



Pyrometallurgical recycling of different lithium-ion battery cell systems: Economic and technical analysis

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ABSTRACT

The global trend towards electromobility raises questions about the treatment of lithium-ion batteries from battery-electric vehicles at the end-of-life stage. The paper examines two pyrometallurgical recycling routes (a direct and a multi-step process) for different lithium-ion battery cell compositions (NMC333/C, NMC811/C, LFP/C, NMCLMO/C) from a techno-economic perspective. Based on lifecycle inventories per recycling process and battery type, the profitability of these two recycling processes is investigated by conducting a total cost of ownership analysis for typified pyrometallurgical recycling plants on a pre-industrial scale. The results reveal that the cell chemistry will have a major impact on the profitability of recycling. In particular, it appears to be difficult to operate recycling profitably under current conditions for low-cobalt and low-nickel battery types. A sensitivity analysis shows different levers and their respective limitations for increasing the process profitability of recycling different lithium-ion battery cell systems.

1. Introduction

Mitigation of the global climate change is a major challenge and requires strong commitments to climate and environmental protection at national and international levels. To reach the European net zero goal, the mobility sector is required to reduce emissions by 90% by 2050 (European Commission, 2019). Battery electric vehicles (BEVs) are a key component of this strategy (European Commission, 2020) and their market penetration is expected to increase significantly by 2030 (IEA - International Energy Agency, 2020).

With increasing electrification of the mobility sector, research on lithium-ion batteries (LIBs) is gaining importance. Production costs (König et al., 2021; Vekić, 2020), storage capacity, and longevity (Hoyer, 2015), but also the recyclability (Ketterer et al., 2010) of LIBs, which strongly depends on their respective composition (Yu et al., 2021), will be decisive factors for ecological and economic sustainability of electromobility in the long term. Recycling becomes indispensable to counteract resource scarcity (Bongartz et al., 2021; Bobba et al., 2020).

Though the LIB market for BEVs already contains a variety of different battery compositions and expects further battery cell compositions to emerge in the near future (Vekić, 2020) which makes it difficult to predict in detail future material volumes gained from end-of-life batteries (Hoyer, 2015).

Market diversity currently not only concerns battery cell compositions, but also recycling process types (Blömeke et al., 2022; Chen et al., 2019; Harper et al., 2019). The latter have a strong influence on recycling efficiency for different materials (Werner et al., 2020), but also and especially on the environmental impacts of recycling (Mrozik et al., 2021). Hydro- and pyrometallurgical processes currently dominate the global industrial LIB recycling landscape for the recovery of valuable metals (Werner et al., 2020; Harper et al., 2019). Within those two general streams of technology, different types and combinations of processes exist (Werner et al., 2020). The present work focusses on pyrometallurgical industrial LIB recycling processes, as smelting of spent batteries is yet more established due to higher throughput capabilities as well as higher robustness to variations in material composition (Chen

Abbreviations: BEV, Battery electric vehicle; EG8, Pay group 8 according to collective agreement (Germany); LFP, Lithium iron phosphate; LIB, Lithium-ion battery; LMO, Lithium manganese oxide; NMC, Lithium nickel manganese cobalt oxide; NMCLMO, Mix of LMO and NMC532; NPV, Net present value; OLS, Ordinary least squares; TCO, Total cost of ownership; WACC, Weighted average cost of capital.

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et al., 2019; Harper et al., 2019).

For the environmental perspective of LIB recycling, a variety of research on different process routes already exists (Iturrondobeitia et al., 2022; Jiang et al., 2022; Rajaeifar et al., 2021; Rey et al., 2021). However, little is currently known about recycling efficiency and output or profitability of LIB recycling processes on a pre-industrial and industrial scale, especially for pyrometallurgical recycling routes (Heimes et al., 2022; Werner et al., 2020; Hoyer, 2015). Concurrently, Germany having already introduced a legal obligation for battery manufacturers to take back spent batteries free of charge (Deutscher Bundestag, 2009), profitability of LIB recycling becomes relevant for a growing number of parties and actors in research and the economy (Wrålsen et al., 2021).

The aim of this paper is to contribute to the existing research gap between lab and pre-industrial scale pyrometallurgical LIB recycling by using pre-industrial data to estimate realistic data and to enable a techno-economic comparison of treating different LIB cell chemistries. More precisely, the paper addresses the following research questions.

- (1) Are the two main options for pyrometallurgical processes profitable for the recycling of LIBs which have different cell compositions?
- (2) How can the profitability of these recycling processes be improved and what parameters need to change for this improvement to happen in the future?

This paper is structured as follows: Section 2 gives a brief overview of materials and methods. After explaining the two pyrometallurgical recycling processes selected here as examples, possible process variations are presented in a morphological box. Furthermore, the total cost of ownership (TCO) method used for the economic analysis is described. Section 3 contains a technical part elucidating the approach used for establishing lifecycle inventories for four individual battery types and two pyrometallurgical recycling processes, and an economic part introducing the TCO model as well as the underlying price assumptions. In Section 4, the results are presented and discussed. Finally, Section 5 provides a conclusion as well as an outlook that outlines possible topics for subsequent research.

2. Material and methods

The techno-economic analysis focuses on three different NMC batteries, specifically NMC333, NMC811 and a mix of lithium manganese oxide (LMO) and NMC532 (NMCLMO), as well as LFP batteries. All battery systems considered contain graphite as anode material. The focus was chosen due to the different cobalt and nickel content in order to investigate the influence of cathode materials used for LIBs on the profitability of pyrometallurgical recycling. This idealized consideration of individual battery types instead of current or future battery market mixes allows the highlighting of individual effects of changing cathode materials in LIBs. Especially because it is not yet known, which battery cell chemistry will prevail in future, the focus on individual battery types can be helpful.

Currently, many companies are involved in battery recycling, either hydro- or pyrometallurgical (Werner et al., 2020; Heimes et al., 2022). Table 1 shows selected lithium-ion battery industrial recycling processes and the respective process units.

The diversity of the battery cell systems and their complex structure and composition have led to numerous options of recycling routes. The process chains can include mechanical and thermal pretreatment steps as well as pyro- and hydrometallurgical methods to recover valuable materials from the batteries. In hydrometallurgical process steps metals are extracted and refined in aqueous media. Usually the following steps are involved: leaching, purification, and subsequent recovery of target metals. Hydrometallurgical treatments enable high purity material recovery with lower energy consumption compared to smelting processes. The high temperatures in pyrometallurgical processes lead to increased

Table 1

Selected overview of lithium-ion battery industrial recycling processes based on (Brückner et al., 2020; Pinegar and Smith, 2019; Velázquez-Martínez et al., 2019).

Company	Process units
Accurec Recycling GmbH	Therm. Treatment → Mech. Processing → Pyrometallurgy
AkkuSer Oy	Mech. Processing → Pyro- or hydrometallurgy
Batrec Industrie AG	Mech. Processing → Therm. Treatment → Hydrometallurgy
Düsenfeld GmbH	Mech. Processing → Hydrometallurgy
Glencore	(Mech. Processing →) Pyrometallurgy
Inmetco	(Mech. Processing →) Pyrometallurgy
Nickelhütte Aue	(Mech. Processing →) Pyrometallurgy → Hydrometallurgy
Recupyl	Mech. Processing → Hydrometallurgy
Retriev Technologies Inc.	Mech. Processing → Hydrometallurgy
Umicore	(Mech. Processing →) Pyrometallurgy

energy requirements. Usually, in industrial pyrometallurgical battery recycling not only pure LIB battery material is processed but a mix of various materials containing nickel, cobalt and copper in different amounts and compounds are fed into a furnace. However, to enable distinct analyses the present work considers separate treatment of different LIB types.

In this chapter, two pyrometallurgical recycling routes for end-of-life LIBs that are relevant in this work are summarized from a technical point of view. In addition, the TCO method is explained.

2.1. Pyrometallurgical recycling processes

Two different pyrometallurgical process chains, in the following referred to as direct and multi-step smelting process, will be explained in more detail and subsequently analyzed from an economic perspective.

In the direct smelting process, energy storage systems are first disassembled to battery module level or battery cell level (Abdelbaky et al., 2021; Tytgat, 2013). Afterwards, mechanically untreated battery modules or cells, reducing agents, and slag additives are fed into a shaft furnace. Three different temperature zones are used in this furnace. In the first of these, the battery modules or cells are preheated in order to minimize the risk of possible explosions. In the following zone – the so-called “pyrolysis zone” – the separator and electrolyte are volatilized at a temperature of up to 700 °C. The energy gained from the combustion of organic materials such as electrolyte and separator helps to reduce the overall energy demand. In the final zone, the batteries are smelted and reduced (Dunn et al., 2012). The flowchart in Fig. 1 depicts this process. Several different products and byproducts are thereby generated (Velázquez-Martínez et al., 2019).

In contrast to the direct smelting process, the second pyrometallurgical recycling route involves additional mechanical pretreatment of the battery modules and cells. The recycling path also starts with the discharging and dismantling of energy storage systems to battery module or cell level, as shown in Fig. 2. The modules or cells are then pyrolyzed to volatilize the organic components. Subsequently, the thermally treated modules are mechanically (e.g., comminution and several sieving steps) separated into different fractions, primarily to obtain active mass as a fine fraction. For smelting in the electric arc furnace, the fine fraction has to be pelletized prior to being fed into the furnace (Sommerfeld et al., 2020).

A metal alloy, Li-containing slag and flue dust can be obtained as products (Sommerfeld et al., 2020). Both recycling routes described above lead to comparable product phases during the smelting process. Nevertheless, the distribution of metals and the mass balances differ due to prior mechanical processing in the multi-step recovery route.

