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Material and energy flow analysis for environmental and economic impact assessment of industrial recycling routes for lithium-ion traction batteries

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A B S T R A C T

Electric vehicles powered with renewable energy are considered a key technology to decarbonize the mobility sector. However, the currently used lithium-ion batteries contain environmentally harmful, scarce, and expensive materials. The recycling of spent traction batteries could mitigate the environmental impact of electric mobility by substituting primary raw materials with recovered secondary materials. Moreover, it would counter the issues related to resource scarcity and expensive materials. Therefore, the automotive industry needs to establish effective processes for taking back and recycling of batteries. While many studies have analyzed the environmental and economic impacts of lithium-ion battery recycling, the lack of transparency of the energy and material flows as well as the missing comparability between different recycling routes contradicts an in-depth life cycle engineering. Therefore, this paper aims to provide transparent material and energy flow analysis on process unit level based on physical and chemical relationships and use this to assess the environmental and economic impacts of three widely used recycling routes. The analysis focuses on pyrometallurgical, mechanical, and thermal-mechanical pretreatment, and subsequent hydrometallurgical material recovery. Furthermore, we assess the environmental and economic impacts of each recycling route. The results indicate that mechanical recycling has the highest economic benefit and avoids most environmental impacts especially due to graphite and lithium recovery. A thermal-mechanical pretreatment has environmental benefits but results in lower profit. The pyrometallurgical pretreatment results in large amounts of slag, for which the hydrometallurgical processing reduces the avoided environmental impacts significantly. The assessment results support transparent decision-making regarding the implementation and further engineering of recycling infrastructure.

1. Introduction

Electric vehicles can enable significant emission reductions in the mobility sector when powered with renewable energy. However, their advancing market penetration and the increasing demand for traction batteries may become problematic from a sustainability perspective due to the high need for scarce materials, such as lithium, cobalt, and nickel, along with environmental, economic, and social concerns in their supply chain (International Energy Agency, 2020; Reuter, 2016). Considering greenhouse gas (GHG) emissions, the production of lithium-ion batteries (LIB) and particularly the extraction and refining of the active materials account for approximately 40% of the total life cycle emissions of an electric vehicle (Sun et al., 2020). Besides, several social issues regarding the extraction and refining of raw materials have been reported. For example, about 60% of global cobalt mining is located in the Democratic Republic of Congo, where the risks of child labor and poor working conditions are particularly high (Fu et al., 2020). Additionally, the geographic concentration of the raw materials in combination with the rapid demand increase may lead to supply bottlenecks (Mayyas et al., 2019).

The circulation of critical and scarce materials through the recycling of spent LIBs allows for reducing the material-related environmental, economic, and social impacts of battery production and for improving supply security (Cerdas et al., 2018; Ciez and Whitacre, 2019).

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Therefore, European legislation obliges companies to take back the batteries which they put on the market and to recycle them with a minimum recovery rate of 50% regarding total mass (European Commission, 2020). New regulation proposals even extend the minimum recovery rate to 65% by 2025 (European Commission, 2020). Furthermore, material-specific recovery rates are discussed, e.g., 90% for cobalt, nickel, and copper, and 35% for lithium. Additionally, minimum quotas for secondary materials in new batteries will be enforced from 2030 on.

While car and battery manufacturers, as well as recycling companies, have started to establish industrial recycling routes for spent LIBs, their low profitability due to the currently low amount of returned LIBs is still an issue. However, the amount of spent batteries is expected to increase significantly by 2030 when more electric vehicles will reach their end-of-life (Dunn et al., 2021; International Energy Agency, 2020). Hence, companies need to decide which recycling processes to implement or how to adjust and expand the current capacities. In this context, various recycling routes are possible. They are usually composed of mechanical, thermal, pyrometallurgical, and hydrometallurgical processes. The various process combinations can differ considerably regarding the recovered materials, achievable purities, recovery rates, required infrastructure, process emissions, investments, costs, and revenues. However, a comprehensive and transparent analysis of the processes, their environmental and economic impacts, and the recoverable materials is missing.

Therefore, this paper provides a material and energy flow analysis (MEFA) for the economic and environmental assessment of three industrial recycling routes for spent LIBs. It extends previous studies with similar boundary conditions by providing a transparent, detailed, and adjustable analysis for material and energy flows. Based on the MEFA, the achievable recovery rates, related GHG emissions, required investments, and potential profits of the three recycling routes are scrutinized. The models on unit process level enable a targeted Life Cycle Engineering of recycling process chains by visualizing the effects of decisions along the recycling process route, which is not covered by available tools yet. Overall, this study seeks to provide transparent and comprehensive decision support for companies that aim to implement or adjust LIB recycling or decide on strategic partnerships.

The paper proceeds with an overview of the current industrial recycling routes for LIBs and their assessment in Section 2. The modeling and assessment approach for this study is introduced in Section 3. Subsequently, the economic and environmental impact assessment results are presented and relevant levers for improvement are discussed in Section 4. The paper concludes with a summary of the main findings and their implications as well as an outlook in Section 5.

2. State of research

This section starts with a summary of the common recycling routes for LIBs based on scientific literature and patents. Next, existing economic and environmental assessment studies are analyzed to identify research gaps.

2.1. Overview of industrial recycling processes

In Germany, three research projects, LiBRi, LithoRec, and EcoBatRec, are considered pioneers for sustainable recycling of traction batteries from electric vehicles, developing and investigating individual recycling routes. These three recycling routes can be distinguished: pyrometallurgical pretreatment (Route 1), mechanical pretreatment (Route 2), and hybrid pretreatment (Route 3). All pretreatment routes are followed by a hydrometallurgical material recovery (Fig. 1). The developed processes have been commercialized by the companies Umicore, Duesenfeld, and Accurec. In all routes, the spent LIBs entering the recycling process are first discharged and disassembled before they undergo pyrometallurgical, mechanical, or thermal treatment. In Route 1, the disassembled battery packs are melted in a pyrometallurgical process and the materials are separated based on their density in an alloy and a slag phase. The Route 2 includes a mechanical pretreatment in which the battery modules are crushed and the materials are further separated based on physical properties such as density or magnetism (Hanisch et al., 2016). The Route 3 combines mechanical and pyrometallurgical processes in combination with an upstream thermal treatment (Sojka et al., 2020). All recycling routes contain a subsequent hydrometallurgical material recovery. The types and quality of the output materials are not only influenced by the upstream process and the input materials (e.

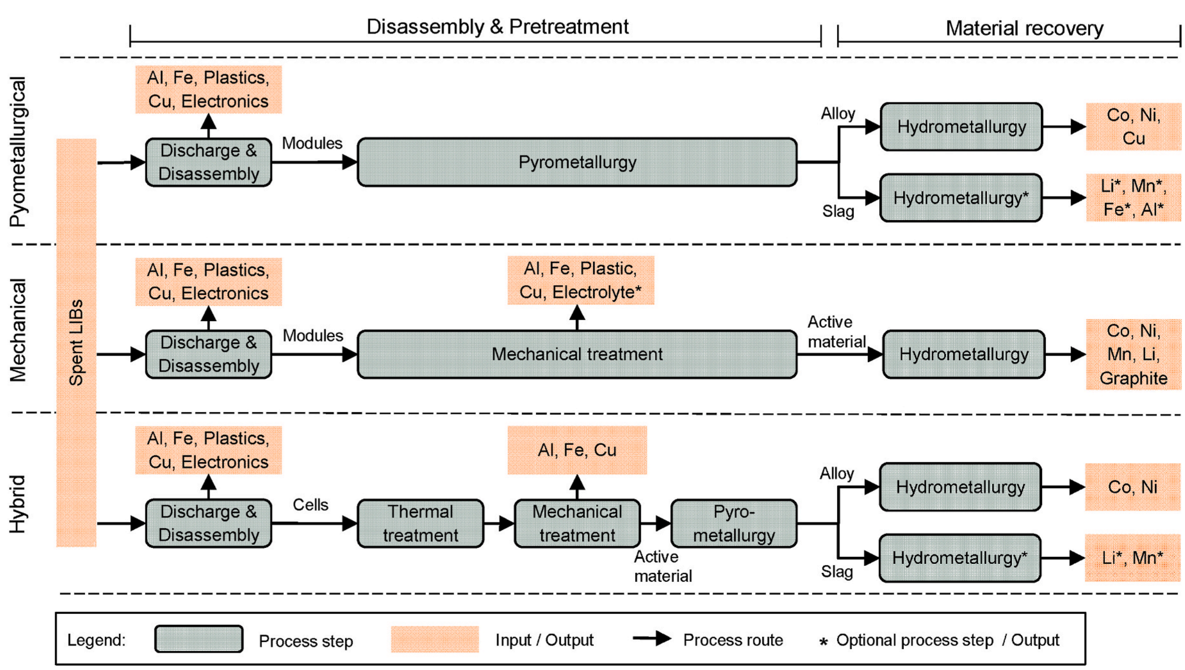

Fig. 1. Recycling routes for exemplary industrial recycling operations for LIBs, adapted from Doose et al. (2021), Sommerville et al. (2021), and Velázquez-Martinez et al. (2019).

g., leaching, precipitation, and extraction agents), but also by the specific process parameters (e.g., temperature, pressure, pH, and inert gas atmosphere) (Brückner et al., 2020; Harper et al., 2019; Velázquez-Martínez et al., 2019). A detailed process description is provided in Section SM.1 of the supplementary material (SM).

