The International Journal of Life Cycle Assessment
https://doi.org/10.1007/s11367-025-02478-z

DATA AVAILABILITY, DATA QUALITY

Process simulation-based LCA: Li-ion battery recycling case study

Yvonne Kaye Perocillo[1] · Eric Pirard[2] · Angélique Léonard[1]

Received: 3 October 2024 / Accepted: 17 April 2025

Abstract

Purpose Data availability remains a bottleneck for life cycle assessment (LCA) despite being an established method in evaluating potential environmental impacts. The use of digital process simulations (process simulation-based LCA) to generate data had been explored as a solution but mainly applied as an assessment tool. This study aims to identify the strengths and limitations of this approach by expounding on its potential to be an eco-design tool starting at the conception phase of a process.

Methods To achieve this, a hydrometallurgical and a pyrometallurgical recycling process were designed from literature and assessed in separate case studies. Recycling processes, especially for Li-ion batteries, are actively researched around the world today because of the critical metals it contains, and the role batteries play in achieving sustainability targets. A literature review of current recycling approaches for LIBs recycling was first conducted in order to design the processes to be assessed. Next, the goal and scope of the LCA case study were defined prior to simulating the chosen processes using HSC Chemistry®. Data generated from the simulation was used for the inventory, and the impacts were assessed using the Environmental Footprint (EF) 3.0 method in Simapro v9.6.

Results and discussion The results showed that sulfuric acid and process emissions contributed most to the potential environmental impacts of the simulated hydrometallurgical treatment, while coke for the pyrometallurgical recycling process. This aligns with known concerns for these processes, i.e., hydrometallurgy can be reagent intensive, and pyrometallurgy specially smelting use carbon-containing materials as reductant, which shows that digital simulations could be a good source of information for the LCA. Another strength of simulation is the possibility to conduct digital experiments that could be time-consuming in a laboratory. Thermodynamic feasibility of the process being designed could also be confirmed. However, reaction kinetics are not completely considered thus the need for data validation through actual experiments is a recommended next step for this approach.

Conclusion From the results, it can be concluded that the use of digital process simulation-based LCA can be a good approach in eco-designing processes. There is, however, a clear need for collaboration between process engineers and LCA practitioners to make sure that a well-informed digital simulation is used for the assessment, i.e., it should include validation of the simulation results based on the scaled-up data from laboratory and/or pilot-scale.

Keywords Life cycle assessment (LCA) · Eco-design · Process simulation · Battery recycling · Li-ion battery

1 Introduction

Life cycle assessment (LCA) is increasingly used nowadays to establish potential environmental impacts and/or benefits of products and processes from various industries. Initially conceptualized as an assessment tool (Bjørn et al. 2018), this method is also now used as an effective guide for eco-design, i.e., integrating environmental aspects into a product's or process' design (UNEP 2001). This is because the "life cycle thinking (LCT)" concept in which LCA is based on also governs eco-design.

Communicated by Benedetta Esposito.

✉ Yvonne Kaye Perocillo
yperocillo@uliege.be

[1] Products, Environment, and Processes (PEPs), Chemical Engineering, University of Liége, 4000 Liége, Belgium

[2] Georesources, Mineral Engineering and Extractive Metallurgy (GeMMe), University of Liége, 4000 Liége, Belgium

A complete LCA would be able to provide comprehensive information of the impacts of the raw materials extraction, the production and manufacturing processes, the use phase, and even up to the end-of-life (EOL). These are necessary in order to distinguish hotspots in the process in terms of environmental impacts and, in turn, make process and product design modifications while balancing these insights with economic, social, and even technical requirements. However, much like other assessment tools, data availability and quality is one of its common limitations.

When applying LCA in the metals and mining industry, the challenge of data availability has resulted in some unit processes becoming a “black box”, meaning that cumulative and/or ambiguous data are used during the assessment (Segura-Salazar et al. 2019). This is usually a result of high confidentiality within the industry or also could be the data needed for the LCA are not necessarily important in the daily operations of the plant and thus not commonly monitored (e.g., electrical consumption of small pumps). This could lead to results that have higher possibility of missing the main potential impact sources of the system being assessed. The earliest attempts found to address this weakness was in the late 1990 s in which digital solutions or the use of computer-related technologies were suggested to bridge the data gap (Spengler et al. 1998; Reuter 1998).

In 1998, Reuter demonstrated the potential of using computer simulations and models of different metallic production systems to address this weakness in LCA for primary resource processing (Reuter 1998). In the same year, Spengler et al. (1998) suggested a similar approach that they applied not just in LCA but for a multicriteria-based investment decision support system called KOSIMEUS. Even 2 years earlier, in the chemical industry, Kniel et al. (1996) suggested a detailed application of this combined method of process simulation, LCA, and economic analysis for the process optimization of a nitric acid plant.

With the speed of technological evolution in the past 20 years, the methods of digitally simulating and modelling processes have also become more advanced and sophisticated. There has been an increase in utilizing coding-language based tools like the Python-based Brightway, which is designed to easily implement and build advanced LCA models (Mutel 2017). This led to some interesting LCA approaches like linking building energy models (Heeren et al. 2015) and manufacturing process models (Bernstein et al. 2019) to LCA.

In the case of process plants, there are dedicated software available in the market that can create digital simulations of both existing, and future processes like HSC Chemistry®, SuperPro Designer v9, Aspen Plus, USIM-PAC®, and ProSim. If effectively used by process experts that have good understanding of the systems being modelled, it is expected to generate reliable simulated data and values for the input and output flows of the process, which are needed for the life cycle inventory (LCI) step of LCA. These simulated data, in the absence of laboratory or field data especially in the conceptualization phase, help address the ambiguity of “black boxed” processes, providing a more defined foreground data. This helps practitioners relate the resulting potential impacts to specific contributors instead of an averaged process.

Currently, this combination of digital simulation and LCA, or process simulation-based LCA, is not just used as a tool to optimize and assess existing process systems but also as a means to evaluate processes that are still in development. Through simulation, it is possible to visualize the possible results of process parameter changes that can be difficult to observe on laboratory tests. When combined with LCA, the results of investigating these parameters and incorporating it to the design of the process could help achieve a technically and environmentally efficient process design. This approach has been applied in both the chemical, and the metals and mining industry with the following references as examples (Kniel et al. 1996; Reuter and van Schaik 2015; Gnansounou et al. 2015; Cosate De Andrade et al. 2016; Pell et al. 2019; Elomaa et al. 2020b, a; Bartie et al. 2021; Rinne et al. 2021b, a; Aromaa et al. 2022; Ng et al. 2023). It should be noted that based on this published literature, confirming the observations from the simulation-based LCA on the actual scaled-up pilot or industrial plant process is not yet available to the public as of this writing.

In this study, the application of process simulation-based LCA is focused on the materials industry, specifically secondary processing or recycling. Even though it is not the primary approach for waste management, i.e., only after prevention and reuse (European Parliament and Council of the European Union 2008), recycling has become the immediate solution when industries are confronted about their waste and emissions. It cannot be denied that the goal of recycling has merits, that is, to return materials previously locked in waste stockpiles back into the economic loop. However, recycling could require high energy inputs and additional raw materials. It could also emit dangerous process gases and wastes that could be more difficult to treat and neutralize (Rada et al. 2018). It is therefore important to make sure that recycling processes are designed within the context of sustainability, or at least with environmental consideration.

In recycling, EOL management of lithium-ion batteries (LIBs) is one, if not the most, actively researched nowadays. Other than they contain critical raw materials (CRMs), the increasing interest on this topic is driven by the vital role of batteries for decarbonization, i.e., storage and efficient use of renewable energy (Zhang and Ramadass 2012), and as a power source for electric vehicles (EVs). With the prominence of EVs increasing in the last decade, and considering the lifetime of LIBs is between 10 and 15 years, it is estimated that by 2030 about 110,000 tonnes of LIBs will reach their EOL (EU Urban Mobility Observatory 2022). Since lithium (Li), cobalt (Co), manganese (Mn), nickel (Ni), and graphite that mainly compose LIBs are CRMs (Geoscience Australia 2022; Government of Canada 2022; U.S. Geological Survey 2023; Grohol and Veeh 2023), the need to keep these materials in the usable loop becomes more important.

Governments, researchers, and process owners around the world are increasing their efforts in developing and optimizing LIBs recycling processes to improve not just the recovery, but also the reusability of the recovered materials in battery production. In the European Union (EU), the battery directive (European Parliament and Council of the European Union 2006) was created and updated to ensure that batteries produced and/or used in EU have low carbon footprint during its life cycle (European Parliament and Council of the European Union 2023). They have also included provisions to improve collection, reuse, and recycling rates and CRM recovery efficiency from waste batteries.

Also being addressed in these efforts are the technological and process cost challenges of recycling LIBs. For example, pyrometallurgical recycling of LIBs leads to the loss of graphite, Li, and Mn, plus generation of hazardous gases. Also, pyrometallurgical processes normally have high energy requirements. On the other hand, hydrometallurgical recycling when used can result to high recovery and purity of substances. However, it involves a complicated process flow that requires the use of large amounts of chemicals and reagents. Also, due to the nature of the process, high volume of wastewater can be generated (Asadi Dalini et al. 2021).

The main objective in this study is to inspect the advantages and disadvantages of applying the simulation-based LCA approach as an eco-design tool for recycling processes that are still in the conceptualization phase. Building on the interest in LIBs recycling, a hydrometallurgical and a pyrometallurgical recycling process were designed from literature and assessed in separate case studies.