When treating LIBs pyrometallurgically to recover valuable metals, different characteristics of both batteries and processes have to be

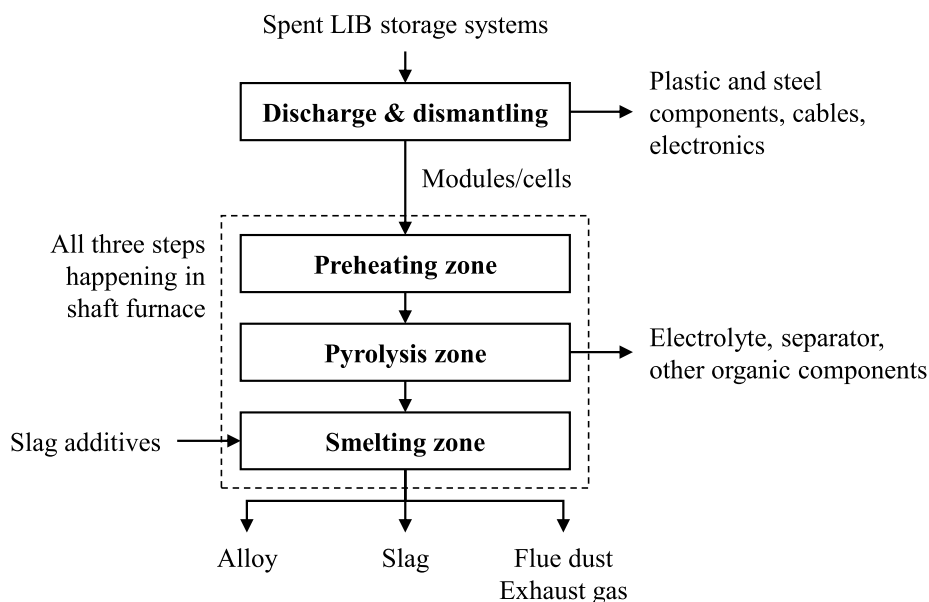


Fig. 1. Schematic illustration of the direct smelting process, adapted from (Chen et al., 2019; Velázquez-Martínez et al., 2019; Arnberger et al., 2018).

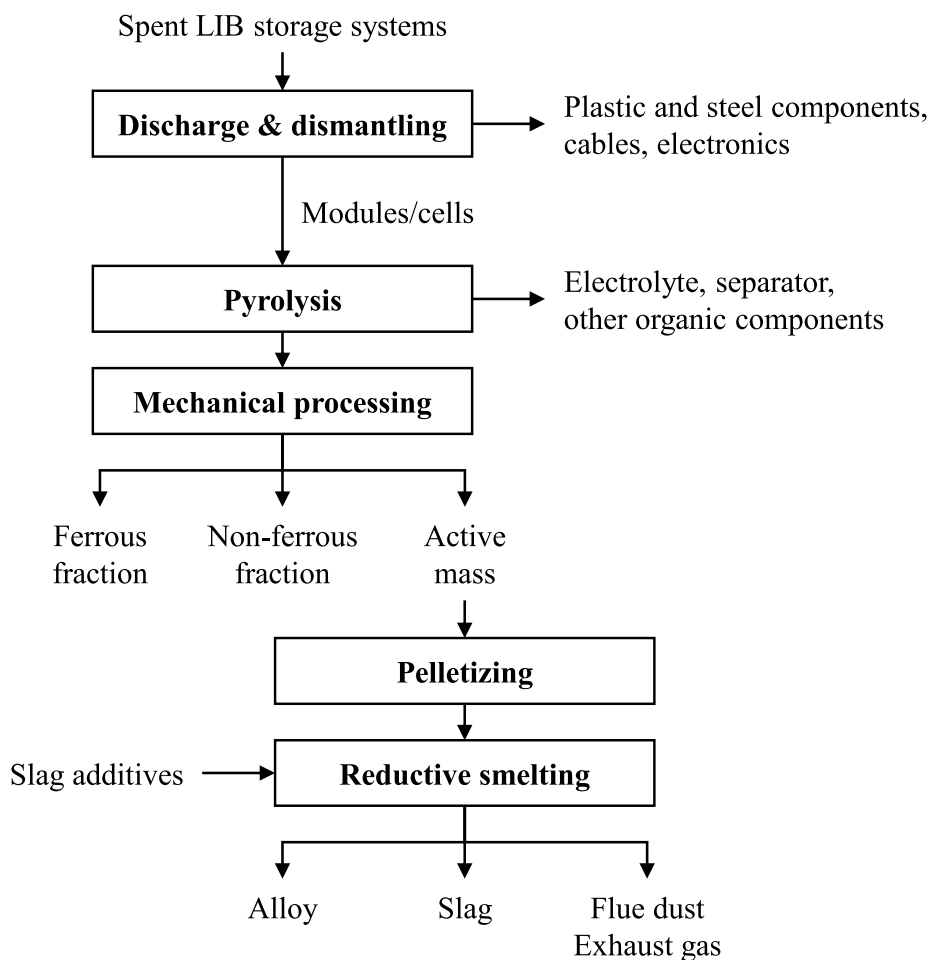


Fig. 2. Schematic illustration of the multi-step smelting process, adapted from (Sommerfeld et al., 2020; Chen et al., 2019; Arnberger et al., 2018).

considered. To provide an overview, Table 2 presents in a morphological box different key characteristics and a variety of possible characteristic traits of the pyrometallurgical treatment options shown in Figs. 1 and 2. From the wide variety of possible battery cell chemistries, four

representative types were selected, which are considered as examples in this paper.

Especially regarding the cell chemistry of LIBs, there are further different specifications that are not mentioned in this morphological

Table 2

Morphological box of relevant characteristics and their specifications with respect to pyrometallurgical LIB recycling routes (own table).

LIB	Casing	Steel	Aluminum	Plastic	
	Battery cell chemistries considered	NMC333	LFP	NMCLMO	NMC811
Pyrometallurgical recycling process	Manual dismantling	None	Module level	Cell level	
	Pretreatment	None	Thermal	Mechanical	Thermal/mechanical
	Reducing agent	Needed		Not needed	
	Melting unit	Shaft furnace	Electric arc furnace	Bath furnace	
	Slag additives	With		Without	
	Further processing: slag	Landfill	Pyro-metallurgical	Hydro-metallurgical	Construction material
	Further processing: metal phase	Marketable (intermediate) product		Pyro-metallurgical	Hydro-metallurgical
	Further processing: flue dust	None	Pyro-metallurgical	Hydro-metallurgical	Circulating

Variants to be considered in this paper are highlighted in bold.

box. The characteristics highlighted in bold are considered as variants in the following study.

2.2. Total cost of ownership and net present value

In order to examine the profitability of the pyrometallurgical LIB recycling processes, a TCO analysis is performed for typified recycling plants. The TCO method allows a comparison of the total costs associated with owning a product or conducting a process (Ellram and Siferd, 1993). TCO models include the purchase prices of the products under consideration as investments and operating costs including the costs of maintenance, disposal, and all other components associated with the object under consideration over its entire useful lifetime (Ferrin and Plank, 2002; Degraeve et al., 2000; Degraeve and Roodhooft, 1999; Ellram, 1994, 1995). Just as each object has its individual cost structure, there is not one general TCO model but rather specific models corresponding to the objects under consideration with a unique set of associated activities and cost drivers needing to be designed (Bhutta and Huq, 2002; Ferrin and Plank, 2002; Degraeve and Roodhooft, 1999). To obtain robust results, detailed price data for all cost drivers as well as the inclusion of uncertainty are essential (Geissdörfer et al., 2009; Degraeve et al., 2000; Ellram, 1995). The work presented in the current article therefore includes various scenarios as well as sensitivity analyses to address existing uncertainties. The underlying TCO model, the structure of which is explained in more detail in Section 3.2, includes not only costs but also revenues to account for all cash-effective variables associated with the recycling routes considered. For a better understanding, the economic calculations are based on contribution margins for each process step.

The annual TCO is derived from the net present value (NPV) using the total planning horizon T and the discount rate i , as shown in Eq. (1).

$$TCO = NPV \frac{(1+i)^T \cdot i}{(1+i)^T - 1} \quad (1)$$

The NPV represents the sum of all discounted cash flows of an investment or a decision and therefore comprises all cash-effective variables associated with the object under consideration (Hirshleifer, 1958).

Moreover, the NPV method is established for TCO models (e.g., Kappner et al., 2019). Here, these are the initial investment I , operating costs $C_{Operating,t}$, and revenues R_t , as shown in Eq. (2), where T is the total planning horizon, and i represents the discount rate. $C_{Operating,t}$ and R_t are calculated on an annual basis.

$$NPV = -I + \sum_{t=1}^T \frac{R_t - C_{Operating,t}}{(1+i)^t} \quad (2)$$

The TCO model in this article does not include resale values of investments, as all assets are supposed to be used throughout their entire lifetime. Investments occur only in the initial period since it is assumed that buildings and plants will not need to be replaced during the period under consideration. Despite the importance of environmental considerations in LIB recycling (Mrozik et al., 2021), the TCO model presented focuses exclusively on internal costs without including potential external effects as costs.

3. Theory and calculation

Before starting the economic evaluation using a customized TCO model, the underlying technical analysis resulting in lifecycle inventories for both recycling routes is presented. Section 3.2 dealing with the TCO model gives a comprehensive overview of all components taken into account as well as the respective data used within the calculations. Since material prices are particularly important input factors for the TCO analysis presented here, the calculation scheme for material prices and the different price development scenarios are explained in detail in Section 3.3.

3.1. Lifecycle inventories

The derivation of material flows starts with energy storage systems (LIB packs), which are then treated along the two routes presented. Table 3 provides an overview of the inputs and outputs of the process flow diagrams.

The emissions of the cleaned exhaust gases are not considered in the following technical or economic analysis. There are two reasons for this:

Table 3

Overview of input and output mass- and energy flows along the process steps of multi-step and direct recycling route (own table).