Each recycling route has specific advantages and disadvantages (Table 1). In general, more materials can be recovered with a mechanical pretreatment than with a pyrometallurgical pretreatment due to the high temperatures in pyrometallurgy, which lead to the combustion of plastics and graphite. Additionally, some materials, including aluminum and lithium, are concentrated in the slag, which is currently not economically recyclable. Furthermore, the melting process in the pyrometallurgy results in higher energy consumption compared to the crushing, shredding, and classification processes in the mechanical pretreatment. In contrast, pyrometallurgy is more robust and achieves greater throughputs. Problematic for both pretreatments are the similar physical and chemical properties of the active materials, which make their separation difficult (Chen et al., 2019; Harper et al., 2019; Mossali et al., 2020). In order to achieve battery-grade materials, impurities must be minimized and physical and chemical properties must be maintained (Harper et al., 2019). Therefore, hydrometallurgy is operated to concentrate the active materials for further use. In addition to these core processes, other (less mature) processes such as direct recycling exist, in which the material structure of the cathode and anode is largely retained, thereby avoiding subsequent synthesis processes.

2.2. Assessment of recycling routes

The main characteristics of existing studies focusing on the environmental and economic assessment of LIB recycling are summarized in Fig. 2. An expanded version of this table with further details is available in Table SM.1.1.

Within the research projects LiBRi, LithoRec, and EcoBatRec an environmental life cycle assessment (LCA) according to ISO14040/14044 was conducted based on existing processes on laboratory or pilot scale and generic electric vehicle traction batteries. While the LiBRi project assumed a mix of lithium nickel manganese cobalt oxide (NMC), lithium nickel cobalt aluminum oxide (NCA), and lithium iron phosphate (LFP) based batteries, the LithoRec and EcoBatRec project assessments were based solely on NMC chemistry. All three routes were reported with a net credit indicating that the benefits of the recycling outweigh the associated burdens (Buchert et al., 2011; Buchert and Sutter, 2016a, 2016b). The environmental impacts of the LithoRec route were further evaluated by Cerdas et al. (2018), who set up the overall mass and energy balance from dismantling to hydrometallurgical processing (with a focus on mechanical processing) and quantified process-specific material and energy flows. The economic feasibility of the LithoRec process was assessed by Thies et al. (2018), who determined investments and operating costs based on the pilot plant. Their findings indicate that the economic feasibility of the LithoRec process is primarily influenced by the development of battery returns and market prices.

The first publicly available closed-loop battery recycling model reflecting cost and environmental impacts is the EverBatt model (**Dai et al., 2019**). The spreadsheet-based tool facilitates the evaluation of the environmental and economic impacts of recycling for different battery chemistries. EverBatt focuses on battery pack recycling and comprises a benchmark assessment of virgin versus recycled material. **Ciez and Whitacre (2019)** extended existing environmental and economic assessments of LIB recycling by differentiating between various battery chemistries and cell designs. The comparison of pyrometallurgical, hydrometallurgical, and direct recycling technologies is done on cell level and does not provide insights on battery module and pack level. An LCA study of LIBs considering the whole product life cycle is done by **Sun et al. (2020)**. The assessment of an NMC-622 battery uses mainly primary data of two leading battery recycling companies with an annual treatment capacity of 3000 t of spent batteries. The authors find that especially the recycling of wrought aluminum avoids environmental impacts due to its high mass share. Additionally, **Hao et al. (2017)** assess the hydrometallurgical treatment on process unit level.

Table 1
Advantages and disadvantages of industrial battery recycling technologies, based on Chen et al. (2019), Harper et al. (2019), Mossali et al. (2020), and Thompson et al. (2021).

	Recycling technology	Advantages	Disadvantages
Pretreatment	Pyrometallurgy	High throughput	High energy consumption
		Simple and mature process	Loss of organic materials
		No need for pretreatment	CO_2 and hazardous gaseous emissions (need for off-gas treatment)
		No wastewater production	Fewer recoverable materials
		(Generation of exothermic reaction reducing energy consumption)	Alloy (and slag) require further processing
			Only Co rich LIB chemistries profitable
	Mechanical processing with optional thermal treatment	Recovery of organic materials possible	Explosion risk
		Moderate energy consumption	Complex delamination (binder elimination)
		Good technology readiness	Need for off-gas treatment
		High and scalable throughput	Active materials require further processing
		Mobile application possible	
		Material structure remains	
Material recovery	Hydrometallurgy	High recovery efficiency and quality outputs	Wastewater production (need for wastewater treatment)
		Good technology readiness	Complexity of procedure
		Moderated energy consumption	Need for pre-treatment (sorting and size reduction)
		No (few) gaseous emissions	Selectivity of reagents
		Recovery of most LIB constituents	Incomplete binder/electrolyte recycling (impurities)
		Mild reaction conditions	

Publication	Assessment subject	Battery type (cell chemistry)	Recyling process	Modeling level	Assessment dimension
Buchert et al. 2011	Pack	NMC NCA LFP	D P H	PU	ENV
Buchert et al. 2016a	Pack	NMC	D M H	PU	ENV
Buchert et al. 2016b	Pack	NMC	D M P	PU	ENV
Ciez and Whitacre 2019	Cell	NMC	D M P H DP	C	ENV ECON
Dai et al. 2019	Pack	NMC NCA LFP LMO + LCO	D M P H DP	PU SP	ENV ECON
Dunn et al. 2012	unclear	LMO	D M P H DP	SP	ENV
Hao et al. 2017	Pack	NMC	H	PU	ENV
Hendrickson et al. 2015	Pack	NMC LFP	D P H	C PU	ENV ECON
Hoyer et al. 2015	Pack	NMC LFP	D M H	PU	ECON
Lander et al. 2021	Pack	NMC NCA LFP LMO	D P H DP	C	ECON
Mohr et al. 2020	Cell	NMC NCA LFP + SIB	M P H		ENV
Rinne et al. 2021	unclear	mixed NiMH + LIB	(M) H	SP	ENV
Sun et al. 2020	Pack	NMC	H	C	ENV
Thies et al. 2018	Pack	NMC	D M	C PU	ECON
Thompson et al. 2021	Pack	NMC	D M H	PU	ECON

Legend:
NMC $LiNi_xMn_xCo_xO_2$; NCA $LiNi_xCo_xAl_xO_2$; LFP $LiFePO_4$; LMO $LiMn_2O_4$; LCO $LiCoO_2$; SIB Sodium-ion battery; NiMH NiMeH; D Dismantling; M Mechanical; P Pyrometallurgy; H Hydrometallurgy; DP Direct physical; C Cumulated; PU Process unit; SP Single processes; ENV Environmental; ECO Economic

Fig. 2. Overview of publications that quantify the environmental and economic performance of LIB recycling.

Cumulated models evaluate recycling as a whole. Process unit models evaluate individual processes, e.g. precipitation, and single process models evaluate individual process steps, e.g. precipitation and filtration. Mohr et al. (2020) set up a parameterized process model of state-of-the-art pyrometallurgical and hydrometallurgical recycling for different cell chemistries. The literature-based life cycle models are used as a benchmark for the evaluation of an advanced hydrometallurgical recycling process based on primary data from the company Duesenfeld. The authors state that the amount of avoided environmental impacts highly depends on the processed cell chemistry and the considered cell design. In particular, the material and energy flows of the hydrometallurgical processes are highly relevant. Rinne et al. (2021) focus on the environmental assessment of the hydrometallurgical treatment of mixed nickel-metal hydride (NiMH) and lithium-ion battery streams. The extensive life cycle inventory is realized by physically simulated material and energy flows based on experimental lab-scale data. The study indicates that recycled metals have a significantly lower global warming potential (GWP) compared to primary metals, but problem-shifting to other impact categories might occur. Further, the authors claim that the benefits of the hydrometallurgical treatment depend on the nickel and cobalt content of the input stream. An extension of the economic assessment of different mechanical and hydrometallurgical processes is given by Thompson et al. (2021). The authors analyze the effect of the disassembly depth as well as different recycling routes for mechanical and hydrometallurgical recycling routes.

Most studies describe LIB recycling as economically beneficial and with the potential to avoid environmental impacts, although the potential benefit varies. More energy-intensive recycling routes, such as pyrometallurgical processing, are found to have higher environmental impacts compared to mechanical and hydrometallurgical recycling routes. However, the models are not transparent or detailed enough to support the decisions of companies for investing in recycling infrastructure or further engineering of recycling processes since most research is based on cumulated or process unit results. The modeling of single processes of the entire recycling route is missing or based on outdated process technologies (Dunn et al., 2012; Rinne et al., 2021). Moreover, many studies consider the recycling of battery cells, which is not the best reference for companies who need to handle battery packs at the beginning. Therefore, a transparent and detailed MEFA, as well as subsequent environmental and economic assessments of industrial LIB recycling processes are conducted in the following.

3. Modeling and assessment approach

The modeling and assessment approach for this study is based on MEFA and LCA, extended by an economic assessment. The MEFA method extends the material flow analysis, which investigates the material flows entering into, passing through, and leaving out of the system, by energetic flows. Based on the conservation of materials, all incoming and outgoing material flows of a defined process must be in equilibrium (Torres et al., 2008). The study follows the four steps of an LCA: goal and scope definition, inventory analysis, impact assessment, and interpretation.

