

Supplementary Material 1

1 Motivation and Aim of the Study

The growing number of Battery Electric Vehicles (EV) is presenting recycling companies with a number of future-oriented decisions. In particular, the handling and recycling of lithium-ion battery systems (LIB) from end-of-life vehicles at the end of their service life is proving to be a central challenge. However, used batteries also offer economic opportunities. This is particularly true in an environment in which manufacturers of key components of electromobility are trying to secure their access to raw materials of strategic economic importance in the long term. The materials and elements from Lithium-Ion batteries of EVs can be a valuable secondary source for this. In addition to material recycling, alternative recycling strategies such as reuse/repair, repair and remanufacturing offer the possibility of extending the economically usable life of a battery system and at the same time relieving the growing demand for resources for the production of energy storage systems.

In order to adequately evaluate this development, we would like to define the main factors influencing the number and timing of future battery returns along with the selection and benefits of an appropriate closed-loop circulation strategy and estimate their development.

According to current studies and market analyses, NMC-based cell chemistry will continue to dominate in the short to medium term. However, different NMC variants already exist today: NMC 111, NMC 532, NMC 622, NMC 811 [1–3]. In order to be able to estimate the potential of battery system Closed-loop circulation, it is necessary to assess the possible development of the market shares of the different NMC derivatives.

2 Theoretical Background

The State-of-Health (SoH) indicates the condition of the battery system and characterizes its ability to meet the specified performance specifications. The specification of SoH is related to the performance values of a new battery system. External factors such as operating temperature, overcharge/discharge, high charge/discharge rates and improper charge/discharge cycles can have a negative influence on the SoH. The following is a brief explanation of relevant factors:

- *Cycle Number:* The cycle number is one of the most important performance parameters of a battery cell, as it gives an indication of the expected working lifetime of the cell. A common threshold, and consequently a termination criterion for service life of battery, is often the number of cycles a cell can go through before its capacity drops to 80% of its original specific capacity or the internal resistance has doubled. Each charge/discharge cycle and the associated reactions of the active material are accompanied by a slow deterioration in performance that is hardly noticeable to the user. The progressive degradation (caused by calendar aging, or by the actual use of the battery cell) is caused by a variety of mechanisms. Generally, a distinction is made between the two loss mechanisms (1) loss of active material and (2) loss of lithium. These in turn are caused by a variety of reaction mechanisms.
- *Operation at high and low State of Charge (SoC):* The SoC is the level of charge of a battery relative to its capacity. The SoC value is calculated as the percentage of the releasable capacity

relative to the nominal capacity. This means that a battery is fully charged at 100% SoC and fully discharged at 0% SoC. Battery management systems (BMS) in EVs allows the driver to use about 80% of the total battery capacity (from 90 to 10% SoC) [4]. However, using the battery in the SoC range between 70-20 %, allows the cycle number of a battery to be extended to 6000 cycles (Table 1) [5].

Table 1S: SoC Range and its effect on a battery cycle life ^[5]

SoC from...to...%	Cycle Number	SoC from...to...%	Cycle Number
100 – 0	500	80 – 0	3000
100 – 10	500	80 – 10	3000
100 – 20	1000	80 – 20	3500
90 – 0	1500	70 – 0	5000
90 – 10	1500	70 – 10	5500
90 – 20	2000	70 – 20	6000

- *Current density (C-Rate):* The capacity of a battery depends on the discharge/charge current. C-rate is a measure of the speed at which a battery is discharged in relation to its maximum capacity. For example, 1C is defined as the power that would reach the total capacity of a battery over one hour and it means for a 50-kWh battery, 1C corresponds to 50 kW. The capacity of a battery depends on the discharge/charge current. For example, a recommended maximum continuous discharge rate for NCA (LiNiCoAlO₂) battery is 1C and for LMO (LiMn₂O₄) based battery it is possible to go up to 10C for an optimum life [6]. In addition, lately a charging speed has become more important than achieving a maximum capacity. Fast charging requires high current densities. A high current density means a high diffusion speed of the lithium ions through the electrolyte. Very high current during the charge process results in lithium plating, which is an undesirable local deposit of metallic lithium on the surface of the anode [7, 8]. Even under ideal charging conditions, the pores of the electrodes can clog after a few charging cycles due to the continuous growth of the SEI layer (solid electrolyte interface). These deposits lead to premature aging and thus to malfunction of the batteries.
- *High Temperatures:* Temperatures above 30 degrees Celsius result in a high load for LIBs. Battery systems lose performance more quickly if the ambient temperature exceeds certain limits.

3 Survey Evaluation

In this section, we provide a prognosis through assessment of the expert opinions. For calculation purposes, we use the average of numerical answers received from experts and combine them with the relevant influencing factors and development potential, in order to calculate the realistic scenarios.

Numerical questions:

- Assuming a technologically progressive development, how long do you estimate the lifetime of an EV battery system in 2030?
 - Average Answers: 12.2 years
- Please indicate the market shares of the individual NMC cathodes in 2030 under the assumption of a progressive scenario, which should reflect the trend of steadily increasing nickel shares.

Table 2S: NMC Market share today and in 2030

NMC- type	Market Share	
	2019 (current) ^[30]	2030 (Experts)
NMC 111	45%	9 %
NMC 532	30%	14 %
NMC 622	20%	30 %
NMC 811	5%	47 %
Total	100%	100%

3.1 Realistic Scenario 1: Battery Life

“Hypotheses” from experts and state-of-the art battery life are considered as 2 components and a formulation is configured in order to get a realistic number:

$$\text{Realistic Development Potential (RDP) \%} = (\text{impact factor} * \text{score}) / (\text{total impact}) \quad \text{Eq. 1}$$

$$\text{RDP \%} = \frac{(4*0.5) + (3*0.5) + (2*1) + (1*1)}{4+3+2+1} = 0.65 \rightarrow 65\%$$

Difference between the progressive lifetime (average of the experts' answers) and the lifetime according to the state of the art results in a progressive increase of battery lifetime which is equal to 4.2 years. According to calculated % RDP, a lifetime based on a realistic scenario can be calculated as:

$$\text{Realistic expected lifetime} = 8 + (4.2 * 0.65) = 10.73 \sim \mathbf{11 \text{ years.}}$$

3.2 Realistic Scenario 2: Future Battery Chemistry

In this part, in order to calculate a realistic scenario for a future battery chemistry, a formulation has been created for the effects of each influencing factors (IF).

$$I_F = ((\text{total votes for "not agree"} \times \text{score}) + (\text{total votes for "agree"} \times \text{score})) * \text{weighting} \quad \text{Eq. 2}$$

Where scores are equal to (from Table 1 in the main manuscript):

Agree = 1 point, disagree = 0 point; Favors Ni-rich cathodes - "YES"

Agree = 0 point, disagree = 1 point; Favors Ni-rich cathodes - "NO" **Fehler! Verweisquelle konnte nicht gefunden werden.**

RDP % for battery chemistry then can be calculated through:

$$RDP \% = \frac{Sum I_F}{Total Weighing} \quad Eq.3$$

According to formulas the realistic development ratio was calculated as **62.6%**.

4 References

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