Process step	Process type	Input	Output
Discharge	No differentiation	No external input needed, energy requirements for discharge are covered by energy recovered	
Disassembly	No differentiation	Energy storage systems, energy	Modules, scrap: steel, copper, aluminum, plastics, electronics
Thermal pretreatment ^a	Direct process	No thermal pretreatment	
	Multi-step process	Modules, nitrogen, energy	Thermally treated modules, exhaust gas, pyrolysis oil
Mechanical treatment ^a	Direct process	No mechanical treatment	
	Multi-step process	Thermally treated modules, energy	Active mass, flue dust, scrap: steel, copper, aluminum
Pyrometallurgy ^a	Direct process	Modules, slag additives, energy	Metal alloy, slag, exhaust gas, flue dust
	Multi-step process	Active mass, slag additives, energy	Metal alloy, slag, exhaust gas, flue dust

^a Including exhaust gas cleaning.

First, reliable information on these emissions is not available. Second, results are unlikely to change since similar emissions for all alternatives considered can be expected.

LIBs are available in different versions. Battery packs consist of modules which in turn consist of cells. The cells can be cylindrical, prismatic or pouch cells, and consist of various components, of which the cathode material is of particular interest (Chen et al., 2019). An overview of different weight compositions of the LIB packs, modules and cells can be found in Fig. 3.

Corresponding calculations are based on the BatPaC 5.0 model developed by Knehr et al. (2022). The element distribution along the recycling routes was derived using a modeling tool by Friedrich and Peters (2019) with own adjustments to the distribution in thermal treatment and mechanical processing steps. Distribution coefficients of the smelting process were calculated using FactSage 8.0. Further information concerning mass balance and assumptions regarding energy consumption of the processes are shown in Appendix A.

3.2. TCO model

The TCO model presented in this paper aims to holistically evaluate the profitability of pyrometallurgical recycling of different battery types. To enable such a comparison, separate TCO calculations are conducted per battery type for both process types. All calculations rely on the same TCO model, shown in Fig. 4, which comprises the revenues generated at the different stages of the recycling process as well as the initial investment and operating costs. To maximize transparency, the TCO calculation structure follows the structure of the recycling process.

Initial investments include investments for machines and technical equipment required for the recycling routes as well as the respective buildings needed. Validated data for the investments is derived from previous studies and offers that assume a multi-step recycling process on a pre-industrial scale. Accordingly, upfront investments refer to pyrometallurgical facilities with a maximum capacity of 375 kg material per hour in total including active mass or battery modules respectively as well as slag additives. Upstream process steps are dimensioned accordingly to achieve maximum utilization of the pyrometallurgy. As pyrometallurgical recycling capacity is kept the same for both recycling routes but pretreatment steps differ, total recycling plant capacities vary between process routes. Hence, total capacity of the recycling plants amounts to 829.74 kg LIB packs per hour for the multi-step process and 539.37 kg LIB packs per hour for the direct recycling process. Based on

practical experience, a lifetime of 25 years is assumed for the recycling plant, with no major replacements for machinery and equipment during this time. Only investments directly related to the pyrometallurgical recycling process, and their preparatory steps were considered. As a result, for the multi-step route, an initial investment of approximately 29 million € is assumed, of which about 73% is related to machinery and technical equipment. Pyrometallurgy is the most capital-intensive process step, accounting for 53% of the total initial investment.

The initial investment for the technical equipment includes general equipment such as cranes, workshop equipment and furniture, as well as safety equipment including exhaust gas treatment and gas detectors, as far as they are required in each process step. In addition, the main components of the additional technical equipment required for each section are listed in Table 4.

Due to the lack of detailed data, the initial investment for the multi-step process was adjusted for the process steps that are not part of the direct recycling process (mechanical preparation and pretreatment) to obtain upfront investments for a direct process pyrometallurgical recycling plant on a pre-industrial scale, i.e., the investments for the pyrometallurgical process step are comparable for both processes considered. Total upfront investment amounts to approximately 18.5 million €. A detailed overview of the distribution of upfront investments per process step for the two pyrometallurgical recycling routes is presented in Table 5.

Operating cost contains fixed and variable costs. Fixed costs comprise workforce as well as insurance and maintenance costs. For the latter, flat-rate shares of the respective investment amounts are taken into consideration for calculating annual costs. Annual insurance costs are calculated as a flat-rate percentage of total investments per process step. Data for maintenance and insurance cost flat-rate shares are based on Bärwaldt and Kwade (2012) and were adjusted according to expert input. The respective parameters applied for calculation of maintenance and insurance costs are listed in Table 6.

Workforce costs include wages of the employees working in the recycling plant as well as the respective social contributions to be paid by the employer. The number of employees per process step that is assumed for the calculation of workforce costs results from internal discussion, taking into account the personnel requirements assumed by Bärwaldt and Kwade (2012) for their disassembly plant. Personnel requirements include a buffer for compensating absences as well as general area and overhead personnel. Analogously to Bärwaldt and Kwade (2012), a distinction is made between simple tasks and more complex tasks to account for different skill levels required. For the allocation of workers performing the two different task types, an allocation key is applied to the total employees per process step, which is derived from to the allocation of task types in a large-size LIB disassembly plant (Bärwaldt and Kwade, 2012). Thus, 93% of the workers are assumed to perform simple tasks, while 7% of the employees are assigned to a complex task level. Bärwaldt and Kwade (2012) apply separate cost schemes per task complexity. The respective cost data were adopted and brought to current levels by applying the real gross salary growth rates in Germany from 2012 to 2020 (Statista, 2021). The final cost data were validated by comparing the range between simple and complex tasks to IG Metall (EG8) salary information currently applied (IG Metall, 2018). Hence, we use realistic and sector-specific personnel costs that also include employer's contribution to social insurance. Analogously to Bärwaldt and Kwade (2012), a premium of 15% is applied to the total salaries for the remuneration of the management level. For the direct process, only personnel costs for discharge, disassembly, pyrometallurgy, and general areas are considered. Furthermore, disassembly personnel is adjusted due to the inferior capacity of the direct recycling plant in terms of LIB packs per hour. Table 7 summarizes the calculation scheme for determining the annual workforce costs.

Variable costs as well as operating revenues depend on annual run-time of the recycling plant and annual productivity. The recycling plants are assumed to run 250 days per year in three-shift operation, for a total

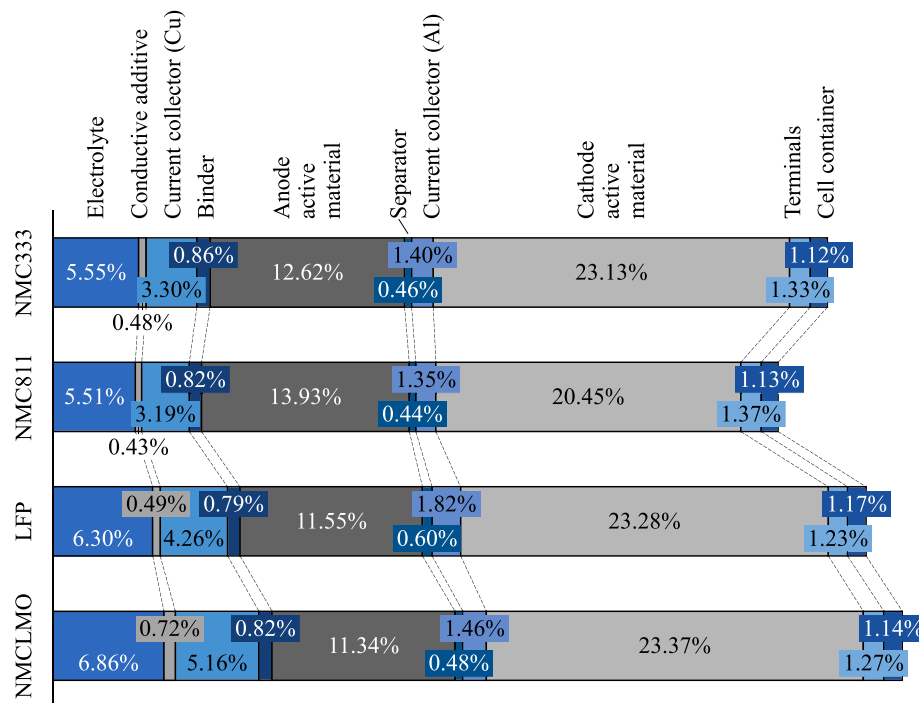


Fig. 3. Battery pack weight compositions at cell level (NMC333, NMC811, LFP and NMCLMO), configured with the BatPaC 5.0 model (own figure).

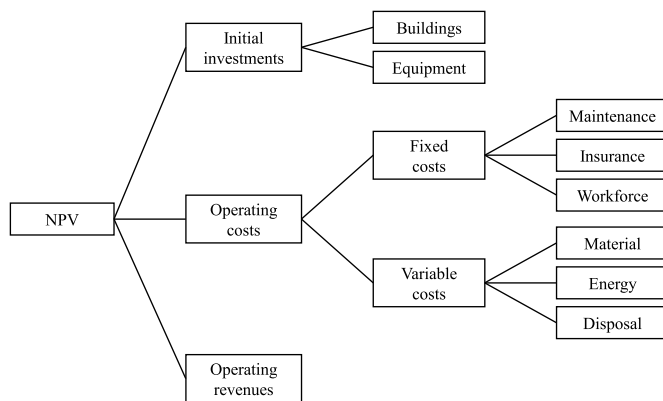


Fig. 4. TCO model (own diagram).

Table 4

Main components of technical equipment (own table).

Process step	Technical equipment (main components)
Discharge	Discharge station
Disassembly	Disassembly systems; flooding system for damaged batteries
Thermal pretreatment	Rotary kiln (heating system, infrastructure for gases/electricity)
Mechanical treatment	Machines for size reduction and screening; auxiliary equipment for processing
Pyrometallurgy	Furnace with spare capacity (incl. Infrastructure for gases/electricity); auxiliary equipment for processing, ladles, etc.; slag post-treatment

of 6000 h per year. Considering possible interruptions due to maintenance or technical problems, it is assumed that 95% of the total runtime is productive, resulting in 5700 productive hours per year.