3.1. Goal and scope

The study is performed to provide transparent and comparable insights regarding the environmental and economic impacts of state-of-the-art industrial LIB recycling routes. The results can be used by industry and research to assess the resource flows on the single process

Table 2
Composition of an EV battery pack with 95 kWh capacity (BatPaC, Version March 2022)

Component	Material	Mass [kg]	Mass share [%]
Cells		349.28	60.6
Cathode	Aluminum foil	15.44	2.7
	Nickel	49.13	8.5
	Cobalt	16.44	2.9
	Manganese	15.33	2.7
	Lithium	9.68	1.7
Anode	Copper foil	39.85	6.9
	Graphite	81.65	14.2
Pouch	Aluminum[a]	0.00	0.0
Separator	Plastics	10.85	1.9
Electrolyte	Electrolyte	54.24	9.4
Miscellaneous	Others	56.67	9.8
Modules		32.20	5.6
	Aluminum	12.04	2.1
	Steel	16.86	2.9
	Plastics	2.38	0.4
	Electronics	0.92	0.2
Pack		194.89	33.8
	Aluminum	145.64	25.3
	Steel	36.5	6.3
	Copper/Tin	9.11	1.6
	Electronics	3.60	0.6
	Others (e.g. seals, elastomer)	0.4	0.0
Total		576.38	100.00

[a] The mass of aluminum of the pouch is not specified separately in BatPaC data but can be customized in the tool.

level and to understand the related environmental and economic impacts. Furthermore, the results can be used to initiate process improvements and to promote decision-making of car and battery manufacturers as well as recyclers regarding investments in recycling infrastructure.

The functional unit of the study is the processing of one ton of spent lithium-ion traction batteries from electric vehicles to recover the contained materials. The related reference flow is 25,000 t of spent LIBs at the recycler per yr. In the study, we evaluate the recycling of a battery pack with a capacity of 95 kWh from the BatPaC model. It consists of pouch cells with an NMC-622 cathode and a graphite-based anode (Table 2). Note that the module and pack periphery of the considered battery has a rather high mass share of (39% of total pack mass), compared to 29% for the NMC-622 battery used in the GREET model (Dai et al., 2018).

The system boundaries for the assessment are illustrated in Fig. 3. The foreground system comprises the discharging and disassembly of the batteries, different pretreatment processes, and hydrometallurgical material recovery. These processes are assumed to be carried out in Germany. The entire assessments including material and energy prices, investments, and GHG emission calculations are based on the yr 2021. It is assumed that the process capacity is fully utilized. The background system comprises auxiliary processes that are derived from generic datasets of the ecoinvent 3.8 database. The construction of the recycling plants, as well as the collection of the spent LIBs, are not considered within the environmental assessment. However, the process equipment is considered for the economic assessment. A detailed description of the pyrometallurgical process route (further called Route 1), the mechanical process route (Route 2), and the thermal-mechanical process route (Route 3) follows in Section 3.2.

The 'avoided burden' method is used, giving environmental credits for recycled materials that can replace virgin battery materials. It considers the avoided production of virgin materials (Fig. 3 bottom right) and the avoided alternative end-of-life paths (e.g., landfill). For recycling to be environmentally sound, the credits for the avoided burdens should be larger than the impacts from the recycling process itself (Cerdas et al., 2018; Geyer et al., 2016). In this context, the recovered materials are considered equivalent to virgin battery-grade materials and are fully taken into account accordingly (100% substitution). The slag utilization without further hydrometallurgical treatment as construction material (Routes 1 and 3) is not considered as recycled content and does not create economic or environmental costs or revenues (burden-free). The calculations of the GHG balances of the used and avoided resources are based on the ecoinvent 3.8 database using the impact category IPCC 2013, climate change, GWP 100a, cut-off. The detailed data are provided in Sections SM.5 and SM.6 including assumptions regarding the considered material types and recycling processes. A second indicator for the assessment is the recovery rate, also called recycling efficiency. The total recovery rate is the proportion of all materials recycled to the overall mass input. Additionally, the material-specific recovery rates indicate the proportion of the mass input that is recovered.

For the economic assessment, the impacts are calculated differently. While the foreground system remains the same, the impacts or profit are not specified by the avoided burdens rather than by the market conditions. In this context, a gate-to-gate consideration is applied. Within the

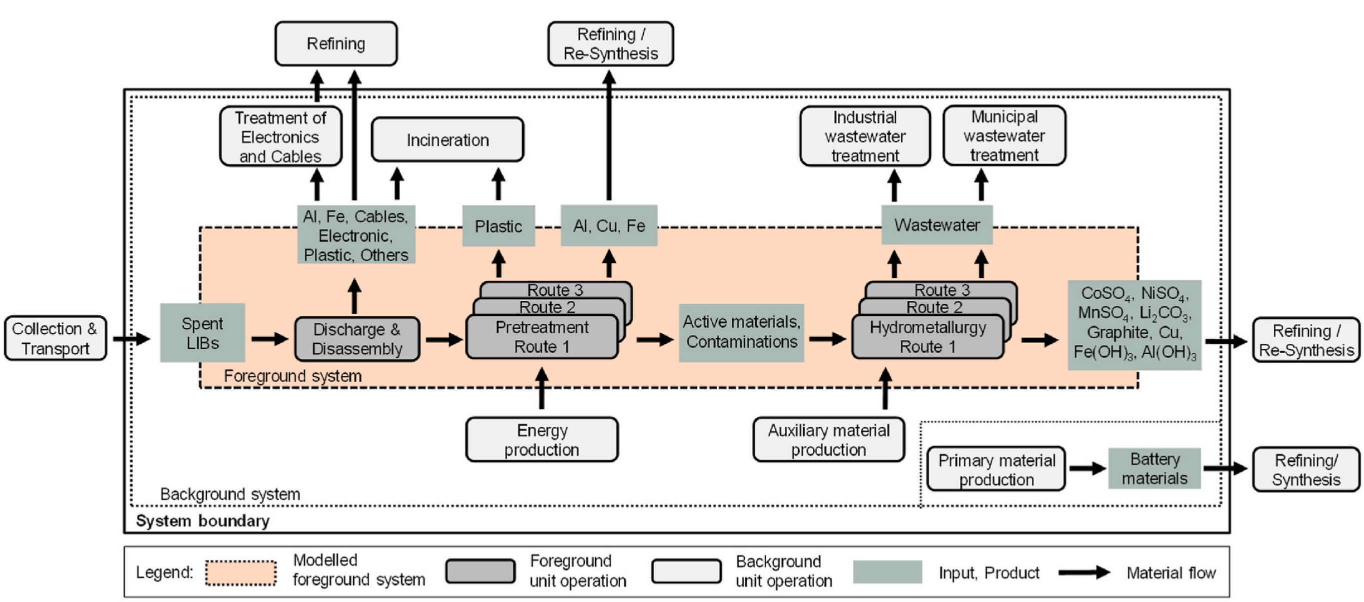

Fig. 3. System boundaries for the analysis of different battery recycling routes.

economic assessment, market prices/costs for all input and output factors as well as resources are multiplied with the respective quantities. Furthermore, costs for machinery and labor are considered, whereas costs for property and buildings are neglected. For the estimation of the investments for recycling equipment, economies of scale are taken into account (Hamelinck et al., 2004). The initial investment with an initial capacity is either taken from industrial reference offers, previous projects, or literature. While prices are determined for the year 2021, some investments are taken from the year 2016. Therefore, price indices are used to scale the investments to the reference year. We used the price index for investment goods in Germany for 2016 (100.6) and 2021 (107.7) (Statistisches Bundesamt, 2021). Based on the needed investment of a machine, the annual depreciation is calculated. Here, a depreciation period of five years is assumed. The annual maintenance costs of a machine are assumed to correlate with the initial investment. Therefore, a predefined maintenance rate between 2% and 5% of the needed investment is used (Thies et al., 2018). For the calculation of the labor and material costs, further assumptions are necessary. The entire recycling plant is running 330 days a year. Due to the high demand for workers and the resulting high costs of the night shift, the disassembly operates in two shifts with 8 h per day while the pretreatment and hydrometallurgical processes are operated in three 8 h shifts per day. Last, an overhead rate of 17% is considered (Dai et al., 2019). However, it should be noted that the overhead rate varies widely. All costs and investments are given in EUR. For values given in USD, an exchange rate of 0.87 EUR = 1 USD is used (date October 06, 2021).

3.2. Life cycle inventory

The life cycle inventory (LCI) for the three recycling routes is derived from a MEFA. As a basis for the MEFA, the three industrial recycling routes with their individual processes and input and output flows are illustrated in Figs. 4–6 and described below. LIBs arriving at the recycler are usually deeply discharged to deactivate the cells and to recover the contained energy. In the next step, the deep-discharged battery packs are disassembled to separate the pack periphery from the battery modules. The pack periphery mainly consists of housing, battery management, electronic components, cables, and connectors. These components are sorted and forwarded to established recycling routes. During manual disassembly of the battery system, more than 30% of the battery's total mass is already separated. In Route 3, the battery modules are further disassembled to cell level. In this step, the module housing, electronics, and cooling are recovered. Based on experiments, an average disassembly time of 2 h for Routes 1 and 2 and 3 h for Route 3 with two workers is assumed.

Fig. 4 depicts the pyrometallurgical Route 1. It is divided into three stages, disassembly and pyrometallurgical pretreatment (based on Verscheure et al. (2014)), the hydrometallurgical processing of the alloy, and optionally, the hydrometallurgical processing of the slag. The fraction balance of the furnace is provided in Table SM.2.4. The main output is slag, which consists of limestone, silica sand, aluminum, lithium, manganese, and approximately 70% of the contained iron. Cobalt, nickel, copper, and the remaining 30% of the iron are contained in the alloy. According to Verscheure et al. (2014), the flue dust contains up to 14% of the lithium contained in the battery. However, a further treatment process is not considered here.