2 Methods

The LCA for the scenarios assessed in this study are conducted in 4 phases as described for LCA in ISO 14040 (2006), i.e., goal and scope definition, followed by the LCI, then life cycle impact assessment (LCIA), and lastly interpretation of the results. Note that each LCA phase is iterative as shown in Fig. 1. Also shown is how process simulation relates to the LCA methodology by providing process foreground data during LCI. The process parameter information in most cases is collected from laboratory or field data. However, in this case, since the processes are still being conceptualized, the parameter information is collected from literature, supplemented by previous experience of the researchers. Note that conducting another iteration based on the recommendations generated from the results of the assessment is optional (dotted lines).

2.1 Process simulation of the recycling processes

The process simulation part of this study is divided into three (3) main steps (Fig. 2). First, a literature review was conducted to gain information on the possible composition of the feed that is waste LIBs for the processes that will be conceptualized. Next, literature review and design of the processes were accomplished prior to the last step, which is the process simulation itself which was conducted using the Sim module of HSC Chemistry v10.5.

2.1.1 Waste Li-ion battery model

As noted in previous sections, there is an increasing interest in the EOL management of LIBs which includes recycling. Currently, there are five major types of cathode active material (CAM) chemistries commercially used for LIBs, i.e., lithiated cobalt oxide (LCO), lithium iron phosphate (LFP), lithium manganese oxide (LMO), nickel cobalt aluminum oxide (NCA), and nickel manganese cobalt oxide (NMC) (Korthauer 2019). Data from 2020 showed that NMC has the largest market share among the different types of cathode chemistries (Salgado et al. 2021; Flash Battery SrL 2022), and NMC 622 cells are the most commonly used especially in different types of EVs due to its performance and lower Co content, which is usually a concern due to its cost, and associated geopolitical risk (Saaid et al. 2024). From this, it can be assumed that it will make up a significant amount of the spent LIBs to be recycled in the next decade.

The battery cells are the basic units that make up the battery pack used in EVs as shown in Fig. 3 (Thompson et al. 2020). There are additional components added in between these levels like the module packaging, battery management system, casing, and cooling systems when they are installed in the vehicle. These components are usually recovered by manual dismantling to be reused or refurbished as well as reduce the impurities of the feed to subsequent recycling processes (Elwert et al. 2018; Koroma et al. 2022). Thus, in this study, spent NMC-based LIBs are considered to be the process feed, specifically spent NMC 622 LIBs cells.

For all types of LIBs, the main components of their cells are the anode, cathode, separator, electrolyte, and the cell housing (Ellingsen et al. 2014), and each one is composed of different types of materials. During recycling treatment of spent battery, a material called black mass (BM) is usually recovered. It is a fine substance (usually < 1 mm) which contains much of the anode and CAM from the battery (Wang et al. 2016; Zhang et al. 2018), and it is recovered during pre-treatment of the

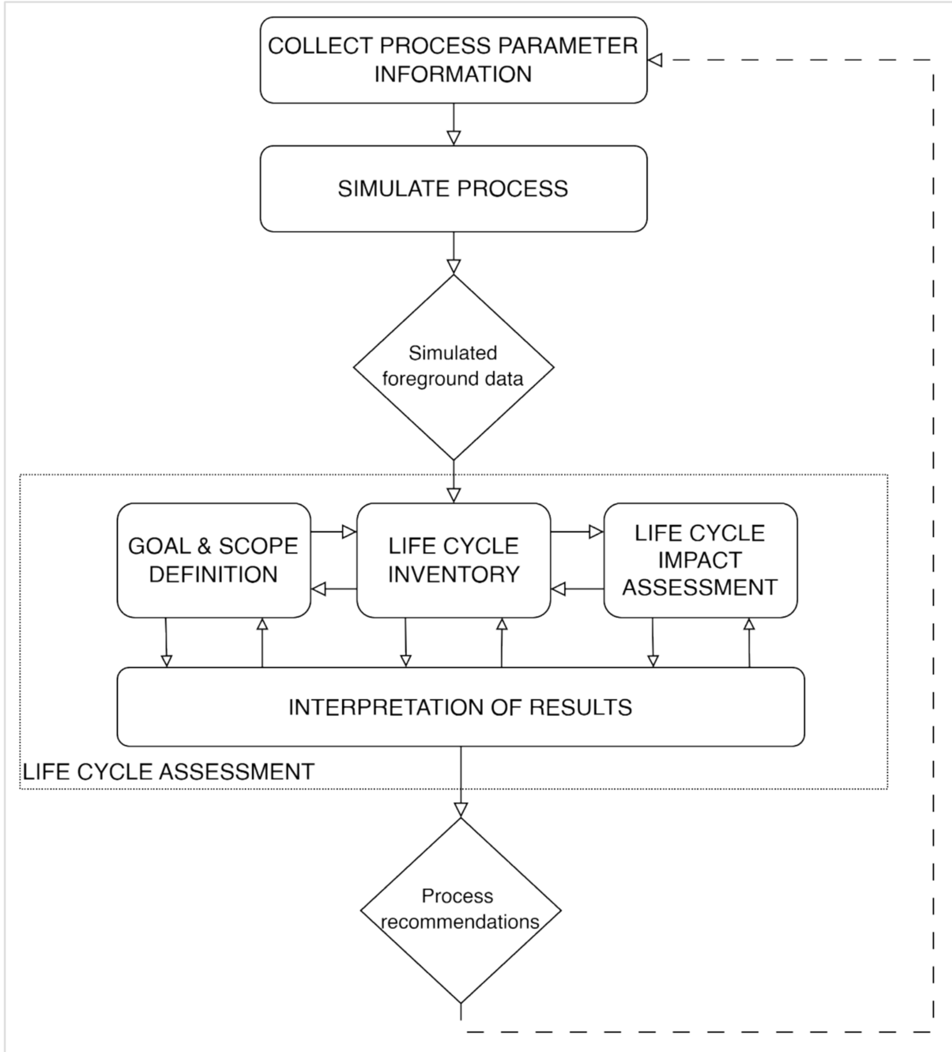


Fig. 1 Graphical representation of the process simulation-based life cycle assessment (LCA) methodology used in this study

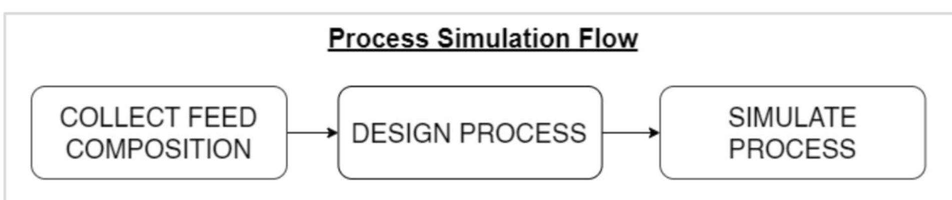


Fig. 2 Simplified flowchart of the specific steps to conduct the process simulation for each case study

spent LIBs. This is conducted in order to remove as many impurities as possible that could affect the subsequent processes, e.g., leaching.

To calculate and determine the composition of the battery cells and the black mass (BM) that will be used in the simulation, data from Wang et al. (2016) and Woeste et al. (2024) were used. The final compositions used in the simulation are summarized in Table 1, and more details are available in Online Resource No. 1—Tables 2 and 3.

2.1.2 Li-ion battery recycling

Nowadays, waste LIBs recycling, being highly studied, have several process variations implemented in different scales around the world. These processes can be generally grouped into physical, pyrometallurgical, hydrometallurgical, solvometallurgical, biometallurgical, and direct reuse approaches, and they can even be combined to maximize the recycling efficiency (Velázquez-Martínez et al. 2019;