Variable costs include material, energy, and disposal costs, while operating revenues are generated when marketable output is produced in the process steps during the recycling process. Electricity is assumed to be consumed during the entire runtime, whereas consumption of

other process inputs and waste generation is assumed to occur only during the productive runtime. Material, waste, and energy inflows and outflows for the four battery types during the different recycling process steps are derived from the lifecycle inventories in Section 3.1 and multiplied by the corresponding market prices which are derived in Section 3.3. Table 8 provides a simplified overview of variable costs and operating revenues generated during the different process steps.

After calculating the annual cash flows, these are discounted to their NPV by applying a discount rate of 6.4%, which corresponds to the average weighted average cost of capital (WACC) for the materials industry (PwC - PricewaterhouseCoopers GmbH, 2022).

3.3. Material prices and price development scenarios

Where possible, commodity exchange prices were used for materials and process auxiliaries as well as for marketable output fractions. Otherwise, prices were derived from available statistics and literature. Electricity costs were taken from utility data. Where data were not available, prices were estimated based on own experience. The value of mixed output fractions, such as alloys, is the weighted sum of the prices of the contained pure materials corresponding to the associated mass fractions. A price markdown of 50% which emerged from a discussion with experts was applied to account for the loss in value of the material mix compared with the individual pure materials.

To reflect different future price developments, four price scenarios were defined. Starting from the same initial price for the first year of the period under consideration p_i , the price development scenarios are characterized by different annual price growth rates $g_{p,i}$ for each material. The four scenarios are a fixed one with no price change over time, a lower case one, a base case one, and an upper case one. The formation of different price growth scenarios is reasonable against the background of uncertain and volatile future commodity prices.

The price growth rate for each scenario consists of a calculated part and a default part, both of which add up to the corresponding annual price growth rate. The calculated part was derived from respective past data sets, provided that data availability allows the calculation of an annual price growth rate. The reference to past data on material prices

Table 5

Initial investments for multi-step and direct process (own table).

	Multi-step process initial investment			Direct process initial investment		
	Buildings	Machines	Total	Buildings	Machines	Total
Discharge	2.40%	0.59%	2.99%	3.76%	0.93%	4.69%
Disassembly	4.65%	3.33%	7.98%	7.28%	5.20%	12.48%
Thermal pretreatment	3.97%	10.84%	14.81%	–	–	–
Mechanical treatment	10.68%	10.57%	21.25%	–	–	–
Pyrometallurgy	5.55%	47.42%	52.97%	8.68%	74.15%	82.83%
Total	27.25%	72.75%	100.00%	19.72%	80.28%	100.00%

Table 6

Parameters used for calculating annual maintenance and insurance costs (own table).

	Building maintenance	Maintenance of machines and technical equipment	Insurance
Calculation basis	Investments in buildings	Investments in machines and technical equipment	Total investments
Flat-rate share	1.5%	3%	1%

serves as the most plausible estimate possible of future developments. The default values form the starting price growth rates and ensure differentiation between price development scenarios, even if sufficient data were not available for a particular material. The default values were set at no price change for the lower case, an annual price growth rate of 1.635% for the base case and 2.811% for the upper case. These values correspond to a total price increase by factors of 1, 1.5, and 2, respectively, over the 25-year period considered. The price growth rate in the fixed scenario is always zero. To avoid negative prices in the long run, negative annual price growth rates are set to zero as a safe-side estimate. This boundary condition only affects the price growth rate of steel scrap in the lower price development scenario (actually -0.01146) and the price growth rates of limestone in all price development scenarios except the fixed one (actually -0.02850 , -0.01215 , -0.00039). If no historical price development was available for a specific material, based on which the calculated part of the annual price growth rate could have been determined, the calculation is continued exclusively with the default value. In addition to the fixed case, this also leads to an annual price growth rate of 0 in the lower case. This affects the price growth rates of electronics scrap, refractories, lining, and waste (slag). Table 9 shows all initial prices as well as price growth rates for the four price development scenarios for the materials and process auxiliaries used in the TCO analyses of the recycling routes considered in this work.

Prices for cobalt, nickel and copper are available from commodity exchanges (SMM - Shanghai Metals Market, 2021). After aggregating weekly prices from 2012 to 2021 to average prices on an annual basis, a log-linear ordinary least squares (OLS) regression of these average prices was performed over the time dimension. This results in annual price

growth rates. The average prices of the data available for 2021 constitute the starting points for the price development scenarios.

Material values of output scrap fractions, the slag additives quartz (100% SiO₂) and limestone (95% CaCO₃ and 5% MgCO₃), and the process auxiliary gases oxygen and nitrogen are based on the German foreign trade balance (Destatis - Statistisches Bundesamt, 2021). It contains transactions, each specified by total weight and total value, for various goods. A linear OLS regression of the transaction values on the transaction weights on an annual basis for the years 2008–2020 provides useable information gained from the data. The price per ton thus obtained for the most recent year available forms the initial value for the price development scenarios. A log-linear OLS regression of these prices over the time dimension results in annual price growth rates for each material. Since the selling price for electronics scrap is not available from the German foreign trade balance, it is taken from ESG Edelmetall-Service GmbH & Co. KG (2022) and thus results in 2240 €/t. The order of magnitude of the price could be validated from the findings of Bärwaldt and Kwade (2012).

Table 8

Overview variable costs and revenues considered per process step (own table).

Process step	Process type	Variable cost types	Revenue types
Discharge	No differentiation	None	None
Disassembly	No differentiation	Energy, disposal	Marketable outputs (scrap: steel, copper, aluminum, plastics, and electronics)
Thermal pretreatment ^a	Direct process	No thermal pretreatment	None
	Multi-step process	Material (nitrogen), energy, disposal	None
Mechanical treatment ^a	Direct process	No mechanical treatment	None
	Multi-step process	Energy	Marketable outputs (scrap: steel and copper)
Pyrometallurgy ^a	No differentiation	Material (oxygen, refractories, slag additives, lining), energy, disposal	Marketable outputs (metal alloy)

^a Including exhaust gas cleaning.

Table 7

Workforce cost calculation scheme in the multi-step process (own table).

Process step	Number of employees			Annual costs per employee		Annual workforce costs incl. 15% executive mark-up
	Total	Simple tasks (93%)	Complex tasks (7%)	Simple tasks	Complex tasks	
Discharge	14	13.02	0.98	48,313 €	62,117 €	793,395 €
Disassembly ^b	17	15.81	1.19			963,408 €
Thermal pretreatment ^a	18	16.74	1.26			1,020,079 €
Mechanical treatment ^a	24	22.32	1.68			1,360,105 €
Pyrometallurgy ^a	24	22.32	1.68			1,360,105 €
General areas	18	16.74	1.26			1,020,079 €
Total	122	113.46	8.54	48,313 €	62,117 €	6,517,170 €

^a Including exhaust gas cleaning.

^b Due to the lower capacity of the direct recycling plant in terms of LIB packs per hour, the number of disassembly employees in the direct process amounts to 11 in total resulting in 623,381 € of personnel costs.

Table 9

Material prices and parameters of the price development scenarios (own table).

Material <i>i</i>	Price p_i	Price growth rate $g_{p,i}$			
	Start value	Fixed scenario	Lower scenario	Base scenario	Upper scenario
Cobalt	45,342.92 €/t	0.00000	0.06925	0.08560	0.09736
Nickel	17,208.65 €/t	0.00000	0.01381	0.03016	0.04192
Copper	8675.87 €/t	0.00000	0.01324	0.02959	0.04135
Aluminum	1019.35 €/t	0.00000	0.00214	0.01849	0.03025
scrap					
Copper scrap	3722.22 €/t	0.00000	0.03350	0.04985	0.06161
Steel scrap	215.14 €/t	0.00000	0.00000	0.00489	0.01665
Electronics	2240.00 €/t	0.00000	0.00000	0.01635	0.02811
scrap					
Plastic scrap	269.79 €/t	0.00000	0.00279	0.01914	0.03090
Refractories ^a	235.00 €/t	0.00000	0.00000	0.01635	0.02811
Quartz	1493.91 €/t	0.00000	0.00485	0.02120	0.03296
Limestone	125.85 €/t	0.00000	0.00000	0.00000	0.00000
Lining ^a	36.46 €/pc.	0.00000	0.00000	0.01635	0.02811
Waste (slag)	105.00 €/t	0.00000	0.00000	0.01635	0.02811
Oxygen	0.11 €/m ³	0.00000	0.00505	0.02140	0.03316
Nitrogen	0.08 €/m ³	0.00000	0.01355	0.02990	0.04166
Electricity	191.10 €/MWh	0.00000	0.03062	0.04697	0.05872

^a Estimates based on own experience.

Electricity prices are based on Eurostat data for 2007 to 2020 in the range of two to 20 GWh of energy consumption per year (Eurostat, 2021). This results in an electricity price of 191.10 €/MWh. The solid waste generated consists mainly of slag from pyrometallurgy. According to Briese et al. (2012), the corresponding disposal costs amount to 90 to 120 €/t. For further calculations, an initial average price of 105 €/t is assumed for the disposal of solid waste.

To enable an analysis of individual effects in the sensitivity analysis that is as differentiated as possible, an attempt was made to avoid mixing different influencing factors such as inflation. Hence, wages are kept constant over time in all scenarios.¹

4. Results and discussion

This section provides an overview of the results of the TCO calculations based on the information and models presented in the previous sections. Furthermore, the effects of the different price development scenarios are discussed. A sensitivity analysis is used to address the potential uncertainties of the results. Due to the separate calculations per battery and process type, there are differences in the results for battery compositions and for process types. However, the following analysis focuses mainly on differences in profitability resulting from different battery types recycled throughout the routes.