Fig. 5 shows the mechanical Route 2. It is divided into a mechanical pretreatment (based on Hanisch et al. (2016) and Kwade and Diekmann (2018)) and a hydrometallurgical recovery of the active materials. This process is commonly known as a 'cold' recycling since the process temperatures are rather low compared to pyrolysis and pyrometallurgical processes. This facilitates the further recovery of organic components such as plastics, electrolyte, and graphite by different crushing and classification processes. The disassembled modules are crushed in an inert atmosphere before the materials are separated based on their physical properties by various classification processes such as zick-zack-sifter and sieving. After the crushing process, a thermal treatment evaporates the volatile components (originating mainly from the electrolyte), which are then condensed and supplied to the chemical industry. In the sieving process, the active materials are separated as "black mass" and further processed in hydrometallurgical processes. The sieve overflow contains the collector foils and the separator which are further separated in a zick-zack-sifter.

Fig. 6 describes the hybrid Route 3. The recycling route is divided into a thermal-mechanical pretreatment (based on Accurec GmbH (2018), Hanisch et al. (2016), and Sojka et al. (2020)), followed by a pyrometallurgical treatment (based on Verscheure et al. (2014)), and a hydrometallurgical recovery of the active materials. The disassembled modules or cells are pyrolyzed to deactivate the cells for further mechanical treatment by removing safety-critical volatile compounds. Next, the cells are crushed before the materials are separated based on their physical properties. The black mass of active materials is separated in a sieving process before the contained metals are separated in a subsequent pyrometallurgical process following Route 1. Lithium can also be recovered via an evaporation process prior to pyrometallurgy, which is not realized in the industry yet (Elwert et al., 2018).

The hydrometallurgical processing in all three routes is derived from a patent of Duesenfeld (Hanisch et al., 2019). It not only contains very current and detailed data but it is also assumed that the hydrometallurgy has to be robust by separating impurities from the targeted active materials. Furthermore, the processes were adapted to different input materials. In general, the hydrometallurgical treatment of the black mass can be classified into four steps. First, the solid material is leached in sulfuric acid. If necessary, this is carried out with additional heat to remove fluorine components that are harmful to subsequent processes and output qualities. In a second step, impurities such as iron, aluminum, or slag formers are precipitated. Later, the lithium is also recovered by precipitation. The third step is the key process in which the target metals are separated by solvent extraction. Specific solvents, e.g. D2EHPA, Cyanex 301 GN, and Cyanex 272, are used to separate materials with similar physical properties in high quality. In the fourth step, the extracted materials are converted to salts by crystallization. The recovered metal salts include a defined water fraction (see Table SM.2.3) and can be used for new active material production.

Table 3 provides a list of the cumulated material and energy flows for Routes 1–3. Corresponding material Sankey diagrams are provided in Section SM.4. The baseline scenario corresponds to the three state-of-the-art recycling routes with an annual capacity of 25,000 t spent LIBs, not considering slag recycling at Routes 1 and 3 and electrolyte recovery in Route 2. The advanced recycling routes further include an additional slag treatment in Routes 1 and 3, and an additional electrolyte recovery in Route 2 to recover more materials compared to the baseline routes. The MEFA diagrams and the LCI show clearly that particularly the hydrometallurgy has large mass flows. Especially, the quantities of sulfuric acid, water, and sodium hydroxide, which account for a large proportion of the total mass, are noteworthy. The Sankey diagrams illustrate the additional efforts within Routes 1 and 3 since two separate hydrometallurgies with cumulatively larger mass flows are required there.

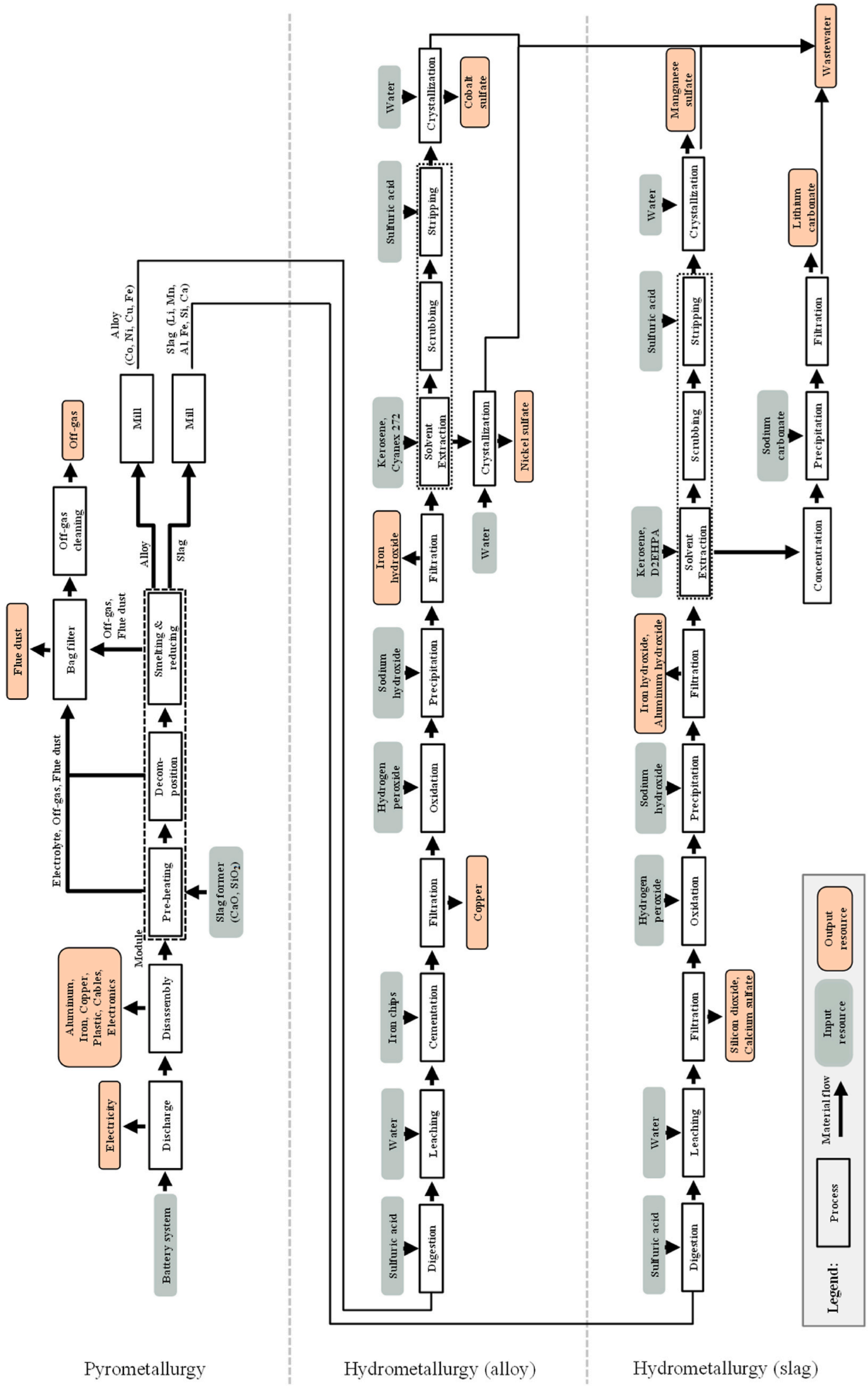

Fig. 4. Pyrometallurgical and hydrometallurgical process chain (Route 1) based on Hanisch et al. (2019) and Verscheure et al. (2014).