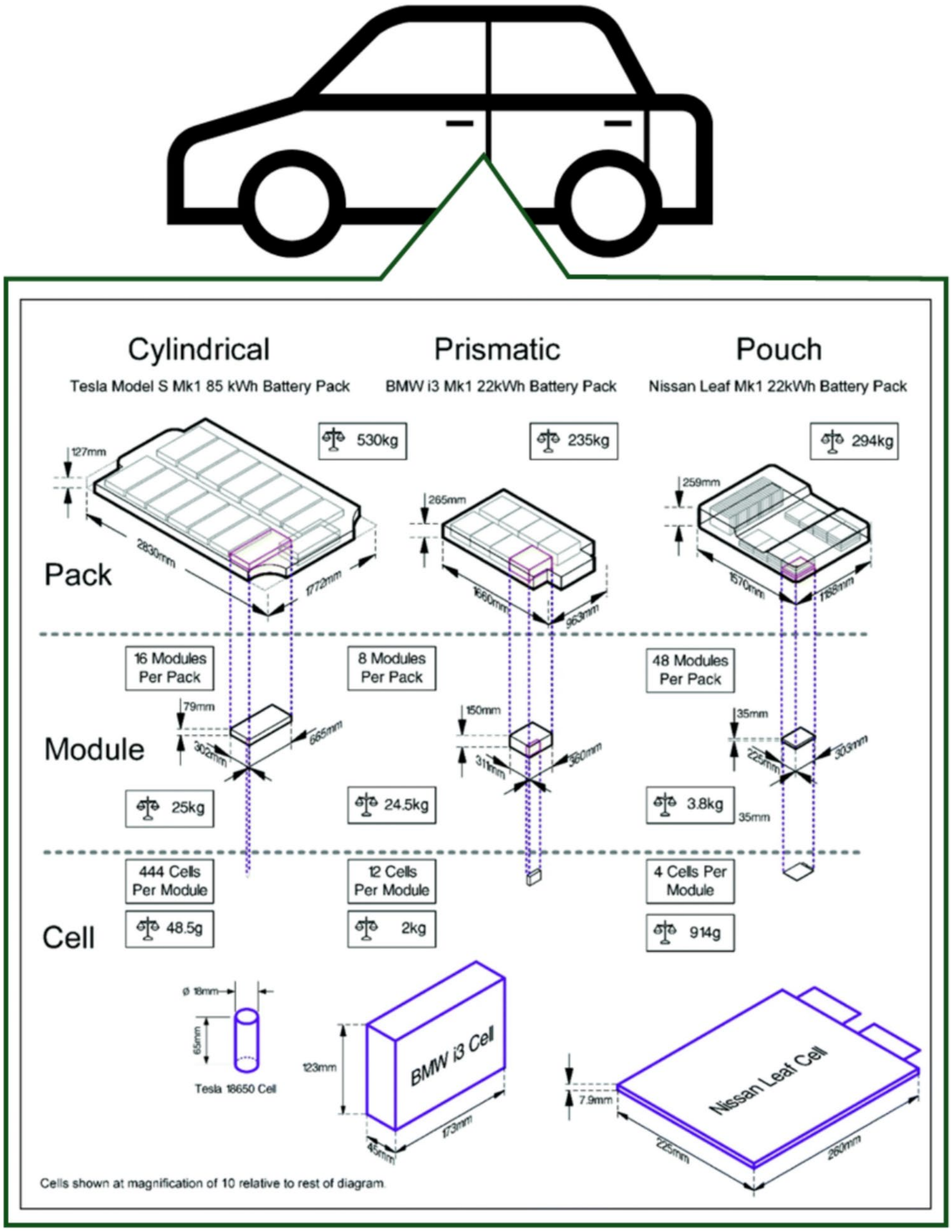


Fig. 3 An illustration of the different types of battery cells used in electric vehicle's battery packs. Image adapted from the figure of "different types of battery cells and how they are organised to form modules and packs" by Thompson et al. (2020) as permitted by its Creative Commons Attribution-Non Commercial 3.0 Unported Licence

Table 1 Composition of waste Li-ion battery (LIBs) cells and black mass (BM) used in the digital process simulation of the recycling processes for each case study

Content	NMC LIBs cells	Black mass
Aluminum (Al)	8.09%	1.82%
Aluminum oxide (Al_2O_3)	0.43%	0.10%
Copper (Cu)	16.31%	1.30%
Carbon (C)	22.46%	29.24%
NMC622 ($LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$)	19.86%	62.37%
Lithium hexafluorophosphate ($LiPF_6$)	14.66%	0.05%
Polyvinylidene fluoride (PVDF)	2.84%	5.12%
Ethylene carbonate (EC)	10.40%	-
Polyphenylene ether (PPE)	4.96%	-

Harper et al. 2019; Jung et al. 2021; Makuza et al. 2021; Toro et al. 2023; Wagner-Wenz et al. 2023; Xu et al. 2023; Zanoletti et al. 2024). Among these processes, hydrometallurgy and pyrometallurgy remain the most successfully implemented on a commercial scale, both having technology readiness levels (TRL) of 9. Though there are some emerging technologies like direct recycling that are also now commercially used, albeit on a smaller capacity compared to the other two, its TRL is only at an 8 (Jung and Zhang 2022; Wagner-Wenz et al. 2023). These processes (i.e., type 1 direct recycling) however are also employing additional processes that are based on hydrometallurgy (Velázquez-Martínez et al. 2019) which could be contradictory to the core definition of direct recycling which is recovering cathode and anode active materials from waste batteries using physical processes, without chemical alteration. This definition of direct recycling can be considered a second type and has a TRL of 4 which suggests that some prototype of the process is in operation but not yet scaled up (Wagner-Wenz et al. 2023; Gupta et al. 2024).

In this regard, the processes chosen to be conceptualized for this study in recycling waste LIBs are hydrometallurgical (Case Study No. 1) and pyrometallurgical (Case Study No. 2) approaches. Recall that the objective of this study is to examine the strengths and weaknesses of process simulation-based LCA when used as an eco-design tool for recycling processes that are still in the conceptualization phase, which is also addressing the data availability challenge. Thus, the processes with the highest level TRL became the main inspiration for a new process being planned to be developed or conceptualized in the context of this case study.

As previously mentioned, the subject of LIBs recycling is actively being researched around the world, and there are several variations of pyrometallurgy- and hydrometallurgy-based recycling processes available in the literature. The specific process flows to be simulated and assessed were decided based on a review of previously published works. For Case Study No. 1, it was based on the generalized process flow common to the current hydrometallurgy recycling processes being researched or done commercially that was reviewed by Jung et al. (2023). The same approach was applied to Case Study No. 2, that is, the common points of the current recycling processes for LIBs that mainly uses pyrometallurgy reviewed by Makuza et al. (2021) were consolidated for the simulation.

For both reviews, the main criteria in choosing the processes to be modelled were that they are either already generally applied in industry or have potential for scale-up. For the pyrometallurgical recycling process, since the usual concern is related to energy, the one chosen had to have the lower energy requirement compared to the other pyrometallurgical processes. For the hydrometallurgical process, another scenario based on Ecoinvent's data for hydrometallurgical recycling was included in order to compare the potential impacts of the data from the existing database to that of the simulation.

The hydrometallurgical process in the Ecoinvent database is described as a chemical treatment after initially shredding the waste battery cells (Hischier 2012). The data was cited to be based on a European recycling company (Fisher et al. 2006) but was modelled to fit as a global average data. This was only applied to the hydrometallurgy-based approach since there is no pure pyrometallurgy-based process available in the Ecoinvent database, i.e., the scope for Case Study No. 2 is only up to the production of the alloy.

Simulated hydrometallurgical recycling of waste LIBs For the hydrometallurgy-based recycling process of the waste LIBs, the critical and strategic raw materials like Co, Li, Mn, and Ni (Grohol and Veeh 2023) from the anode and cathode materials of the battery are required to be concentrated first from the waste LIBs cells as BM in order to minimize impurities during the subsequent processes. Thus, prior to leaching, the battery cells are subjected to pre-treatment, specifically mechanical pretreatment, as seen in Fig. 4. The pre-treatment involves a combination of size reduction, screening, and/or sorting, in order to concentrate as much of the BM and to minimize the effect of impurities (e.g., plastics and scrap metals) in the leaching process. Due to data limitation (i.e., size distribution analysis of the shredded and screened products), the pretreatment was not simulated using HSC Chemistry® but was calculated based on data from literature (Malmgren et al. 2013; Wang et al. 2016).

During the hydrometallurgical part of the process, the BM is leached using a lixiviant, specifically sulfuric acid (H_2SO_4)) to dissolve the target metals into the solution producing a pregnant liquor solution (PLS). Process parameters are then carefully controlled to keep these metals in solution until they are ready to be recovered. Since most of the times impurities could also be leached, the solution is purified by carefully controlling its pH to precipitate the impurities that are then removed through filtration. The purified PLS, assumed to only contain now the target metals, goes through a series of precipitation processes by pH control and specific precipitants, i.e., sodium hydroxide or caustic soda (NaOH) for the mixed Ni-Mn-Co hydroxide salts and sodium carbonate or soda ash (Na_2CO_3) for lithium carbonate (Li_2CO_3). Note from Fig. 4 that there is no wash water used in the filtration operation to recover the target products. This is to avoid redissolving the precipitated salts. More details about the simulation are found in Online Resource No.1. The products of the hydrometallurgical processes will need post-treatment to be usable for battery applications or for other purposes, but it is not included in the scope of this study.

Simulated pyrometallurgical recycling of waste LIBs In the case of the pyrometallurgical process, waste LIBs cells can be directly treated without mechanical pretreatment (Fig. 5). Though discharging the battery cells is an option, pyrometallurgy-based treatment makes it possible to eliminate the need to do so (Makuza et al. 2021). The method chosen to be simulated in this study was reductive smelting of the waste batteries due to its medium energy requirement in an industrial scale compared to using electric arc furnace and converter (Makuza et al. 2021). This only involves one step of smelting the waste LIBs by feeding it into a shaft furnace along with the reducing agents (commonly high in C materials like metallurgical coke), the slag formers (e.g., quicklime (CaO)), and air enriched with oxygen.