4.1. TCO results

The economic analysis shows that personnel costs constitute the most important cost driver for all battery types, recycling routes, and price development scenarios. Taking the first year of operation as a reference, personnel costs account for about 62% of all costs in the multi-step process and about 69% in the direct process. Energy and material costs are other important cost drivers amounting to about 22% (about 9% in the direct process) and about 6% (about 9% in the direct

¹ The effect of wage increases in different price development scenarios was checked. Wages increasing according to the mentioned default price increase rates lead to less profitable processes in the basic and high scenario and to no changes in the fixed and low scenario. Change of sign in the result occurs only for NMCLMO/C in the dedicated process in the high scenario.

process) of total costs in the multi-step process. Differences in relative cost shares between battery types are of minor importance and can be explained by different cell compositions.

As stated in Section 3.2, contribution margin analysis follows a process-oriented approach. Pyrometallurgy is by far the most cost-intensive process step, causing approximately 32% of the total costs in the multi-step process and as much as 54% of the costs in the direct process. However, pyrometallurgy represents also the most important source of revenues. The market value of the metal alloy obtained during pyrometallurgical treatment depends on the cobalt, nickel, and copper content of the recycled batteries. In this work, annual metal alloy revenues in the multi-step process for NMC333 batteries amount to 8 million €, whereas recycling of NMC811 and NMCLMO batteries generate metal alloy revenues of 6.3 and 4.1 million € per year. Since LFP batteries contain neither cobalt nor nickel, copper constitutes the only valuable marketable metal in the alloy. Hence, revenues from LFP metal alloy only amount to 0.5 million € in the multi-step process and 1 million € in the direct process.

Unlike discharge and thermal pretreatment, the mechanical treatment step (in the multi-step process) generates revenues, which however cannot cover the respective total process costs. Disassembly and pyrometallurgy thus constitute the only process steps that generate positive contribution margins for NMC333, NMC811 or NMCLMO batteries. In the case of LFP batteries, revenues from metal alloy cannot cover process costs for pyrometallurgy.

Profitability differences between battery types are also particularly evident at NPV and TCO levels. A positive NPV or TCO value indicates that the recycling process is profitable. Table 10 shows an overview of the results from the economic analysis.

The TCO per ton of LIB divides the TCO annuity by the annual mass input of LIBs into the recycling process and can thus be interpreted as the profit or loss per ton of LIB processed. The annual mass input of LIBs is calculated by multiplying the maximum capacity per hour (see Section 3.2) with 5700 productive hours per year. It may thus serve as an indication of a potential gate fee, i.e., in case of a negative TCO per ton of LIB, the amount a third party would have to pay to the recycler for disposing of LIB batteries for recycling, or vice versa, in case of a positive TCO per ton of LIB, the price a recycling company could pay per ton of LIB received. However, these amounts only refer to the break-even point, so that a recycling company paying/receiving this gate fee is not making a profit or loss. It should also be noted that, in order to reduce complexity, this analysis does not include the cost of battery collection, logistics, storage, and potential wage raise, so the actual cost of pyrometallurgical recycling may be higher. All data relates to pre-industrial scale assumptions, which may be subject to economies of scale or learning curves when applied in an industrial context, i.e., it can be expected that profitability will improve over time. Furthermore, material prices and corresponding growth rates are based on data from 2007 to 2021 and can thus only be considered as an estimate but not as a reliable forecast for the future. Nevertheless, the results already provide an indication of the economic viability of pyrometallurgical recycling processes for different LIB types.

In a fixed price scenario, a gate fee would have to be paid to the recycler for all LIB types regardless of the recycling process considered except NMC333 batteries in the direct process. However, there are significant differences in the amount of potential gate fees for the fixed scenario. For the direct process, the gate fee required for LFP recycling is more than four times higher than the gate fee required for NMC811, and almost twice as high as the gate fee needed for NMCLMO. Between the recycling routes, important differences arise. NMC333 recycling amounts to a TCO of 24 € per t LIB in the direct process as compared to a negative TCO of -644 € per t LIB in the multi-step process. For both routes, it can be clearly stated that LFP is by far the least profitable type, followed by NMCLMO. Due to the high content of cobalt and nickel in the battery cell, NMC333 and NMC811 achieve better economic results in a fixed price scenario, although in most cases gate fees would still

Table 10
Overview NPV and TCO results in fixed scenario (own table).

Investment	Multi-step process			Direct process		
	–28,956,530 €			–18,516,337 €		
LIB type	TCO total in €	TCO in €/t LIB	NPV in €	TCO total in €	TCO in €/t LIB	NPV in €
NMC333	–3,045,306	–644	–37,492,461	72,281	24	889,892
NMC811	–4,715,547	–997	–58,055,729	–1,154,692	–376	–14,216,062
LFP	–10,601,598	–2242	–130,522,197	–5,268,778	–1714	–64,866,867
NMCLMO	–6,951,166	–1470	–85,579,680	–2,799,401	–911	–34,464,993

have to be paid to a recycling company when the fixed scenario is considered.

4.2. Sensitivity analysis

The results explained in Section 4.1 are partly based on assumptions that are subject to uncertainties. To cope with these uncertainties, important input factors are varied in a sensitivity analysis to show how differences in assumptions affect the results of the economic analysis. Since the analysis in this paper covers 25 years, price developments have an important impact. Table 11 shows that different price scenarios stress the difference in profitability when comparing different LIB types.

For NMC333 batteries, any price increase within the analyzed scenarios leads to positive TCOs per ton of LIB. The profitability of NMC333 recycling increases to a maximum of 2647 € per ton of LIB in the direct process in the upper price development scenario. For NMC811, price development scenarios only lead to positive TCOs per t LIB in case of the direct recycling process with a maximum of 800 € per ton in the upper price development scenario. For NMCLMO, future price increases improve profitability, but the break-even point is still not reached in most cases. Only in the upper scenario, a positive result can be obtained in the direct process. For all other process type and price scenario combination, the TCO per ton of LIB remains negative and peaks at –1470 € per ton of LIB. In the case of LFP/C batteries in the multi-step process, future price increases have a negative effect on profitability by increasing the gate fee payable to the recycler to a maximum value of 2422 € per ton of LIB. This can be explained by the fact that LFP batteries do not benefit to the same extent as other battery types from price increases in terms of higher revenues, especially for the metal alloy, but still have to bear higher costs for material and energy for the recycling process. However, in the direct process, price development scenarios slightly improve profitability for LFP battery recycling even though the

maximum is still negative at –1.649 €/t LIB.

There are two more parameters whose impact on the results presented in the previous section should be assessed as part of the sensitivity analysis. The first one is the markdown mentioned in Section 3.3, which describes the loss in value of the metal alloy compared to the equivalent quantity of the respective pure materials. The second one is the degree of automation that describes to what extent human labor is replaced by machines. In the further considerations it is assumed that only simple tasks, as defined in Section 3.2, can partly be replaced by automation. Complex tasks will continue to be completely performed by the human workforce.

For the calculations leading to the results presented in the previous section, a markdown of 50% was assumed as described in Section 3.3. Figs. 5 and 6 give an overview of how the TCO per ton of LIB changes in the respective price development scenarios if a markdown between 0% and 100% is applied for the multi-step process and the direct process. Since copper is the only valuable alloy component for LFP and hence LFP alloy output revenues are inferior to other battery types, LFP recycling profitability is to a lesser extent affected by change in the markdown. For NMC333 in particular, but also for NMC811 and NMCLMO, the markdown has a significant impact on the results of the TCO calculation. For the multi-step process, a markdown of 0% instead of 50% leads to a change in TCO per ton of LIB between 1677 € and 4070 € for NMC333 and between 845 € and 1763 € for NMCLMO, depending on the price development scenario. For the direct process, the corresponding ranges are 2040 € to 4750 € and 1127 € to 2202 €. As Figs. 5 and 6 show, a change in the markdown may well lead to profitability of routes that were not profitable before. The routes for NMC333 were already profitable for all price development scenarios except the fixed one in case of the multi-step process. At lower markdowns, the routes for NMC333 are profitable for all scenarios. Even for NMCLMO, where only the direct process in the upper price development scenario was profitable at a 50%

Table 11
Overview NPV and TCO results (own table).

Investment		Multi-step process			Direct process		
		–28,956,530 €			–18,516,337 €		
LIB type	Price scenario	TCO total in €	TCO in €/t LIB	NPV in €	TCO total in €	TCO in €/t LIB	NPV in €
NMC333	Fixed	–3,045,306	–644	–37,492,461	72,281	24	889,892
	Low	2,456,036	519	30,237,633	4,267,199	1388	52,535,870
	Basic	4,962,731	1050	61,098,949	6,317,121	2055	77,773,604
	High	7,200,373	1522	88,647,807	8,138,782	2647	100,201,086
NMC811	Fixed	–4,715,547	–997	–58,055,729	–1,154,692	–376	–14,216,062
	Low	–3,050,125	–645	–37,551,795	308,140	100	3,793,678
	Basic	–1,815,905	–384	–22,356,615	1,445,897	470	17,801,250
	High	–706,729	–149	–8,700,930	2,458,657	800	30,269,904
LFP	Fixed	–10,601,598	–2242	–130,522,197	–5,268,778	–1714	–64,866,867
	Low	–11,009,112	–2328	–135,539,324	–5,266,673	–1713	–64,840,949
	Basic	–11,260,243	–2381	–138,631,134	–5,170,292	–1682	–63,654,351
	High	–11,453,548	–2422	–141,011,024	–5,070,606	–1649	–62,427,060
NMCLMO	Fixed	–6,951,166	–1470	–85,579,680	–2,799,401	–911	–34,464,993
	Low	–5,284,778	–1117	–65,063,850	–1,410,815	–459	–17,369,329
	Basic	–4,406,246	–932	–54,247,756	–554,475	–180	–6,826,446
	High	–3,607,522	–763	–44,414,218	212,521	69	2,616,471