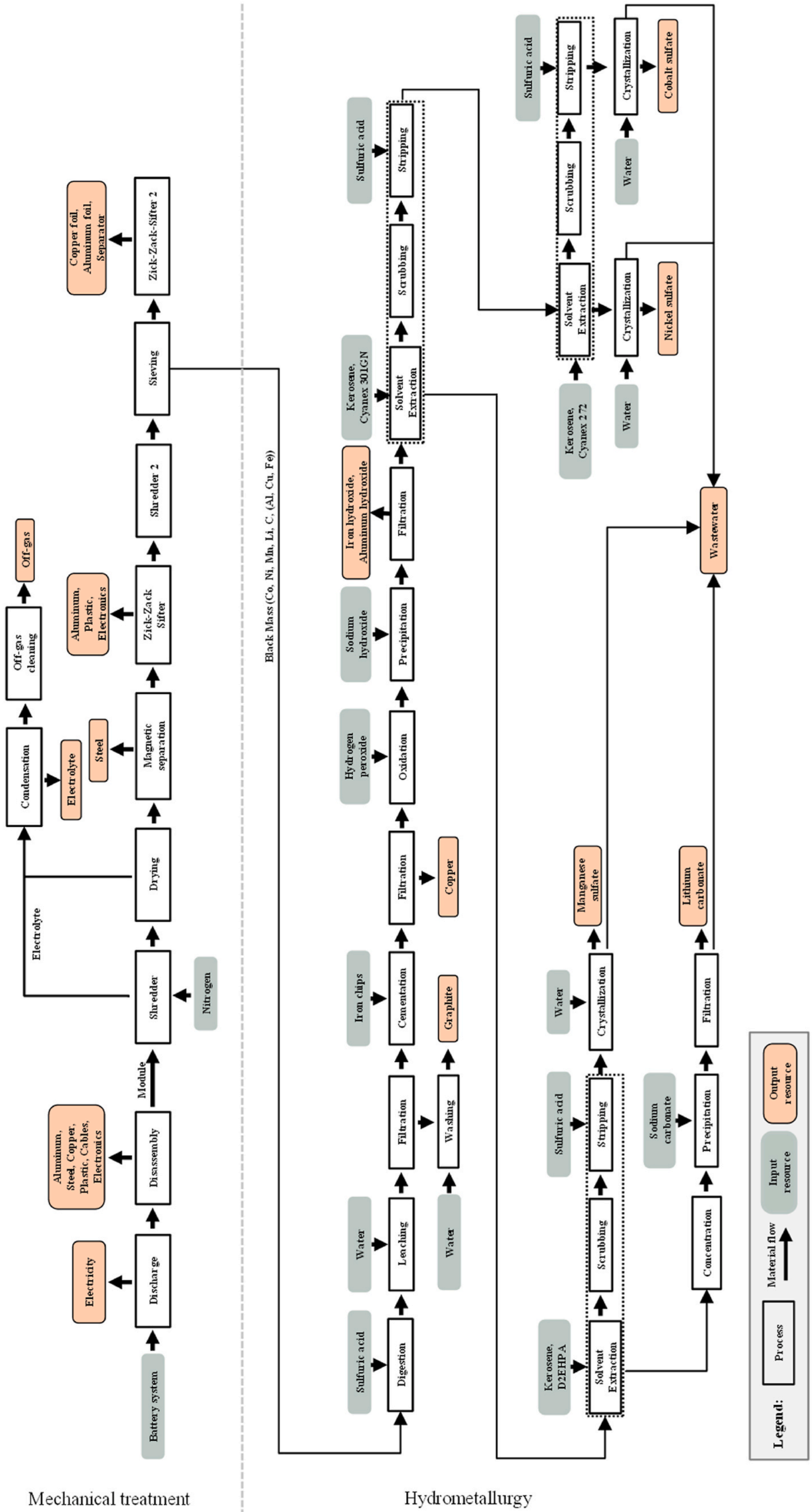

Fig. 5. Mechanical and hydrometallurgical process chain (Route 2) based on Hanisch et al. (2016, 2019).

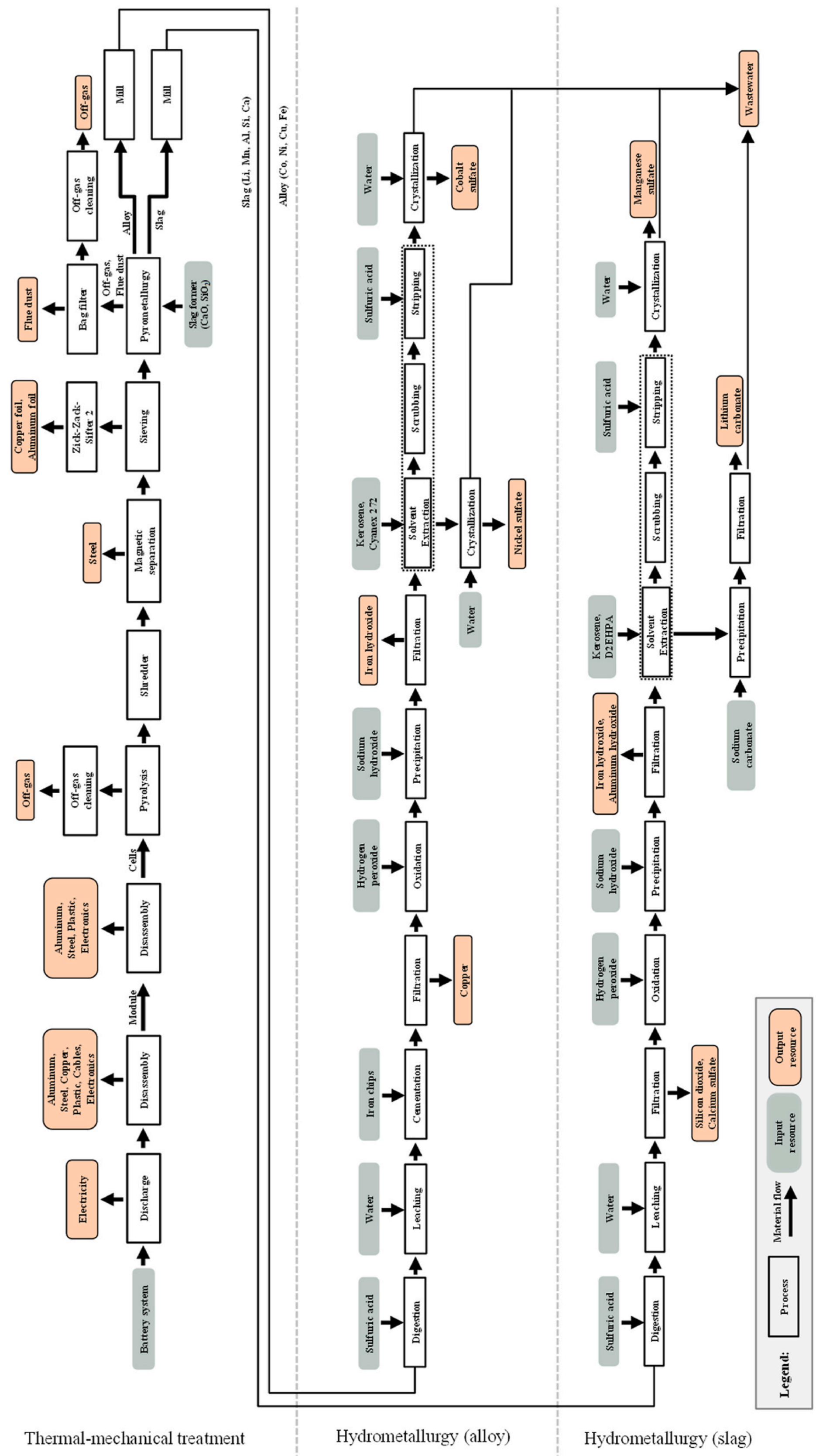

Fig. 6. Thermal-mechanical and hydrometallurgical process chain (Route 3) based on Hanisch et al. (2016) and Sojka et al. (2020).

Table 3
LCI of the three recycling routes for baseline and advanced scenario for 25,000 t spent LIB input per yr. The colors show the material-specific comparison of the treated mass in the different recycling routes. Net resource flows of water and electricity consider a circulation in the recycling system. With incin. = incineration, ind. = industrial, mun. = municipal.

Substance	Unit	Route 1 baseline	Route 1 advanced	Route 2 baseline	Route 2 advanced	Route 3 baseline	Route 3 advanced
Input							
Spent LIBs	t	2.50E+04	2.50E+04	2.50E+04	2.50E+04	2.50E+04	2.50E+04
Nitrogen	m³	-	-	3.46E+05	3.46E+05	-	-
Limestone	t	7.01E+03	7.01E+03	-	-	3.22E+03	3.22E+03
Silicon	t	3.63E+03	3.63E+03	-	-	1.67E+03	1.67E+03
Oxygen	t	2.74E+03	2.74E+03	-	-	1.26E+03	1.26E+03
Sulfuric acid	t	7.81E+03	3.61E+04	7.71E+03	7.71E+03	4.63E+03	1.57E+04
Process water (net)	t	4.51E+03	5.38E+03	1.15E+04	1.15E+04	1.64E+03	1.84E+03
Iron	t	1.67E+03	1.67E+03	3.30E+01	3.30E+01	1.70E+01	1.70E+01
Hydrogen peroxide	t	3.61E+03	6.09E+03	1.34E+03	1.34E+03	1.28E+03	2.01E+03
Sodium hydroxide	t	1.56E+04	7.22E+04	1.53E+04	1.54E+04	9.24E+03	3.14E+04
Kerosene	t	2.80E+00	5.80E+00	4.00E+00	4.00E+00	1.30E+00	2.44E+00
Cyanex 272	t	1.30E+00	1.30E+00	3.00E-01	3.00E-01	6.10E-01	6.10E-01
Cyanex 301GN	t	-	-	1.20E+00	1.20E+00	-	-
D2EHPA	t	-	1.40E+00	7.00E-01	7.00E-01	-	5.40E-01
Sodium carbonate	t	-	3.03E+03	2.09E+03	2.09E+03	-	2.58E+03
Natural gas	m³	1.86E+06	1.86E+06	-	-	8.57E+05	8.57E+05
Electricity (net)	kWh	3.91E+07	7.85E+07	1.73E+07	1.74E+07	1.64E+07	1.89E+07
Output							
Aluminum	t	6.25E+03	6.25E+03	7.39E+03	7.39E+03	7.41E+03	7.41E+03
Steel	t	1.57E+03	1.57E+03	2.28E+03	2.28E+03	2.29E+03	2.29E+03
Copper	t	1.95E+03	1.95E+03	2.09E+03	2.09E+03	2.08E+03	2.08E+03
Electronic scrap	t	1.55E+02	1.55E+02	1.91E+02	1.91E+02	1.55E+02	1.55E+02
Electrolyte		-	-	-	1.86E+03	-	-
Graphite		-	-	2.95E+03	2.95E+03	-	-
Iron hydroxide	t	3.22E+03	4.10E+03	5.88E+01	5.88E+01	2.45E+01	2.45E+01
Aluminum hydroxide	t	-	3.02E+03	3.57E+01	3.57E+01	-	1.43E+01
Kerosene	t	2.76E+00	5.81E+00	3.99E+00	3.99E+00	1.23E+00	2.44E+00
Cyanex 272	t	1.18E+00	1.18E+00	3.00E-01	3.00E-01	5.52E-01	5.52E-01
Cyanex 301GN	t	-	-	1.13E+00	1.13E+00	-	-
D2EHPA	t	-	1.30E+00	6.00E-01	6.00E-01	-	4.92E-01
Nickel sulfate	t	8.98E+03	8.98E+03	7.95E+03	7.95E+03	8.36E+03	8.36E+03
Cobalt sulfate	t	3.17E+03	3.17E+03	2.81E+03	2.81E+03	2.92E+03	2.92E+03
Manganese sulfate	t	-	1.91E+03	1.71E+03	1.71E+03	-	1.34E+03
Lithium carbonate	t	-	1.41E+03	1.62E+03	1.62E+03	-	1.02E+03
Calcium sulfate	t	-	2.10E+04	-	-	-	9.65E+03
Silicon	t	-	3.52E+03	-	-	-	1.62E+03
Waste (incin.)	t	1.72E+00	1.72E+00	4.34E+03	4.34E+03	1.15E+03	1.15E+03
Sewage (ind.)	t	-	1.97E+04	1.83E+04	1.83E+04	-	2.79E+04
Sewage (mun., net)	t	1.94E+04	7.11E+04	-	-	-	-
Electricity (usage)	kWh	2.17E+05	2.17E+05	2.17E+05	2.17E+05	2.17E+05	2.17E+05