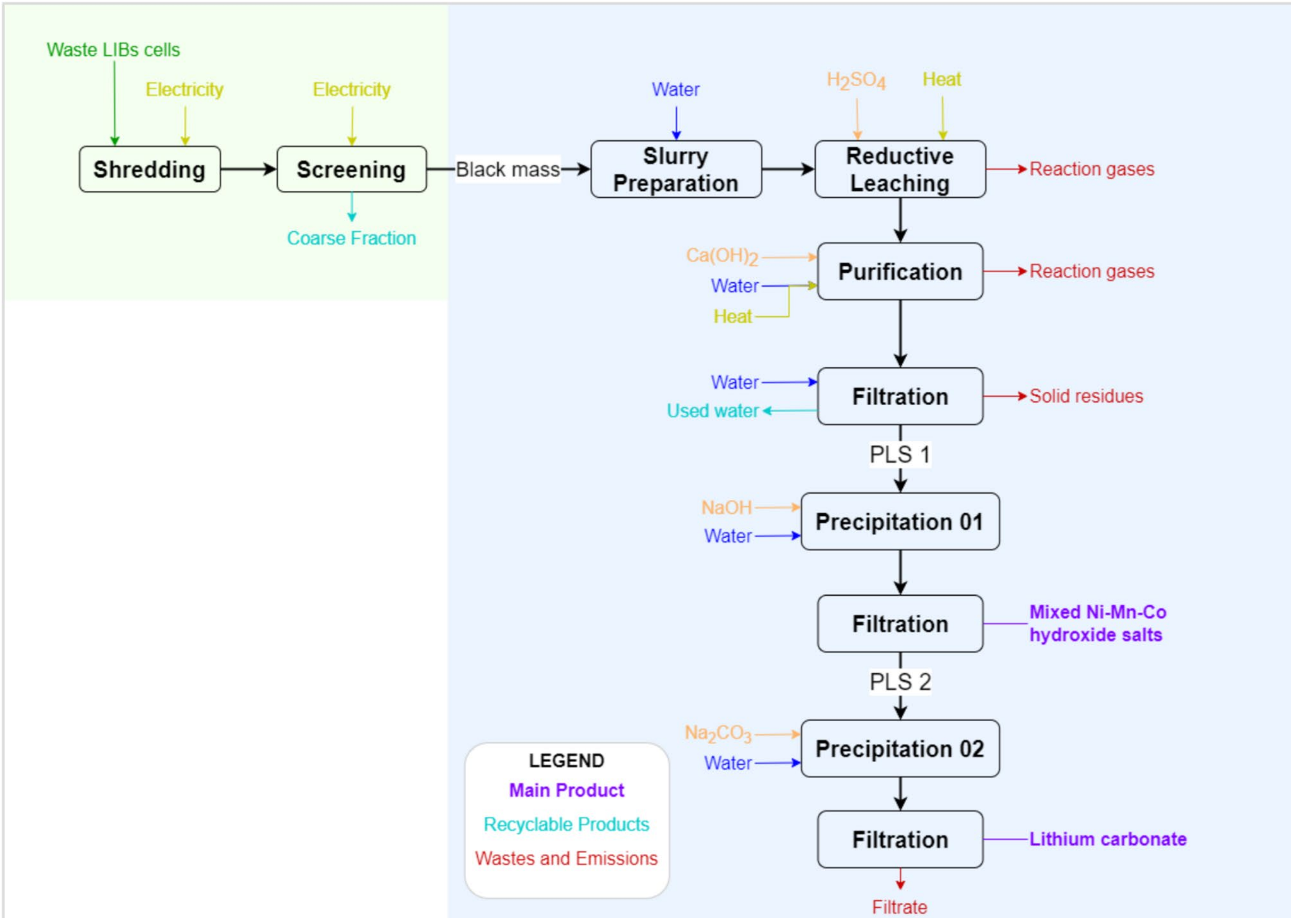
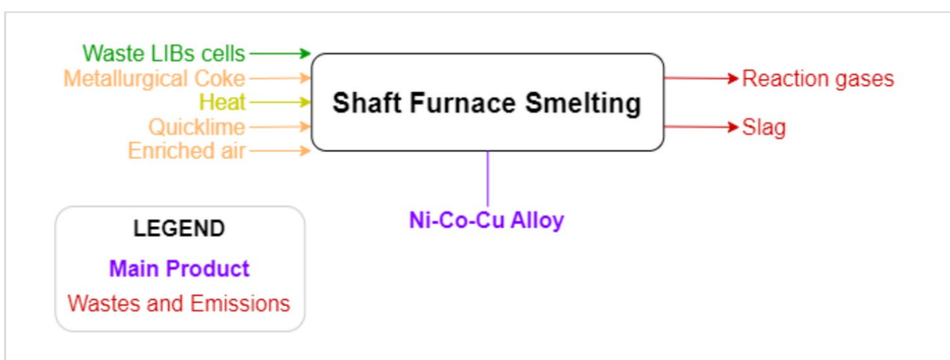


Fig. 4 Simplified process flow for the simulated hydrometallurgy-based recycling process of the waste Li-ion batteries (LIBs). The part in green was calculated using Excel and the one in blue was simulated using HSC Chemistry Sim module

Fig. 5 Simplified process flow for the simulated pyrometallurgical-based recycling process of the waste Li-ion batteries (LIBs)



In recycling waste LIBs through pyrometallurgical processes, the chemical composition of the battery cells allows it to be processed without mechanical pretreatment. The carbon from the anode's active material could serve as a reductant as well as other C-containing components like the plastic parts, supplementing the commonly used metallurgical coke. Also, though aluminium (Al) is lost in this approach, the reaction converting it to the slag component Al_2O_3 releases energy which could also help decrease the energy requirement. The process produces a Ni-Co-Mn-rich alloy, and a slag containing Li. Though it has been a common posit that pyro-treatment results to the loss of Li in the slag or to the flue dust, there has been claims that this is not necessarily the case (Verrecht 2023). This further treatment however is not on the scope of this study. More details about the simulation are found in Online Resource No.1.

2.1.3 Simulation of the recycling processes

As previously discussed, the recycling processes chosen for this case study were hydrometallurgy- and pyrometallurgy-based approaches of treating waste LIBs. Generally, LIBs recovered at EOL are in a pack (Harper et al. 2019). Since the objective of this study was focused on the battery cells, disassembling the pack including discharge of the battery was assumed completed prior to transporting the EOL LIBs cells to the recycling plant and thus out of the scope of this study.

For the simulated hydrometallurgy-based process, the cells required a pre-treatment stage in order to isolate the BM prior to the subsequent process (Fig. 4). On the other hand, the pyrometallurgical process directly uses the battery cells as feed (Fig. 5). For both case studies, the hydrometallurgical and pyrometallurgical treatment portion were simulated on HSC Sim module. Some important operational parameters that were used in the digital simulations are shown in Table 2, and more details about how the process simulation was modelled are available in Online Resource No.1. Note that the composition of the simulated process outputs is based on the process know how of the researchers supplemented with literature data. All logical phases that the products could contain are indicated in the software, but the final composition are determined by the software's thermodynamic-based calculations.

2.2 Goal and scope definition

The main goal of conducting this study was to examine the advantages and disadvantages of applying simulation-based LCA in eco-designing and assessing recycling processes that are still under development specifically for hydrometallurgical (Case Study No. 1) and pyrometallurgical (Case Study No. 2) methods. Most of the LCA studies of LIBs recycling processes found in literature were already at a commercial or industrial scale (Mohr et al. 2020; Rinne et al. 2021b, 2024; Du et al. 2022; Blömeke et al. 2022; Tas et al. 2024; Zhang et al. 2024; Lu and Wang 2024), and some are at least tested in the laboratory (Anwani et al. 2020; Wu et al. 2022; Duarte Castro et al. 2022; Kallitsis et al. 2022; Mousavinezhad et al. 2023; Cao et al. 2023; Liu et al. 2023, 2024; Premathilake et al. 2024). For those that were conceptualized to either industrial or laboratory scale operation, inventory data were based mainly on literature and scaled-up through methods that were not well-defined (Kallitsis et al. 2022; Blömeke et al. 2022; Mousavinezhad et al. 2023; Chen et al. 2023; Liu et al. 2023, 2024; Premathilake et al. 2024; Lu and Wang 2024). It is however known that there is a large variation between laboratory and scaled up processes especially at industrial scale, which in turn could influence the resulting potential impact of the process. Only a few studies were found that employed process simulation to scale literature data like this study (Rinne et al. 2021b, 2024; Kim et al. 2024; Tas et al. 2024). The difference in the case here is that the processes is purely a concept with no laboratory experiments conducted dedicated for the exact unit process combinations.

In LCA, the choice of functional unit (FU) is crucial on the subsequent interpretation of the results. Since the goal

Table 2 Summary of the operation parameters used in the digital process simulation of each study case

Parameters	Values	Reference
Hydrometallurgy-based recycling process simulation		
Black mass slurry density	1,600 kg/m^3	Assumption
Leaching, temperature	90 °C	Guimarães et al. (2022)
Leaching, pH	3.2 ± 0.1	Guimarães et al. (2022)
Precipitant/pH control, %solids	45%	Assumption
Purification, temperature	60 °C	Han et al. (2020)
Purification, pH	6.5 ± 0.5	X. Zhang et al. (2018)
Mixed hydroxides precipitation, temperature	60 °C	Han et al. (2020)
Mixed hydroxides precipitation, pH	11.5 ± 0.5	X. Zhang et al. (2018); Zou et al. (2013)
Lithium carbonate precipitation, temperature	50 °C	Battaglia et al. (2022)
Lithium carbonate precipitation, pH	12.8 ± 0.3	Han et al. (2020); Meshram et al. (2015)
Pyrometallurgy-based recycling process simulation		
Coke-to-feed ratio	1:1	Assumption
Operation temperature	1300 °C	Makuza et al. (2021)

Table 3 A summary of the defined goal and scope for the waste Li-ion battery (LIBs) recycling LCA life cycle assessment (LCA) case studies