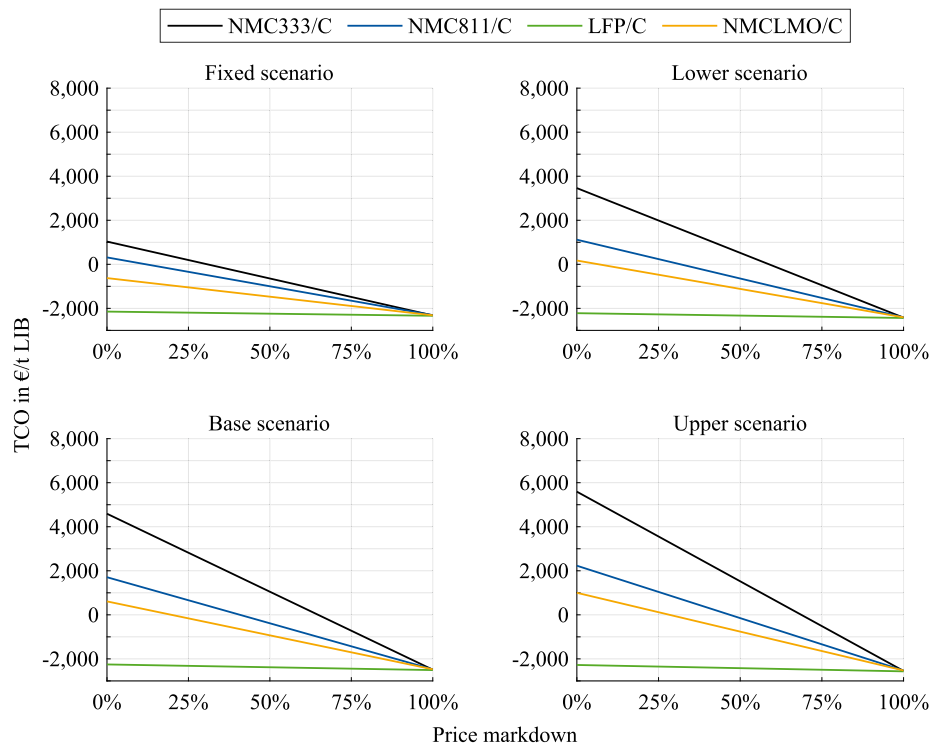


Fig. 5. Impact of the price markdown in the multi-step process (own figure).

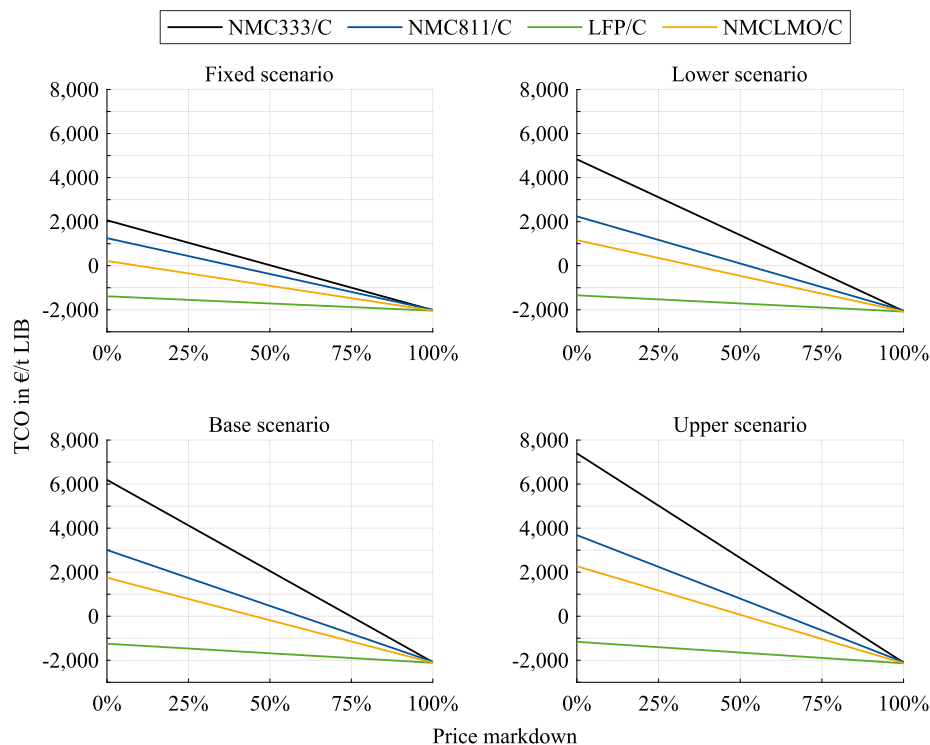


Fig. 6. Impact of the price markdown in the direct process (own figure).

markdown, both process routes eventually become profitable at lower markdowns. For the baseline scenario, the break-even markdown is about 15% in the multi-step process and 45% in the direct process. The break-even markdown for the multi-step process for NMCLMO in the lower scenario is quite low at 7%. For the fixed price scenario, even a markdown of 0% does not make the multi-step process for NMCLMO

profitable. Since the markdown is mainly determined in negotiations with business partners, no more accurate estimates of real markdowns can be given at this point. However, the markdown can make the difference between non-profitable and profitable. The effect of lowering the markdown is highly dependent on the cobalt and nickel content of the batteries, so NMC333 batteries show the largest differences in

profitability when metal alloy markdown decreases.

Based on the workforce costs described in Table 7 in Section 3.2, Figs. 7 and 8 allow an evaluation of the change in TCO per ton of LIB with a variation in the degree of automation. Labor costs only differ for recycling routes but remain constant for all battery types within the same recycling process. Therefore, the same applies to the sensitivity analysis when introducing different automation levels. For the multi-step process, a degree of automation of 100% leads to an increase in the TCO per ton of LIB of 1256 €, while the corresponding value for the direct process is 1126 €. The degree of automation thus has a positive effect on the economic feasibility. Nevertheless, its influence is smaller than the influence of the markdown. Consequently, even with a high degree of automation, only some of the previously non-profitable routes do become profitable *ceteris paribus*. For LFP batteries, none of the processes becomes profitable due to automation. For NMC333, in the fixed scenario, an intermediate degree of automation of 52% for the multi-step process leads to profitability whereas it is already profitable without any automation in the direct process. For NMC811, breakeven is reached in the fixed scenario at a degree of automation of about 79% in the multi-step process and 34% in the direct process. For NMCLMO, values for the TCO around zero are only achieved in at least a lower price development scenario with 89% automation in the multi-step process whereas they are already reached in a fixed price scenario with 81% of automation. In summary, the degree of automation is certainly a parameter that positively influences the result. At present, it is unclear at what point the maximum degree of automation is reached and what degree of automation is realistic. However, the impact of automation on LIB recycling profitability is subordinate. It must also be taken into account that the investments also increase with a higher degree of automation, which weakens the results discussed here. However, these results emphasize the necessity of automation but should not be understood as point-accurate forecasts. Moreover, process optimization should not be based on automation alone.

5. Outlook and conclusions

This study provides a first indication on the economic profitability of pyrometallurgical recycling processes for individual LIB systems with significantly different cell chemistries. The rather idealized consideration of individual battery types instead of current/future battery market mixes was deliberate in order to obtain reliable results and to highlight individual effects at an early stage. Future battery types or batteries that are significantly different from the batteries considered here may lead to results that differ from the figures presented in this paper. For reasons of data availability all technical data is dimensioned at pre-industrial scale and may hence be subject to further improvements if applied to larger scales. Focusing on the recycling process only, the economic analysis does not consider logistics costs for battery collection. Currently, it is unclear which costs arise for the collection of end-of-life LIBs and which volumes can actually be obtained with the current reverse logistics systems for LIBs.

Charges for CO₂ emissions have not been considered in the analysis so far. However, recycling plants may also be subject to CO₂ emission fees in the future so that further costs for CO₂ emissions may then arise in both recycling routes. The accurate amount of these costs is currently difficult to forecast as publicly available reliable data on CO₂ emissions in pyrometallurgical recycling processes is lacking.

With regard to the first research question outlined in Section 1, the present economic analysis, despite its limitations, shows that important differences in terms of profitability of pyrometallurgical recycling exist between different LIB cell chemistries. If current price levels are considered, LIB pyrometallurgical recycling seems to be only profitable for NMC333 batteries in the direct process whereas all other combinations of the cell chemistries and recycling paths considered in the analysis show negative profitability results. Even though the analysis related to pre-industrial scale data, the analysis clearly shows that profitability of pyrometallurgical recycling routes strongly depends on the cobalt and nickel content of the LIBs. Consequently, for NMCLMO recycling, break-even can only be reached in specific price development scenarios whereas LFP recycling is not profitable in any scenario

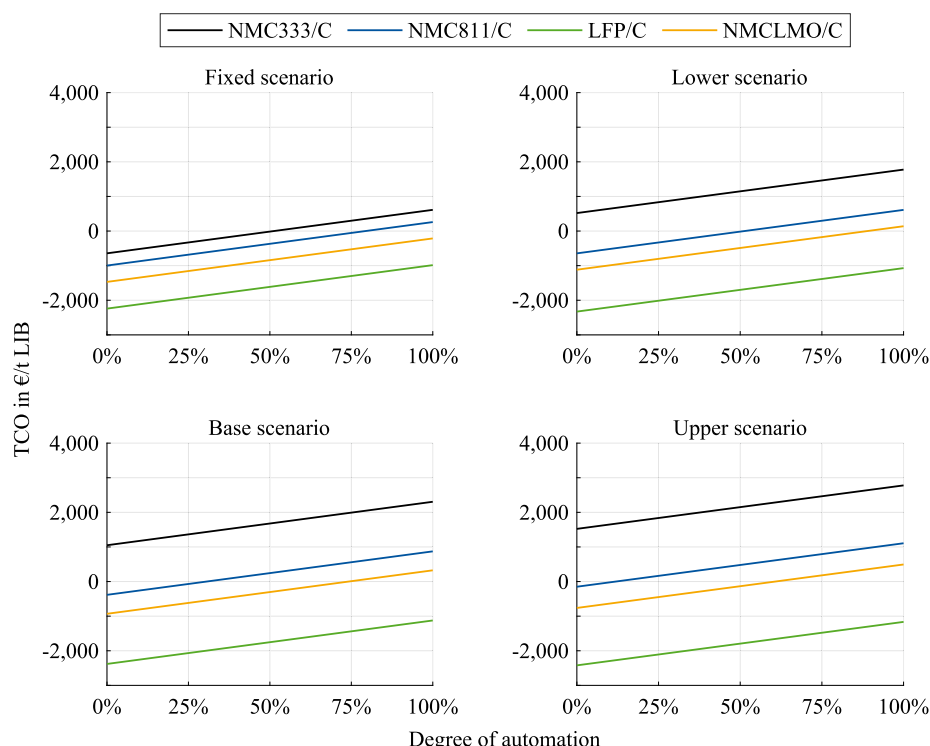


Fig. 7. Impact of the degree of automation in the multi-step process (own figure).