Legend: rel. more advantageous recycling route — rel. less advantageous recycling route

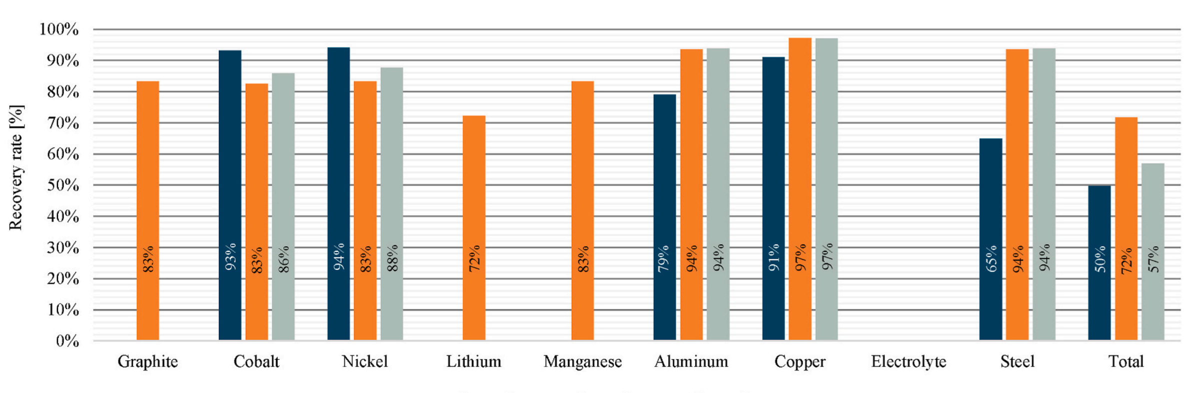

Fig. 7. Total and material-specific recovery rates of three baseline battery recycling routes.

4. Impact assessment results and interpretation

The results of the impact assessment are presented and discussed as follows. First, a baseline scenario with currently performed battery recycling routes is assessed. Next, scenarios regarding the process configuration and recovered materials, process capacity, and prices developments are evaluated and discussed to identify critical parameters and future process improvements. Last, the results are validated based on previous studies.

4.1. Baseline scenario

Fig. 7 illustrates the achievable recovery rates of the three recycling routes in the baseline scenario. While the EU legal minimum recovery rate of 50% is achieved in all routes, the individual values vary. Route 2 offers the highest total recovery rate of approximately 72% because all active materials are recovered. In contrast, only the valuable metals cobalt and nickel, as well as the commodity metals aluminum, copper, and steel, are recovered in Routes 1 and 3. This is also reflected in the different material-specific recovery rates. The highest recovery rates for cobalt and nickel are achieved by Route 1, mainly due to the high efficiency of the pyrometallurgical separation process. Routes 2 and 3 are characterized by incurring losses of active materials throughout the mechanical processing and especially the sieving process. Nevertheless, these routes achieve higher recovery rates for aluminum and copper, because of the higher disassembly depth and the early recovery in the mechanical separation step. Furthermore, only Route 2 allows for the recovery of graphite, manganese, lithium in the baseline scenario due to its low process temperatures which consequently affects positively the total recovery rate.

The recovery rates also affect the results of the environmental and economic assessment. As seen in Fig. 8, Route 3 presents the lowest investments required, closely followed by Route 1. Route 2 requires the highest investment due to the more complex pretreatment and the hydrometallurgical treatment of all active materials. Hence, the depreciation and maintenance of Route 2 results in higher costs. Nevertheless, the total costs for all three routes are similar. For Route 1, the large mass flows in the hydrometallurgy due to the high amount of supply materials for copper and iron recovery lead to a rather high consumption of electricity for crystallization. Hence, electricity costs are up to three times higher. For Route 3, the deeper disassembly leads to significantly higher labor costs. With more recovered materials in Route 2, approximately 17% higher revenues can be realized, making Route 2 with approximately 3330 € profit per ton of battery input the most economic.

All recycling routes have the potential to avoid a significant amount of environmental impacts (credits minus effort). Route 3 achieves similar climate impact credits to Route 2 of approximately 7 t CO_2-eq. per ton spent LIB, but with lower efforts due to the smaller mass flows in hydrometallurgy with resulting lower energy and operating materials expenditures. Route 1 reaches the lowest avoided environmental impact because of fewer recovered materials. Detailed analyses for individual resource flows are provided in Fig. SM.7.3.

4.2. Extended recoverable materials

The baseline recycling routes can be extended for better material recovery. The recovery rates of these advanced recycling routes in comparison to the baseline recycling routes are illustrated in Fig. 9. With the slag treatment in Routes 1 and 3, lithium and manganese are additionally recovered. Moreover, the recovery rate of aluminum is

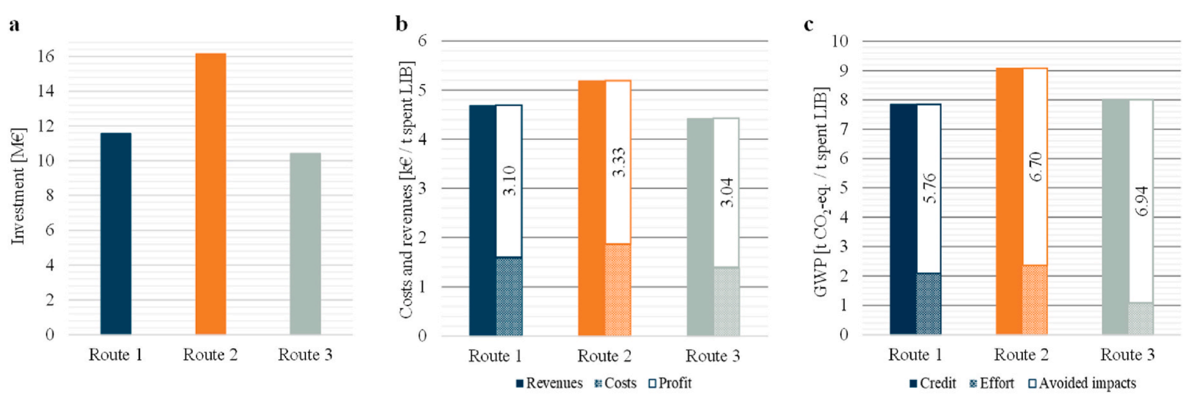

Fig. 8. Assessment of three baseline battery recycling routes regarding a) needed investment, b) costs and revenues, and c) climate impacts.

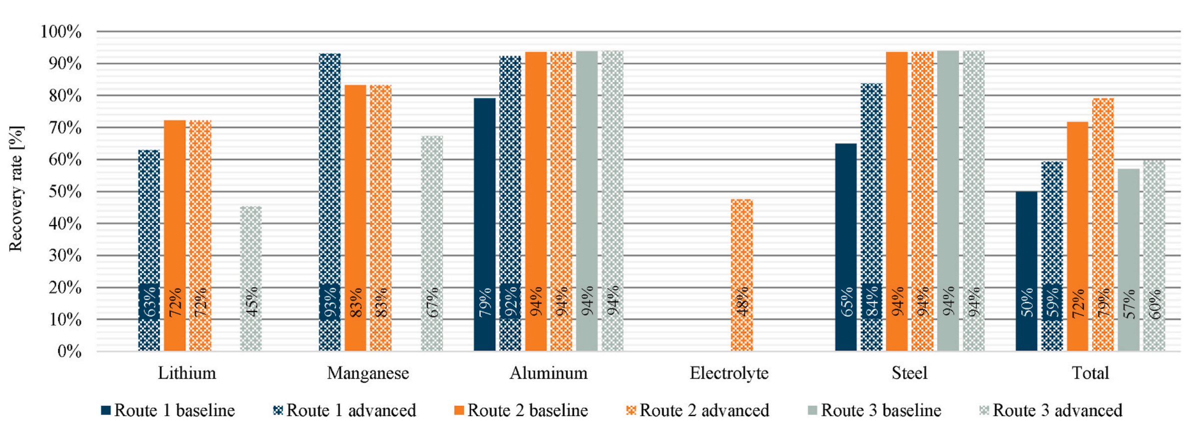

Fig. 9. Total and selected material-specific recovery rates of three advanced battery recycling routes.

