agent. The electrolyte and plastics burn, and the valuable metals are reduced to an alloy of copper, cobalt, nickel and iron, which can be recovered by leaching. This recycling process is commercial, and it is economical for cathode material which contain cobalt or nickel [76].

In intermediate recycling, the batteries are put in a hammer-mill to reduce size, and a shaker table is used to separate plastics and metals. The metals can be separated and sent for recycling. This process is commercially used in Canada, but it is also economical only when the cathode contains cobalt or nickel [76] [77].

Direct recycling recovers battery materials for reinsertion into the battery supply chain with little or no additional processing. The electrolyte is extracted using CO₂ and it can
be recycled, if decided to be economical. Then the cells go through size-reduction, e.g. pulverization, and the cell components are separated using differences in electrical conductivity, density or other properties. The advantage is that almost all battery components can be recovered, and the method is suitable for different cathode materials. However, there is a risk that the recovered materials will not perform as well as original materials, which could lower the battery lifetime and performance, making manufacturers reluctant to use direct recycling [76].

In the future, it is possible, that best battery chemistries and design are found, which would make it possible to make more uniform LIBs, and thus simplify recycling [76]. User-friendly labeling of batteries would help appropriate routing. Recycling could be taken into account in the battery design phase too. Having strict industry standards for battery recycling could ensure the quality of the recycled materials [76].

As can be seen, there are still quite many issues regarding the recycling of LIBs. Therefore, when the batteries meet their EOL in EVs, other options such as second use should be considered.

### 8.4 Life cycle assessment and economic feasibility

Life cycle assessment and economic feasibility studies are important addition for reasoning the potential of second life battery applications. LCA can reveal the most environmentally burdening steps of LIB applications and propose actions. However, previously performed LCA studies on LIBs included varying system boundaries and functional units making their unbiased comparison difficult. In a report by Romare and Dahllöf [78], the need for standard designs of the batteries to enable transparent comparison of LCAs and collaboration with battery and cell manufacturers in order to get more detailed technical descriptions of each production step is highlighted.

In INVADE project, Task 3.7 *Life cycle analysis* will analyse the entire life cycle of all devices involved in the INVADE systems. However, due to the overarching nature of the study, the environmental impact of LIBs will not be studied in details. For this reason, Task 6.4 *Battery safety and lifecycle management* will perform the LCA of first and second life LIBs which will be reported in D6.4 *Advanced state of health diagnostics tool* and combined later with the findings of Task 3.7 and D3.4 *Draft life cycle analysis* and D3.5 *Final life cycle analysis*. 
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