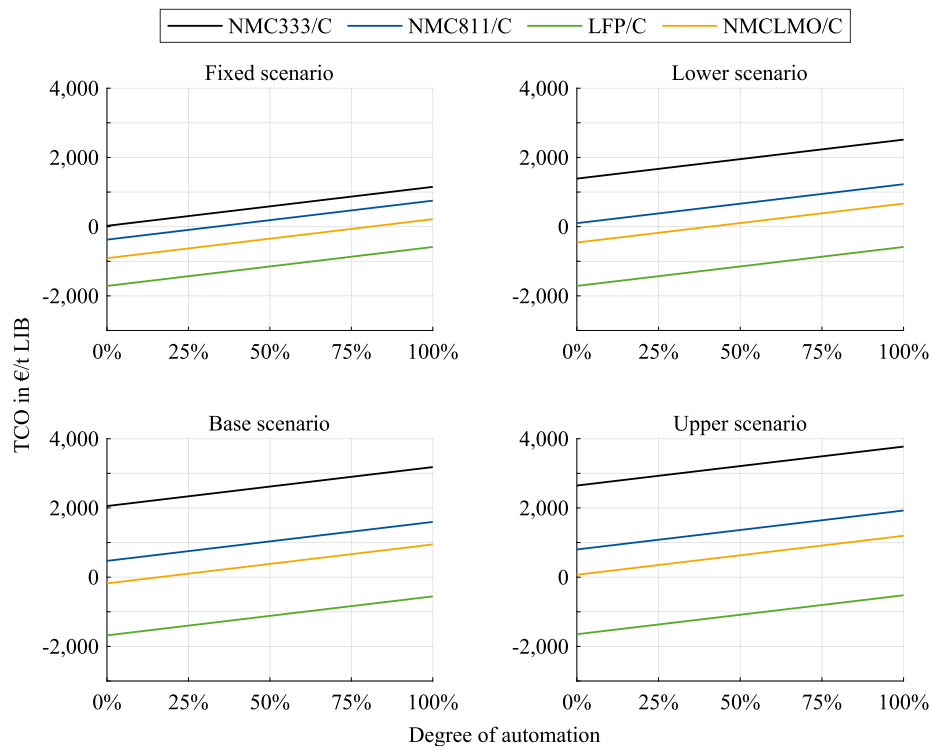


Fig. 8. Impact of the degree of automation in the direct process (own figure).

regardless of price developments or increasing process automation. With regard to the current LIB/BEV market which shows a clear trend towards cheaper battery cell chemistries with lower cobalt and nickel contents, the economic analysis presented reveals a major challenge which needs to be considered in the debate on the future of electromobility. Another important aspect for improving profitability from a technical point of view is the recovery of lithium, which is lost in the slag in the pyrometallurgical recycling processes presented. An economic recovery of lithium from slag might be realized within the framework of European lithium primary recovery projects. Another possibility is offered by the multi-stage recycling process, in which it would be possible to recover lithium with partial graphite recovery before smelting. Nevertheless, it should also be preconceived that other recycling routes besides pyrometallurgy exist.

To answer the second research question presented in Section 1, the analysis examined price developments, different markdowns and increasing automation as potential factors influencing the profitability of pyrometallurgical LIB recycling. It was shown that a higher degree of automation would increase process profitability but price increases and lower markdowns of metal alloy products from pyrometallurgical recycling processes provide more significant levers for profitability at least for NMC333, NMC811 and NMCLMO recycling. For all battery types, economies of scale could potentially increase recycling process profitability. However, since the furnace constitutes the process bottleneck, it is expected that economies of scale are fostered by facility enlargements due to synergy effects.

Developments or circumstances increasing the profitability of recycling routes can be initiated or promoted by regulatory measures such as raw material taxes, recycling subsidies or regulations imposing a minimum use of recycled material for LIB production. At EU level, there is already a minimum recycling quota of 50% for LIBs (EU - European Union, 2006). Increases in recycling volumes enabling economies of scale could be supported by further increasing legal minimum recycling quota for BEV LIBs. However, if LIB recycling will not become profitable for particular battery cell compositions, it needs to be discussed who will finally bear the costs of recycling. In this context, it might be possible

that LIB producers need to put a markup on cheap LIBs in order to cover recycling costs at the end of useful life. Hence, our results indicate that LIB recycling will likely need political support through economic incentives or regulatory requirements – at least until price developments and process improvement will lead to economic feasibility.

To enrich a debate around possible regulatory action supporting pyrometallurgical recycling of LIBs, the ecologic and societal perspectives should also be taken into consideration. Further research could thus concentrate on environmental and societal consequences of pyrometallurgical LIB recycling and its alternatives. However, these aspects cannot be analyzed independently from technical as well as economic considerations to reflect potential trade-offs. The integration of social and environmental aspects into the TCO model in terms of external costs hence constitutes an avenue for further research to enable a combined analysis. Further research may also address the optimization of reverse logistic chains for LIBs to maximize process efficiency and minimize costs and the environmental impact of collection. To obtain a holistic picture of current and future LIB recycling opportunities, several process routes, such as hydrometallurgy, should be considered comparatively. Analyzing automation and economies of scale in more depth is also a promising avenue for future research when more accurate data will become available. In this vein, process learning would be another promising avenue of future research. For future research, we also recommend to consider design changes of LIBs over time whenever data will be available.

CRediT authorship contribution statement

Linda Reinhart: Methodology, Formal analysis, Writing – original draft, Writing (economic analysis), Original Draft. **Dzeneta Vrucak:** Methodology, Formal analysis, Writing – original draft, Writing (metallurgical/technical analysis) - Original Draft. **Richard Woeste:** Methodology, Formal analysis, Writing – original draft, Writing (economic analysis) - Original Draft. **Hugo Lucas:** Formal analysis, (metallurgical/technical analysis). **Elinor Rombach:** Conceptualization, Writing – original draft, Writing (metallurgical/technical analysis) -

Review & Editing, Project administration. **Bernd Friedrich:** Conceptualization, Writing – original draft, Writing (metallurgical/technical analysis) - Review & Editing, Supervision. **Peter Letmathe:** Conceptualization, Writing – original draft, Writing (economic analysis) - Review & Editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

Data availability

The data that has been used is confidential.

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Appendix A

Assumptions and derivation of composition and distribution of the output generated in the two pyrometallurgical recycling processes

In the two pyrometallurgical recycling processes presented, the difference lies in the optional upstream connection of a pre-processing stage before the actual smelting process. Whereas in the multi-step process, dismantled battery modules are thermally pretreated and mechanically processed before pyrometallurgical processing, the direct process contains less process steps so that pyrometallurgical treatment directly follows dismantling to battery module level. Consequently, output streams differ amongst the recycling routes considered.

Battery configuration

The different battery cells (NMC333, NMC811, LFP and NMCLMO) have been configured using the BatPaC 5.0 model by [Knehr et al. \(2022\)](#). In order to guarantee the comparability of the batteries despite different cell chemistries and hence different structural setups, battery capacities and modular structure of the packs with the number of cells (default setting in BatPaC 5.0) were presumed to be the same for the four battery systems. All batteries considered for the analysis contain pouch cells with an aluminum casing. The overall weight of the battery packs ranges from 115.80 kg (NMC811) to 149.57 kg (LFP), a detailed overview of their weight composition can be seen in [Figs. 9 and 3](#).

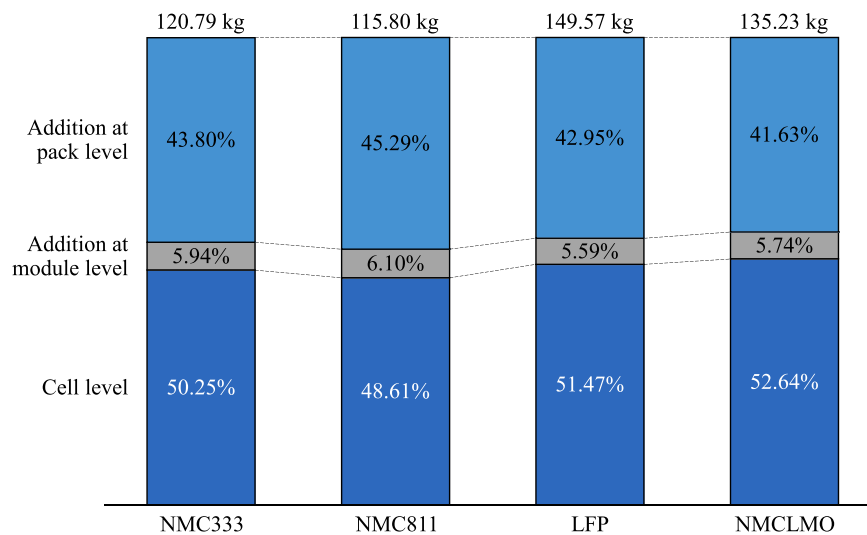


Fig. 9. Battery pack weight compositions at system level (NMC333, NMC811, LFP and NMCLMO), configured with the BatPaC 5.0 model (own figure).