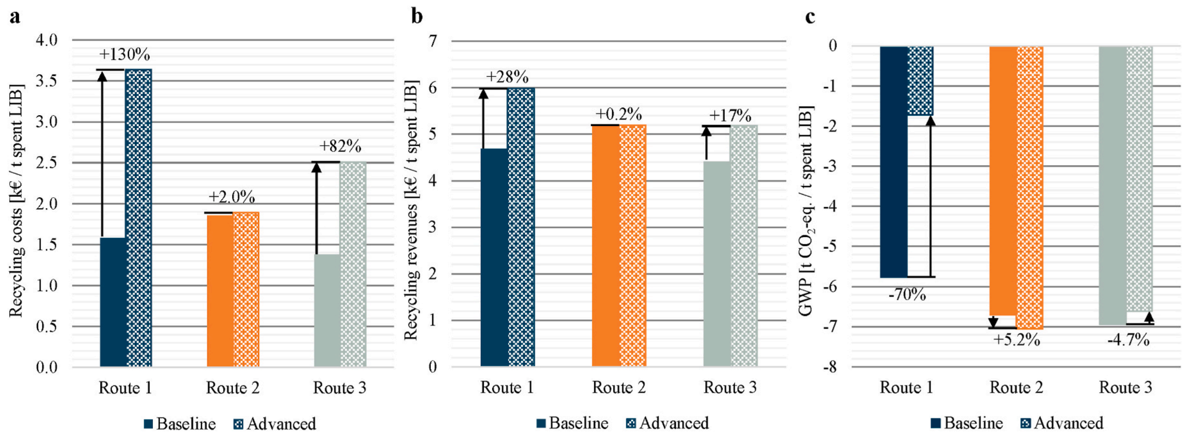

Fig. 10. Route specific assessment of three baseline and advanced battery recycling routes regarding a) costs, b) revenues, and c) climate impacts.

increased. Route 2 still achieves the highest recovery rate for lithium since it avoids the losses inherent to pyrometallurgical processing. Accurec counters this circumstance with the plan of early-stage separation of lithium via evaporation (Sojka et al., 2020). The hydrometallurgical treatment of Route 3 is more efficient due to fewer impurities and slag formers. Furthermore, Route 2 recovers the electrolyte partially, which has a considerable impact on the total recovery rate due to its high mass fraction.

The economic and environmental effects of extending the recoverable materials are illustrated in Fig. 10. The slag treatment has significant effects on the expected investments and operating costs. Especially the slag treatment in Route 1 must process larger material flows with corresponding extended demand for infrastructure, operating resources, and energy. In this context, the deeper disassembly of Route 3 compared to Route 1 is an advantage since much less slag is produced resulting in lower amounts of acid, base, and reducing agent in Route 3 (Table 3). This results in economic advantages of Route 3 over Route 1. Furthermore, the higher revenues justify the additional expense in Route 3, while Route 1 becomes less profitable. In the environmental assessment, both advanced Routes 1 and 3 perform worse. The implementation of an electrolyte recovery in Route 2 is rather inexpensive but does not result in significant advantage from an economic point of view. However, from an environmental perspective, electrolyte recycling is beneficial. As a basis for process-specific engineering, the economic and environmental influence of the respective recycling processes and resource flows are important. A detailed assessment is provided in Fig. SM.7.1–3. It shows that the climate impacts are concentrated particularly on precipitation and evaporation processes due to their high demand for process energy and supply material.

The environmental credits of the recycling depend largely on the material substitution factor of the secondary material in relation to the primary material, a proxy for secondary material quality. Decreasing the substitution factor for the electrode coating materials (Co, Ni, Mn, Li, Graphite) from 1.0 to 0.75 and 0.5 increases the GWP of the recycling by 20% and 41% respectively for Route 2. No reuse of the mentioned materials (SF = 0) still results in an environmental beneficial recycling, due to the high quantities of Al, Cu recovered. Further analysis of the substitution factor of Route 1–3 is provided in Fig. SM.8.1.

4.3. Process capacity

The second scenario focuses on the influence of the capacity of the recycling routes on the environmental and economic performance. In general, higher capacity is assumed to result in lower input-related energy consumption and investments, since the process periphery and infrastructure requirements increase disproportionately slower than capacity. To evaluate these economies of scale, three process capacities are investigated, namely 2,500, 25,000, and 75,000 t of spent LIBs per yr. The three process capacities have been defined together with the industry partner whereby 2,500 t per yr reflect a pilot scale, 25,000 t per yr a medium industrial scale, and 75,000 t per yr a large industrial scale recycling. The stoichiometry of the hydrometallurgy remains unchanged because it is independent of process capacity. In Fig. 11a, the route-specific investments are illustrated. Naturally, they increase degressively with higher capacity. Fig. 11b shows the change in costs per ton of input, which decrease considerably in all routes from the pilot to industrial scale. Consequently, the relative costs at industrial scale are less than half of those at pilot scale. While the depreciation per capacity unit decreases at a similar magnitude for all routes, Route 1 shows the largest relative cost reductions. This is because the depreciation effect is not

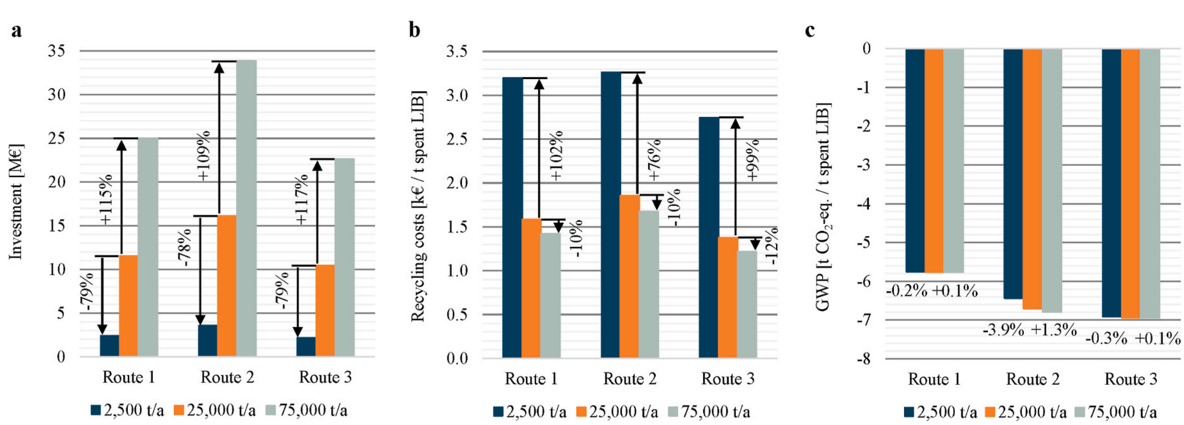

Fig. 11. Route specific assessment of three baseline battery recycling capacities regarding a) needed investment, b) costs, and c) climate impacts.

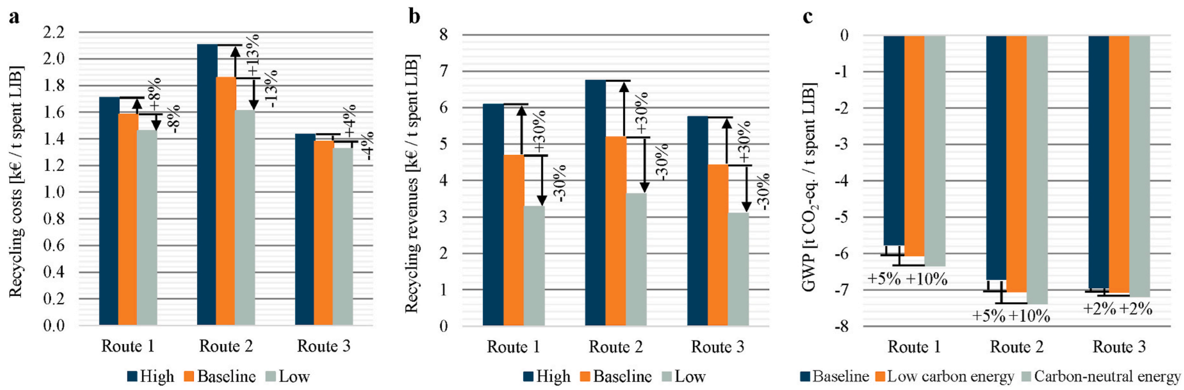

Fig. 12. Route specific assessment of three baseline battery recycling routes regarding a) costs for different material prices, b) revenues for different material prices, and c) climate impacts for different energy carbon intensities.

superimposed by cost effects (e.g., staff), which are at a lower level in Route 1. Due to the high economic impact of the hydrometallurgy and the pronounced economies of scale, large centralized facilities can achieve better economic performance. However, the relatively lower investments per capacity unit need to be balanced with the higher transportation costs. In order to determine the minimal capacity of a recycling plant from an economic point of view, we conducted a break-even analysis. For Route 1, 2, and 3 the minimal capacity is 1,350, 1,100, and 1,170 t per yr respectively. The capacity of Route 1 needs to be higher to achieve break-even due to the high investment in hydrometallurgy and the simultaneous low revenues from recovered materials. However, it should be noted that investment in property and buildings are neglected. Hence, the implemented capacity needs to be greater. The amount of avoided environmental impacts increase in all routes with higher capacities, however, with just minimal increases in Routes 1 and 3 due to the linear relationship of the pyrometallurgical expenses (Fig. 11c). In general, it can be stated that larger recycling plants are both economically and environmentally beneficial. However, as assumed here, a high capacity utilization must be ensured, which seems to be achievable only in the medium term.

4.4. Material price and electricity mix

Besides process capacity, the material and energy market is considered to influence the recycling performance. Especially the prices of battery active materials have been very dynamic recently (US Geological Survey, 2021). The influence of material prices on recycling is investigated by three scenarios regarding the procurement of operating materials and the sales of secondary materials. The baseline scenario contains customary market prices from October 2021. For the “high” and “low” scenarios, cost variations of ±30% are assumed. The electricity price is assumed to be constant at 16.54 € ct/kWh, reflecting the average industry price for electricity in Germany in 2020 (Bundesnetzagentur, 2020).