System/product assessed	*Case Study No. 1* Scenario A: Literature- and process simulation-based hydrometallurgical recycling process that aims to produce mixed hydroxides salts of Ni-Mn-Co and lithium carbonate (Li_2CO_3) Scenario B: Ecoinvent's hydrometallurgy-based recycling process (product not defined) *Case Study No. 2* Literature- and process simulation-based pyrometallurgy-based recycling process that aims to produce a Co–Ni-Cu alloy and a slag that contains Li
Functional unit	Treatment of 1-kg waste Li-ion battery
Scope of the study	• *Geographical boundary* Belgium (BE) and EU • *System boundary* Cradle-to-gate which starts from the transportation of the waste LIBs cells and other raw material inputs to the recycling facility until the recovery of the target CRMs, i.e., mixed hydroxides Ni-Mn-Co salts, and Li_2CO_3 for Case Study No. 1, and Co–Cu-Ni alloy for Case Study No. 2
Multifunctional strategy	Main strategy: Cut-off approach Additional analysis: EOL recycling or avoided burden approach, to assess the potential benefits of the recycling processes
Impact assessment methodology	Environmental Footprint (EF) 3.1 v1.0 (Simapro v9.5)
Assumptions and limitations	• For the inventory of the hydrometallurgy-based recycling method, the discharge water from different unit processes (i.e., wash water used in filtration, including the final filtrate after the recovery of Li_2CO_3) are assumed to be collected in a separate tank and reused as process water instead of being discharged • Reaction gases are assumed to be directly emitted. In practice, these gases are treated prior to discharge to meet regulation standards • No infrastructure is included in the inventory, e.g., chemical plant and specific equipment

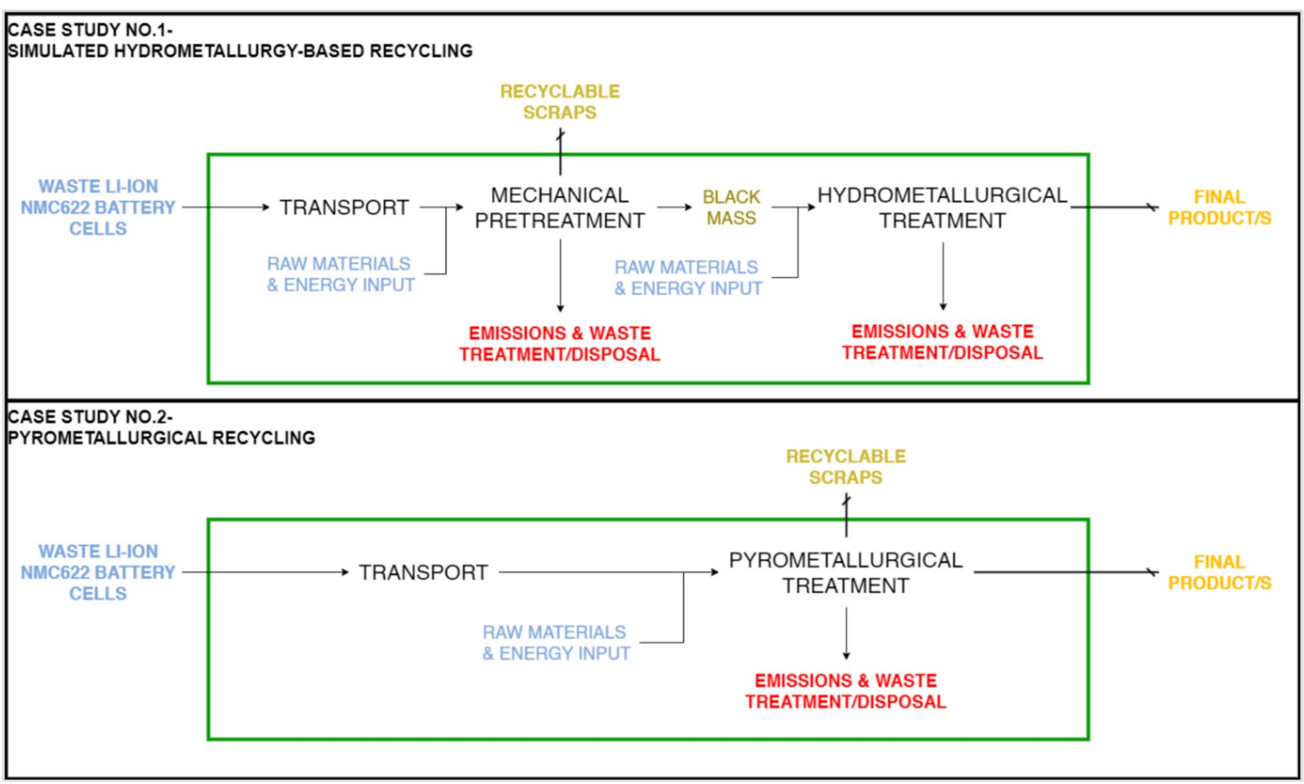


Fig. 6 Simplified graphical representation of the system boundaries (inside green box) for the simulated scenarios. The broken arrowhead represents the "avoided burden" from the production of the recyclable materials and the final products

here is mainly to eco-design recycling processes for treating waste LIBs by assessing the potential impacts, the FU used was the treatment of 1-kg waste Li-ion battery cells using either the hydrometallurgical approach or the pyrometallurgical one as shown in Table 3.

Regarding the scope of the study, cradle-to-gate was the system boundary for the assessment (i.e., from material acquisition to the recovery of the target products, including handling of process wastes and emissions) (Fig. 6), and the geographical boundary was Belgium (BE) and the EU. This means that for raw materials, transport, heat, and electricity that were needed in the process, it was assumed that they are mainly sourced from BE, if not, other European countries.

Since the processes assessed will generate multiple products from each recycling process, setting the strategy of possible multifunctionality was necessary before even starting the assessment. For an attributional LCA, it is important to first classify the outputs of a process into the main product, co-products, recyclable material, and waste (Williams and Eikenaar 2022). Though the treatment of the waste generated will be a burden of the process assessed, the subsequent use life of the recyclable materials and the final products is not necessarily the case.

To model how the products and the recyclable materials contribute to the total potential impacts of the process, cut-off approach or the avoided burden approach is commonly used (Nordelöf et al. 2019). The cut-off approach gives no credit or burden to the recycling process for the subsequent use life of products. On the other hand, the avoided burden approach gives "credit" to the recycling process for the potential of its product to replace primary raw materials, e.g., chemicals, metals, and minerals. There are of course several other variations available (Ekvall et al. 2020), but these two are the main basis for most of them. Initial analysis will be conducted using cut-off to focus on the potential impacts of the processes being assessed. Since this study also aims to assess the potential benefits of these recycling processes, the avoided burden approach will be conducted as a sensitivity analysis.

2.3 Life cycle inventory

This phase of the study involved two main steps. First was the simulation of the processes for each scenario (discussed in Sect. 2.1), and next was the inventory based on the simulated results supplemented with data from literature. Once data was generated from the simulations and calculations, the information was consolidated and added to the LCA software, i.e., Simapro v9.6 with Ecoinvent 3 database, for the inventory.

The background data regarding transportation, production of input raw materials, water, heating, electricity, waste treatment, and emissions were adopted from the built-in Ecoinvent database in the LCA software. Though the database contains extensive data for various products, services and processes, careful examination of each dataset and whether it fits to address the LCA questions posed is still necessary. For example in this case, the Belgian electricity mix needed to be updated based on the latest report about Belgium's energy mix and usage (IEA 2022; Elia Group 2023) since the last update of Ecoinvent's data was from 2020 instead of 2022 (Treyer 2023).

3 Results

The results from the process simulation, inventory and impact assessment phases of the LCA are presented in this section.

3.1 Simulation results and inventory

Data for the inventory of both hydrometallurgical and pyrometallurgical recycling is collected mainly from the results of the digital simulations of the two recycling processes assessed. The products of each process (except for air and water emissions from reaction or flue gases, and used process water, respectively) and their classifications, as required for attributional LCAs, are shown in Table 4. The detailed inventory for both processes is available in Online Resource No.2.

3.1.1 Case study no. 1 process simulation

Other than the general mass flow of inputs and outputs, the chemical composition is also recovered from the simulation making it possible to compute the recoveries. The target CRMs, i.e., Li, Ni, Mn, and Co, recovered based on the simulation of the hydrometallurgical recycling process are about 71%, 81%, 81%, and 81%, respectively (Table 5). It is also noted from the simulation that there are some impurities present in the precipitated solids. The mixed NiCoMn hydroxide is only about 62% pure with the

Table 4 Classification of the waste Li-ion battery (LIBs) cells recycling process outputs for each case study

Product classification (*Attributional LCA*)	Case Study No. 1
Main product/co-products	Lithium carbonate (Li_2CO_3)
	Mixed Ni-Mn-Co hydroxide
Recyclable	Coarse fraction
Waste	Plastic scraps
	Waste electrolyte
	Impurities from purification (gypsum, graphite, etc.)

Table 5 Data summary of the recovery, and purity of the products from the simulated hydrometallurgical treatment of the black mass (BM) recovered from the waste Li-ion battery cells

Products	Elemental recovery				Purity
	%Ni	%Co	%Mn	%Li	
Mixed NiCoMn hydroxide	81.11%	81.11%	81.11%	-	61.79%
Lithium carbonate (Li_2CO_3)	-	-	-	70.82%	44.94%

moisture and impurities removed, while the Li_2CO_3 has only about 45%. Nevertheless, it is assumed for the inventory that the wet weight of the precipitated products, even with the impurities, are the final products which needs further post-treatment and refining before they can be used for any applications.

It is interesting to note that the main impurities of the products come from the coprecipitation of sodium sulfate (Na_2SO_4) with the target metals. The Na^+ ion was from the precipitant and pH regulator for both precipitation steps, i.e., caustic soda and soda ash, respectively. Since the products were observed to have low purity because of this, especially Li_2CO_3, further investigation of the system was conducted using the digital simulation.