Discharge

In the discharging step, energy recovered during discharge and energy requirements for the process are assumed to sum up to zero and will not be further focused in this study.

Disassembly

In the disassembly step, the energy storage systems will be dismantled to module level. [Table 12](#) shows the obtained fractions during dismantling. Besides battery modules, copper, aluminum, and steel (Fe–Ni–Cr alloy) scrap as well as other scrap (electronics, polymers) and waste result as output fractions from disassembly. It is assumed that there will be no losses during dismantling. A detailed overview of output fractions and energy requirements during dismantling is provided in [Table 12](#).

Table 12

Process outputs and assumptions of energy input requirement during dismantling step (own table).

LIB type	Output fractions in kg/LIB pack					
	Modules	Cu scrap	Al scrap	Steel scrap	Other scrap	Waste
NMC333	67.9	1.1	10.0	35.0	3.6	3.3
NMC811	63.4	1.1	9.9	34.6	3.6	3.2
LFP	85.3	1.2	11.3	44.3	3.6	3.8
NMCLMO	78.9	1.1	10.7	37.4	3.6	3.5
Energy requirement: 0.06 kW h/t LIB						

Thermal Pretreatment

During thermal pretreatment in the multi-step process, the organic matter such as solvent of the electrolyte, the separator, binder, and interconnect panels volatilize assuming 100% volatilization. The thermal pretreatment takes place at a nitrogen atmosphere. The thermally treated modules are further processed. As outlined in Section 3.1, the distribution coefficients per battery component during the thermal pretreatment and mechanical processing are based on a modeling tool by Friedrich and Peters (2019) with own adjustments. The distribution of the mass flow can be seen in Table 13.

Table 13

Output fractions and assumptions of energy and nitrogen input requirements during thermal pretreatment (own table).

LIB type	Output fractions in kg/LIB pack	
	Thermally treated modules	Volatilized material
NMC333	58.6	9.2
NMC811	54.5	8.8
LFP	72.9	12.5
NMCLMO	66.9	12.0
Energy requirement: 172.31 kW h/t LIB (Technical) Nitrogen requirement: 9.6 m ³ /t LIB		

Mechanical treatment

After thermal treatment the remaining battery material is mechanically processed in the multi-step process. During processing, losses of 1% of the input components can occur through dust generation. Furthermore, losses of active material are considered and will be found in the other metallic fractions. The distribution of the output mass flow can be seen in Table 14.

Table 14

Output fractions and assumption of energy input requirement during mechanical processing (own table).

LIB type	Output fractions in kg/LIB pack			
	Active mass	Al/Cu scrap	Steel scrap	Waste
NMC333	46.8	7.7	3.6	0.6
NMC811	43.1	7.4	3.5	0.5
LFP	57.9	10.0	4.2	0.7
NMCLMO	52.4	9.6	4.0	0.7
Energy requirement: 1450.13 kW h/t LIB				

Pyrometallurgy

In the pyrometallurgy, the active mass is smelted. The elemental distribution during smelting has been calculated with FactSage 8.0 using the FScopp, FToxid and FactPS databases. Oxygen partial pressure is set to 1e-14 bar. The distribution coefficients for smelting are shown in Tables 15 and 16 for the two process routes. For all battery types, module enclosure is set as stainless steel 304 which contains nickel so that minor shares of nickel also occur in LFP metal alloy.

Table 15

Percentage distribution coefficients of elements during smelting of thermally and mechanically pretreated battery material in the multi-step process, calculated with FactSage 8.0 (own table).

LIB	Outputs	Distribution coefficients in %														
		Al	Cu	Mn	Mg	Fe	Ni	C	Li	Co	P	F	Si	Cr	O	Ca
NMC333	Gas	0.0	0.5	2.1	1.4	0.0	0.0	99.4	3.5	0.0	0.0	42.0	0.0	0.0	75.8	0.0
	Metal	0.0	99.2	85.6	0.0	99.8	100	0.6	0.0	100	5.4	0.0	0.0	85.0	0.0	0.0

(continued on next page)

Table 15 (continued)

LIB	Outputs	Distribution coefficients in %														
		Al	Cu	Mn	Mg	Fe	Ni	C	Li	Co	P	F	Si	Cr	O	Ca
NMC811	Slag	100	0.3	12.3	98.5	0.2	0.0	0.0	96.5	0.0	94.6	58.0	100	15.0	24.2	100
	Gas	0.0	0.5	0.7	1.7	0.0	0.0	99.8	3.7	0.0	0.0	49.1	0.0	0.0	78.9	0.0
	Metal	0.0	99.2	96.3	0.2	99.9	100	0.2	0.0	100	84.8	0.0	0.5	94.8	0.0	0.0
	Slag	100	0.2	3.1	98.1	0.1	0.0	0.0	96.3	0.0	15.2	50.9	99.5	5.2	21.1	100
LFP	Gas	0.0	0.2	1.9	0.2	0.0	0.0	99.8	1.3	0.0	4.4	57.6	0.0	0.0	71.2	0.0
	Metal	0.0	99.7	58.0	0.0	100	100	0.2	0.0	0.0	74.7	0.0	0.0	100	0.0	0.0
	Slag	100	0.1	40.1	99.8	0.0	0.0	0.0	98.7	0.0	20.9	42.4	100	0.0	28.8	100
NMCLMO	Gas	0.0	0.3	2.7	0.4	0.0	0.0	99.7	11.0	0.0	0.0	66.6	0.0	0.0	70.6	0.0
	Metal	0.0	99.6	59.0	0.0	99.8	100	0.3	0.0	100	99.6	0.0	0.1	84.2	0.0	0.0
	Slag	100	0.1	38.3	99.6	0.2	0.0	0.0	89.0	0.0	0.4	33.4	99.9	15.8	29.4	100

Table 16

Percentage distribution coefficients of elements during smelting of battery modules in the direct process, calculated with FactSage 8.0 (own table).

LIB	Outputs	Distribution coefficients (battery modules without pretreatment)														
		Al	Cu	Mn	Mg	Fe	Ni	C	Li	Co	P	F	Si	Cr	O	Ca
NMC333	Gas	0.0	0.4	2.0	0.5	0.0	0.0	99.7	8.3	0.0	0.1	63.7	0.0	0.0	72.3	0.0
	Metal	0.0	99.4	73.4	0.0	99.8	100	0.3	0.0	100	97.6	0.0	0.3	92.1	0.0	0.0
	Slag	100	0.2	24.6	99.5	0.1	0.0	0.0	91.6	0.0	2.3	36.3	99.7	7.9	27.7	100
NMC811	Gas	0.0	0.5	1.0	0.5	0.0	0.0	99.8	9.3	0.0	0.0	68.4	0.0	0.0	75.1	0.0
	Metal	0.0	99.4	86.5	0.0	99.9	100	0.2	0.0	100	100	0.0	4.7	94.8	0.0	0.0
	Slag	100	0.1	12.5	99.5	0.1	0.0	0.0	90.7	0.0	0.0	31.6	95.3	5.2	24.9	100
LFP	Gas	0.0	0.2	1.7	0.2	0.0	0.0	99.8	12.2	0.0	1.2	65.8	0.0	0.0	70.8	0.0
	Metal	0.0	99.7	63.1	0.0	99.9	100	0.2	0.0	0.0	92.4	0.0	0.0	99.9	0.0	0.0
	Slag	100	0.1	35.2	99.8	0.1	0.0	0.0	87.7	0.0	6.4	34.2	100	0.1	29.2	100
NMCLMO	Gas	0.0	0.3	2.7	0.4	0.0	0.0	99.7	11.0	0.0	0.0	66.6	0.0	0.0	70.6	0.0
	Metal	0.0	99.6	59.0	0.0	99.8	100	0.3	0.0	100	99.6	0.0	0.1	84.2	0.0	0.0
	Slag	100	0.1	38.3	99.6	0.2	0.0	0.0	89.0	0.0	0.4	33.4	99.9	15.8	29.4	100

The slag system chosen for smelting battery modules is $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$. The aluminum in the input serves as slag former and the need for quartz (100% SiO_2) and limestone (95% CaCO_3 and 5% MgCO_3) varies for each input stream. Table 17 shows the various requirements of slag additives per kilogram LIB.

Table 17

Addition of slag additives in percent of the battery input mass during pyrometallurgy, calculated with FactSage 8.0 (own table).

LIB type	Direct process		Multi-step process	
	Quartz	Limestone	Quartz	Limestone
NMC333	5.1	14.3	3.7	10.5
NMC811	5.2	14.6	3.8	10.7
LFP	5.6	15.7	4.4	12.3
NMCLMO	5.0	14.1	3.8	10.8

Due to the absence of established industrial processes treating only LIBs, energy consumption during the melting process is based on assumptions in the present analysis. In order to increase comparability amongst battery cell chemistries, data on energy consumption, refractory material and lining are assumed to be at the same level for both process routes. Detailed information on respective input data resulting from prior research projects is provided in Table 18. The reported numbers reflect best estimates from a confidential and publicly-funded research project.

Table 18

Other process inputs in the pyrometallurgy (own table).

Inputs	Requirements per t LIB	
	Requirements per t LIB	Unit of measure
Oxygen	0.07	m ³
Refractories	4.94	kg
Energy	790.06	kWh
Lining	2.81	pc.

Table 19 shows the process energy requirements for smelting the input material (battery module and slag additives). Negative values indicate that an exothermic reaction is occurring. The different results are due to the organic components still present in the direct process. The values shown were

not used as a basis for the economic calculation, but merely provide indications for a later design of the process control.

Table 19

Process energy requirement during smelting different battery chemistries in kWh per kg input material (LIB and slag additives), calculated with FactSage 8.0 (own table).

LIB type	Process energy requirements in kWh/kg LIB with slag additives	
	Direct process	Multi-step process
NMC333	−0.018	0.053
NMC811	−0.142	−0.107
LFP	0.137	0.144
NMCLMO	−0.067	0.142

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