The results in Fig. 12b show that the revenues are directly related to the material prices. However, costs increase/decrease by less than 30% since costs for labor and depreciation are not influenced by the material prices (Fig. 12a). Therefore, the increase in revenues exceeds the increase in costs, leading to increased profits. In general, higher material costs have a positive influence on the battery recycling industry. Consequently, increasing future demand for active materials may increase the price and further improve profitability.

In addition, the influence of the energy transition on the environmental impact of recycling is investigated. The German energy mix for 2020 had a CO_2 intensity of 366 g CO_2-eq./kWh (Icha et al., 2021). The “low carbon energy” scenario assumes a 50% reduction in CO_2 intensities, while the ‘carbon-neutral energy’ scenario assumes electricity use from 100% renewable sources. As illustrated in Fig. 12c, the energy mix has the lowest impact in Route 3, as rather small volume flows are treated and accordingly less electricity is required. In Route 1, the impacts are greater because the hydrometallurgical treatment is larger. The electricity mix has the greatest impact on Route 2 whereby the impact on pyrometallurgical processes is minor, as natural gas is used as the main energy supplier. For the recycling industry, the choice of the energy mix is a simple improvement opportunity with a potential of 2–10% lower environmental impacts per ton of input. A potential simultaneous reduction of the environmental impacts of primary material production was not considered.

4.5. Comparison with previous studies

This subsection critically compares the results of the assessment with the results from previous studies. Since literature presents no comparable, comprehensive, and transparent results (Fig. 2), environmental and economic indicators are validated separately. The underlying values can be taken from Fig. 13.

Ciez and Whitacre (2019) conclude that hydrometallurgical processing has environmental advantages, while pyrometallurgical processing does not pay off environmentally. In our study, all recycling routes pay off and have a significantly higher amount of avoided

Source	Value	Route	Recyling process	Value source	Value assessment	Remark	Assessment dimension
Mohr et al. 2020	Environmental impacts	1	D P H	12 kg CO_2-eq./kWh	35 kg CO_2-eq./kWh	Source on cell level	ENV
Mohr et al. 2020	Environmental impacts	2	D M H	22 kg CO_2-eq./kWh	41 kg CO_2-eq./kWh	Source on cell level	ENV
Mohr et al. 2020	Recycling efficiency	2	D M H	85 to 100%	79%	Source on cell level	ENV
Hoyer et al. 2015	Investment	2	D M	€ 5.8 million	€ 6.1 million	Average degression coefficient 0.72	ECON
Hoyer et al. 2015	Investment	2	H	€ 14.0 million	€ 10.1 million	Average degression coefficient 0.63	ECON
Dai et al. 2019	Profit	1	D P H	0.96 €/kg battery	3.10 €/kg battery		ECON
Dai et al. 2019	Profit	2	D M H	1.35 €/kg battery	3.33 €/kg battery		ECON
Nguyen-Tien et al. 2022	Profit	2	D M H	2.40 €/kg battery	3.33 €/kg battery	Source on cell level	ECON

Legend:
D Dismantling P Pyrometallurgy M Mechanical H Hydrometallurgy ENV Environmental ECON Economic

Fig. 13. Validation of the assessment results in comparison to previous studies.

impacts. The derivation can be justified by the different products under investigation, battery cells and battery packs. The recycling of battery packs leads to higher environmental credits due to the relatively easy disassembly and the recovery of a large mass fraction of aluminum casing (Thies et al., 2018). Mohr et al. (2020) have evaluated a pyrometallurgical and mechanical-hydrometallurgical LIB recycling on cell level comparably to the recycling routes of the model here. In our study, Routes 1 and 2 achieve a larger amount of avoided impacts, which is reasonable for the same arguments. The overall recycling benefit in the study of Mohr et al. is dominated by aluminum, copper, and nickel compounds. This is in line with the result of our model. However, the contribution from cobalt compounds is rated higher our study, which is likely because of the higher GHG emissions in the new cobalt dataset introduced with the ecoinvent 3.8 update. The reported recovery rates in their sensitivity analysis are comparable to the recovery rates achieved in this paper. The difference can be reasoned by the neglect of plastics recycling.

Hoyer et al. (2015) have assessed the necessary investments for a mechanical and hydrometallurgical process comparable to Route 2. Based on the results, we have scaled the investments to 25,000 tones spent batteries per yr according to the average degression factor. The investments for the disassembly and mechanical preparations are comparable to our results. However, the investments for hydrometallurgy differ. The difference can be reasoned by the technological improvements as well as the neglect of wastewater treatment. Within the EverBatt model, profits for the Routes 1 and 2 can be found. While both routes indicated economic profitability in both assessments, the results differ. We found three main reasons for the difference in the profits. First, the profits in our study are higher due to the neglection of costs for buildings and property, which makes up to 0.31 €/kg according to the EverBatt model (Dai et al., 2019). Second, research and development costs are neglected. Third, while both studies consider NMC-622 battery, the battery compositions vary significantly in size. Nguyen-Tien et al. (2022) have assessed a hydrometallurgical treatment and provide similar economic results which are 2.40 €/kg. The assessed recycling process bases on the EverBatt model with the scope of the UK in 2040. Due to the depreciation of investments over time the profit is increasing. Since our does not include cost of infrastructure the results of the two publications show similar results. Hence, although the results differ, valid reasons can be found. Therefore, the profits are valid considering these simplifications. Overall, the recycling routes are based on representative assumptions. Due to the transparent structure of the model, individual product and process adaptions can be made.

Noteworthy limitations of this study include the neglected costs for property and buildings in the economic assessment as well as the production and provision of the building and process infrastructure in the environmental assessment. Furthermore, the environmental assessment only considers the global warming potential and does not assess other impact categories. The secondary data used needs to be updated especially for the pyrometallurgical pretreatment. Therefore, it should be noticed that the real or planned recycling routes of the mentioned companies may differ. In general, future product, processes, and market developments will have a major influence on the potential of recycling and should be considered dynamically. In this context, the increasing use of LFP batteries poses new challenges regarding the process engineering as well as the economical advantageousness due to different materials used compared to NMC batteries. Therefore, future studies should include both the design of the recycling process and the economic assessment of LFP batteries. Further, a consideration of the output quality and associated potential substitution factors of primary materials should be further investigated.

5. Conclusion and outlook

The industrial recycling routes assessed in this paper achieve different economic and environmental performance, which is evaluated using a spreadsheet-based assessment tool developed for the automotive industry. Due to the modular structure, the tool enables future adaptation and thus continuous advancement of the recycling routes and processes. A detailed MEFA based on the single processes of the recycling routes is carried out and integrated into an economic and environmental assessment under consistent and comparable system boundaries and assumptions. The results show that the considered industrial recycling routes are economically and environmentally advantageous compared to the production of virgin battery materials and therefore contribute to a cleaner production of batteries. The assessed routes realize profits of 3000 to 3300 €/t and avoided impacts of 5.0–7.0 t CO2-eq./t spent LIB. A higher disassembly depth in Route 3 results in large environmental credits and has positive economic effects, which is mainly due to the smaller dimensions of the subsequent process steps. However, the process chain becomes more complex with reduced profit. An exclusively pyrometallurgical pretreatment does not enable lithium and manganese recovery, whereas a later recovery of lithium through hydrometallurgical processing of slag reduces the economic benefit and amount of avoided environmental impact due to the need to treat large material flows. This will become problematic in the future if material-specific or higher total recovery rates are enforced, as currently planned in the new battery regulation of the European Union. Besides, it is shown that process capacity and material prices have a high influence on the economic performance of recycling. In this context, a goal conflict between large, centralized facilities and increasing transportation costs exist. This is particularly problematic since spent batteries need to be transported as hazardous goods with corresponding high costs. Due uncertainties concerning the future environmental legislation and the market development, it is important to establish a flexible and expandable recycling infrastructure that ensures high-capacity utilization corresponding to expected future market growth. The lack of standardization and the rapid development, especially regarding electrode composition, pose additional challenges for recycling. It is therefore important for recyclers to consider the dynamic market with product and process development and to ensure homogeneous input streams into the recycling process.

Avenues for future research are the systemic and global relationships between recycling, production, and suppliers, especially when multiple actors are involved. In this context, the combination of supply chain management, sustainability assessment, and process engineering is crucial to achieve a sustainable circular economy for LIBs. Here, advancement potentials are often difficult to quantify due to interdependencies between recycling processes and resulting product qualities. These can be represented and made quantifiable by combining physical models with a subsequent environmental and economic assessment. Finally, the influences of product design on recycling will become particularly interesting when the battery producer also acts as the subsequent recycler.

CRediT authorship contribution statement

Steffen Blömeke: Conceptualization, Methodology, Writing – original draft, Investigation, Visualization, Validation, Software. **Christian Scheller:** Conceptualization, Methodology, Writing – original draft, Investigation, Visualization, Validation, Software. **Felipe Cerdas:** Conceptualization, Methodology, Writing – original draft, Investigation. **Christian Thies:** Conceptualization, Methodology, Writing – original draft, Investigation. **Rolf Hachenberger:** Writing – review & editing, Resources, Methodology, Validation. **Mark Gonter:** Writing – review & editing, Resources, Methodology, Validation, Supervision. **Christoph Herrmann:** Writing – review & editing, Supervision, Resources, Funding acquisition. **Thomas S. Spengler:** Writing – review & editing, Supervision, Resources, Funding acquisition.

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 and model used in the manuscript are available at https://www.doi.org/10.5281/zenodo.6342832.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2022.134344.

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