Recall that the amount of reagents was automatically calculated by the software. For this digital experiment, the automation was turned off and the amount of soda ash used in precipitating Li was manually added instead. When the amount was decreased (i.e., base scenario used about ~287 kg), the purity increased while Li recovery only increased up to a certain point as shown in Fig. 7. On the other hand, when it was increased compared to the base scenario, the recovery also increased but only to a certain point then decreased again with a higher amount of soda ash. On the other hand, the purity decreased. This could mean that the simulated soda ash consumption in the base scenario is not necessarily the optimum dosage to maximize the recovery and purity of the recovered Li_2CO_3. Since the software's simulation is mainly thermodynamics-based, some key factors that could affect the reaction kinetics were not being considered (e.g., particle size, agitation). This means that conducting confirmatory experiments in the laboratory would be necessary to optimize the reagent dosage and maximize process efficiency.

With the maximum Li recovery at only about 71%, the recovery is within the target for the new EU battery

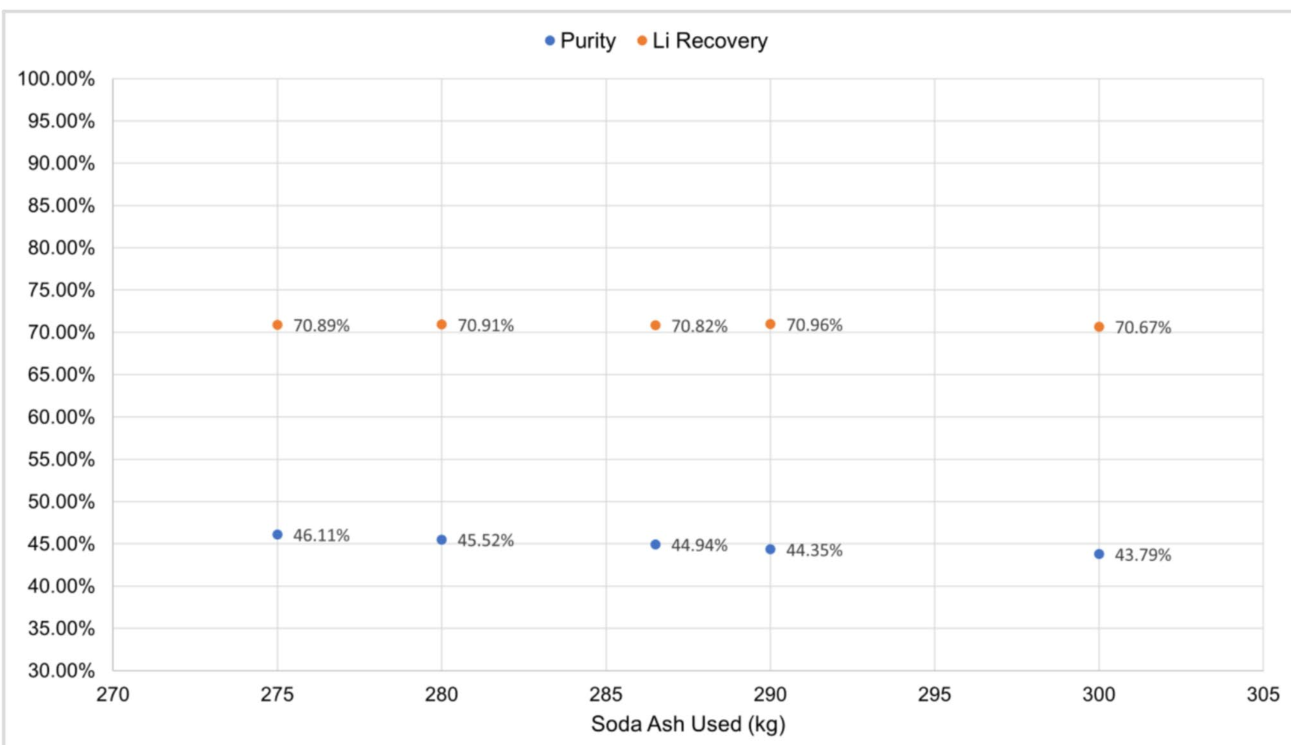


Fig. 7 A scatter chart showing the purity of the recovered lithium carbonate (in %), and the corresponding Li recovery (in %) vs the amount of soda ash used based on the digital process simulation of the hydrometallurgical treatment of black mass recovered from the waste Li-ion battery cells. Graph value details are available in Online Resource No. 2

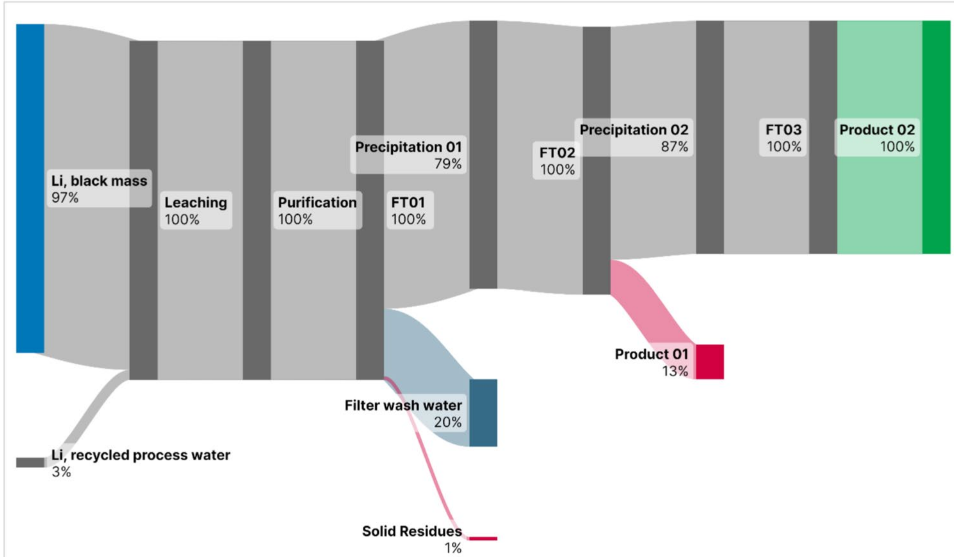


Fig. 8 A Sankey diagram representing the flow of Li from the black mass (BM) up to the different outputs of the simulated hydrometallurgical process. Graph value details are available in Online Resource No. 2. (FT = filter, Product 01 = mixed NiCoMn hydroxide, Product 02 = lithium carbonate)

regulation, that is, a 50% recovery for Li by end of 2027 (European Parliament and Council of the European Union 2023). It is however important to note that the final recovery after post-treatment and refining could be lower due to process losses, thus maximizing the Li recovery at this point would be necessary. Upon tracing the flow of Li in the simulated process, it was observed that the loss is due to diluted Li in the filtrate and solid residues recovered during filtration as shown in Fig. 8. Though the used wash water in FT01 is recycled back into process as process water during BM slurry preparation, not all the water is reused since there is more process water than what is required to maintain the target %solids of the BM slurry. In total, about 20% of Li is discharged with the used FT01 wash water, and about 11% is lost with the solids recovered, i.e., waste solid residues after purification, and the mixed NiCoMn hydroxide salts after the first stage of precipitation.

3.1.2 Case study no. 2 process simulation

For the second case study, i.e., pyrometallurgical recycling process, Li is mostly lost in the slag (Table 6). Though it is a critical material, it is considered waste in this case because combined with its low concentration in the slag (about 40%) and complicated post-treatment, recovery is deemed impractical.

On the other hand, though the recovery for the target metals Ni, Co, and Cu are all 100%, the alloy is only 83% pure in terms of these target metals. This emphasizes the need for refining before the target metals can be reused into any applications. This is due to the presence of other metals (i.e., Mn, Li, Fe) that formed with the alloy.

Recall that all logical phases in the process outputs were added into the simulation, and then the software's thermodynamical calculation determined the final composition. In this case, it indicated that it is thermodynamically possible for Mn, Li, and Fe to be present in the alloy. For the slag, though it was indicated that the oxides of Ni, Co, and Cu are possible phases in it, the thermodynamic calculations favored the formation of the metals in the alloy. Also, recall that thermodynamics is the main basis of the simulation software's calculation, i.e., no kinetics included, so for the process to achieve such recoveries could require long reaction times.

3.2 Impacts assessment

3.2.1 Case study no. 1—hydrometallurgy-based recycling process

The assessment of the potential impacts of the hydrometallurgy-based treatment of waste LIBs based on the digital simulation data (A) and Ecoinvent (B) is shown in Fig. 9.

Table 6 Data summary of the recovery, and purity of the products from the simulated pyrometallurgical recycling of waste Li-ion battery (LIBs) cells

Process outputs	Elemental recovery					Purity (Cu-Co–Ni)
	%Ni	%Co	%Mn	%Cu	%Li	
Co–Cu-Ni alloy	100%	100%	100%	100%	2.31%	83.07%
Slag	-	-	-	-	66.61%	-
Offgas	-	-	-	-	31.08%	-

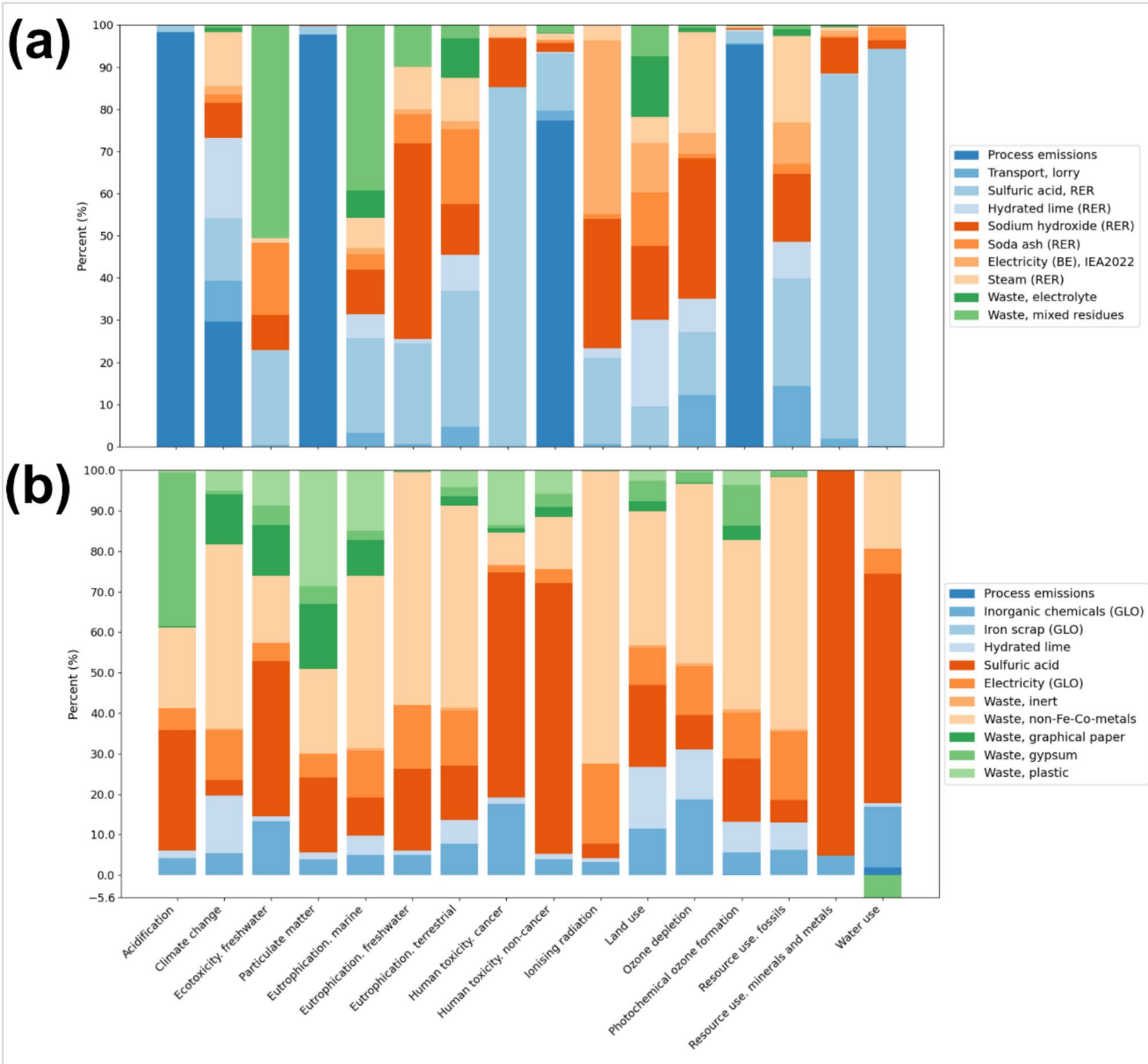


Fig. 9 Bar charts representing the life cycle impact assessment (LCIA) results for the (**a**) simulated hydrometallurgy-based recycling of waste Li-ion battery cells and (**b**) the Ecoinvent data-based model for the same type of technology, assessed using Environmental Footprint 3.0 Method v1.0 and calculated in Simapro v9.6. (RER = rest of EU, BE = Belgium, GLO = global)

For the simulated process (Fig. 9a), most of the impact categories are influenced by either H_2SO_4 or process emissions except freshwater ecotoxicity (ETP) and marine eutrophication (EP), i.e., waste mixed residues treatment contributed most. Other exceptions are freshwater EP, and ozone depletion (ODP) which is mainly influenced by the NaOH used to precipitate the mixed NiCoMn hydroxide, ionising radiation (IRP) which is influenced most by electricity used, and lastly hydrated lime contributes most to land use (LU) potential impacts. Sulfuric acid, the lixiviant used during leaching, contributed mostly to terrestrial EP, cancerous human toxicity (HTP), fossils, and minerals and metals resource use (RU), and water use (WU). On the other hand, process emissions have the highest potential contribution to acidification (ADP), climate change (GWP), non-cancerous HTP, and photochemical ozone formation (POF).

For the assessment of the Ecoinvent data for the hydrometallurgical treatment of waste LIBs cells, treatment of the non-Fe-Co metals scrap (i.e., represents Al and Cu scrap) and sulfuric acid are the highest contributors for most of the impact categories except ADP, and PM impacts. For ADP, the treatment of the waste gypsum contributed most, while

Table 7 A comparison of the total potential impacts of the Ecoinvent dataset and the simulated hydrometallurgy-based recycling process of waste Li-ion batteries (LIBs)

Damage category	Unit	Simulation	Ecoinvent	% difference
Acidification	mol H + eq	1.25E + 00	8.74E − 03	***−14,243.29%***
Climate change	kg CO2 eq	9.42E − 01	8.00E − 01	***−17.70%***
Ecotoxicity, freshwater	CTUe	1.09E + 01	4.32E + 00	***−151.79%***
Particulate matter	disease inc	6.39E − 06	7.38E − 08	***−8553.34%***
Eutrophication, marine	kg N eq	6.79E − 04	8.16E − 04	16.78%
Eutrophication, freshwater	kg P eq	1.20E − 04	2.71E − 04	***55.98%***
Eutrophication, terrestrial	mol N eq	5.01E − 03	6.95E − 03	27.99%
Human toxicity, cancer	CTUh	6.60E − 09	1.09E − 09	***−503.75%***
Human toxicity, non-cancer	CTUh	1.88E − 08	1.35E − 08	− 39.64%
Ionising radiation	kBq U-235 eq	7.70E − 02	6.94E − 02	− 10.92%
Land use	Pt	1.13E + 00	1.35E + 00	16.11%
Ozone depletion	kg CFC11 eq	1.51E − 08	4.72E − 09	***−220.42%***
Photochemical ozone formation	kg NMVOC eq	6.63E − 02	2.46E − 03	***−2602.32%***
Resource use, fossils	MJ	8.35E + 00	7.53E + 00	***−10.88%***
Resource use, minerals and metals	kg Sb eq	1.56E − 07	1.33E − 05	98.82%
Water use	m^3 depriv	1.47E + 00	2.28E − 01	***−543.14%***

Italicized and bolded values signifies the significant difference between the simulate recycling process and the Ecoinvent dataset

the treatment of the waste plastic contributed most to PM impacts. It should be noted that for this dataset, it has been modelled in the global context and the treatment of the generated wastes are a combination of different treatment background data for different geographical locations.

In terms of the total values for each potential impacts, the simulated recycling process is greater than the Ecoinvent dataset for most of the impact categories except marine, freshwater, and terrestrial EP, LU and minerals and metals RU as shown in Table 7. Typically for ADP, EP, and POF (or respiratory inorganic effects) impacts, a difference of 30% between scenarios is considered significant (Jolliet et al. 2016), which in this case applies to all except marine and terrestrial EP. For climate change and fossil resource use, significant difference between the scenarios is observed (i.e., > 10%), as well as with the toxicity-related potential impacts except for non-cancerous HTP (Jolliet et al. 2016). Those categories with significant difference including those with a %difference greater than 100% are emphasized in bold and italicized.

In order to assess whether there is any benefit in recycling LIBs using this process prior to improving it, an additional analysis using end-of-life or avoided burden approach was conducted. Recall that the products of the simulated hydrometallurgical treatment process are Li_2CO_3 and mixed NiCoMn hydroxides. Since they still contain impurities (i.e., mixed NMC hydroxide is about 71% pure, and Li_2CO_3 is about 45% pure), the pure amounts of Li_2CO_3 and mixed NiCoMn hydroxide in the products were calculated for the avoided burden analysis. This is because these values represent the potential amounts that can be recovered from the products after post-treatment, which in turn represents the potential avoided primary materials, i.e., Li_2CO_3 produced from spodumene and brines, and NMC hydroxides produced from reacting primary nickel sulfate ($NiSO_4 \bullet 6H_2O$), cobalt sulfate ($CoSO_4 \bullet 7H_2O$), and manganese sulfate ($MnSO_4 \bullet H_2O$) with sodium hydroxide. The results showed that most of the negative potential impacts or "benefits" is mainly due to the avoided use of NMC hydroxide (Fig. 10).

It should be noted that the primary NMC hydroxide used in the model is the NMC811 hydroxide dataset. NMC811 has higher ratio of Ni, and lower Mn, and Co compared to the NMC622 waste LIBs that was assessed in this study. Due to the close values of the amount between these two NMC types, it was deemed insignificant for this case to use whether NMC811 or NMC622 data, that is, if it exists. This was also confirmed by an additional analysis on NMC811 hydroxide dataset by Ecoinvent (see Online Resource No.2).

3.2.2 Case study no. 2—pyrometallurgy-based recycling process

For the simulated pyrometallurgical-based treatment of waste LIBs, the LCIA results are shown in Fig. 11. Coke has the highest contribution to most of the impact categories specially in the most significant ones based on normalization. The exceptions are freshwater E), non-cancer HTP, and WU categories. For these impacts, it is mostly influenced by the liquid oxygen that was used to produce the enriched air used during smelting, except for non-cancer HTP which is influenced by the process emissions made up of the untreated offgas released during smelting.

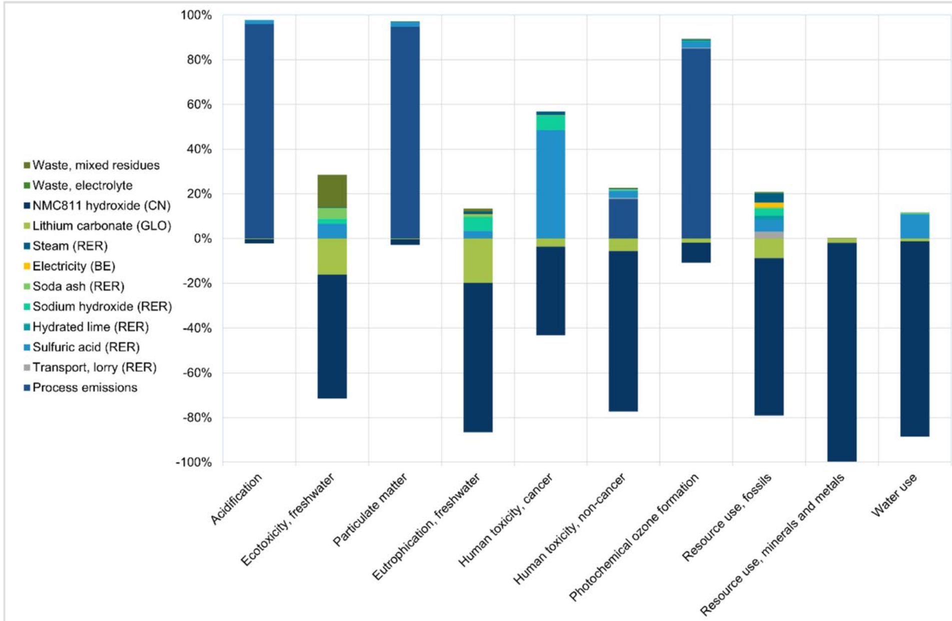


Fig. 10 A bar chart representing the life cycle impact assessment (LCIA) results using avoided burden approach for the simulated pyrometallurgical recycling of waste Li-ion battery (LIBs) cells. The Environmental Footprint (EF) 3.0 Method v1.0 was used in the Simapro v9.6. Impact categories shown are the most significant based on normalization. (GLO = global, CH = Switzerland, RER = rest of EU, CN = China)

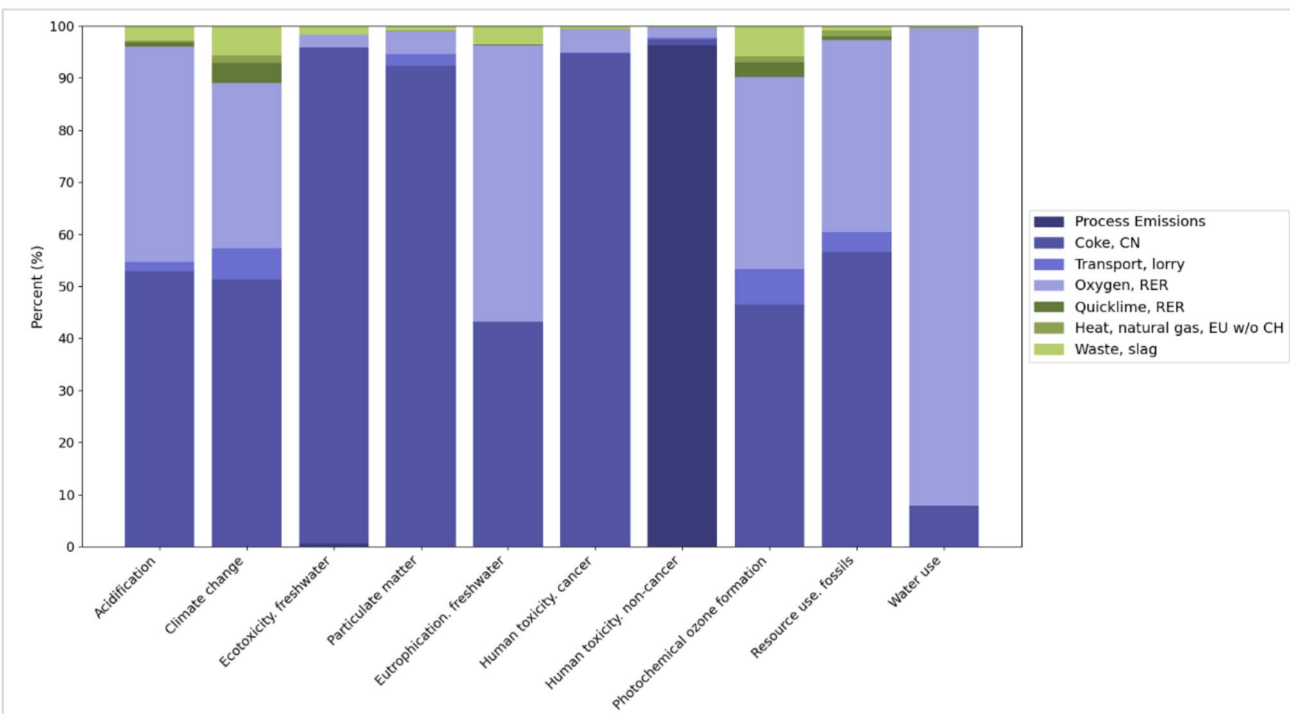


Fig. 11 A bar chart representing the life cycle impact assessment (LCIA) results for the simulated pyrometallurgical recycling of waste Li-ion battery (LIBs) cells assessed using Environmental Footprint (EF) 3.0 Method v1.0 and calculated in Simapro v9.6. Impact categories shown are the most significant based on normalization. (CN = China, RER = rest of EU, CH = Switzerland)

As in the first case study, an additional analysis using the avoided burden approach was also conducted for the pyrometallurgy-based recycling process. Considering the Ni, Co, and Cu content of the alloy product, most of the avoided burden is attributed to the primary production of Cu (Fig. 12). It should be noted that the post-treatment and refining processes needed to achieve the same quality as the primary material was not taken into account in terms of the recycling process' burden and thus could greatly change the results of this analysis. Expanding the system boundary and

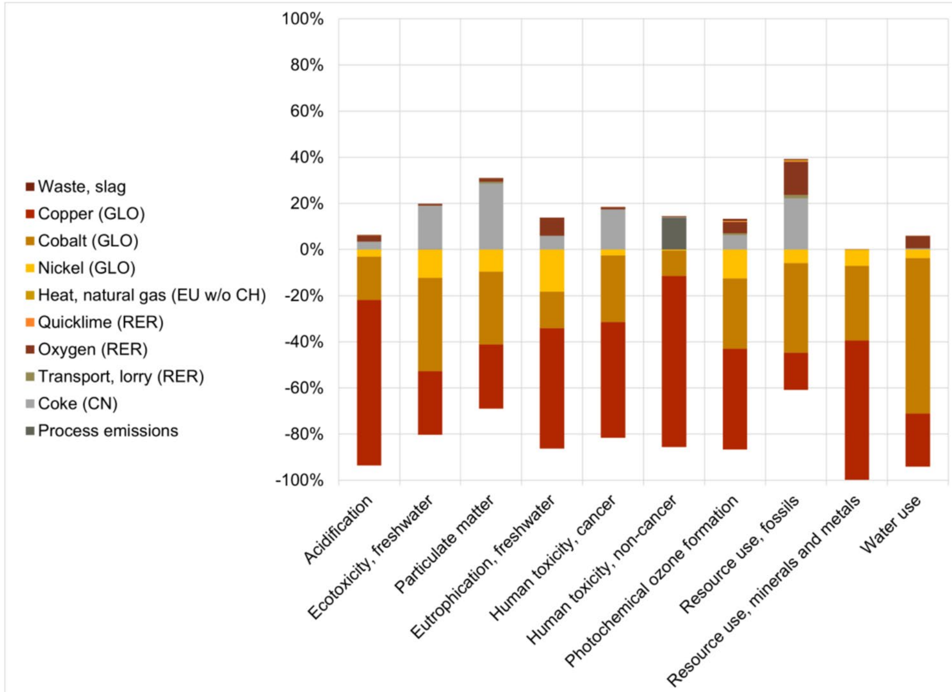


Fig. 12 A bar chart representing the life cycle impact assessment (LCIA) results using avoided burden approach for the simulated pyrometallurgical recycling of waste Li-ion battery (LIBs) cells. The Environmental Footprint (EF) 3.0 Method v1.0 was used in the Simapro v9.6. Impact categories shown are the most significant based on normalization. (GLO = global, CH = Switzerland, RER = rest of EU, CN = China)

reanalysing this model could be an option for the next iteration of this process (see Fig. 1).

3.3 Sensitivity analysis

To confirm how the modelling decisions and assumptions affected the final results of the digital process simulation-based LCA, a sensitivity analysis for both of the simulated recycling processes is conducted. One of the first and biggest assumption was the composition of the battery cells used (i.e., feed for the pyrometallurgical treatment), and the black mass (i.e., feed for the hydrometallurgical treatment) recovered from it which is what was assessed in this analysis. Other modelling choices deemed significant for each case study were also assessed and presented below.

3.3.1 Case study no. 1—hydrometallurgy-based recycling process

For the hydrometallurgical process, the black mass composition used in the simulation was assumed to not contain any sources of Fe (see Online Source No.1). However, the majority of the information found in literature regarding the composition of BM for different types of Li-ion batteries had Fe to some extent (Ellingsen et al. 2014; Pavón et al. 2021; Mousa et al. 2022; Woeste et al. 2024), and it has been noted that higher Fe content, at least more than 1%, could have a negative effect in the subsequent processes in treating BM (Pavón et al. 2021).

Based on the results of varying the Fe content of the BM from the base scenario, i.e., 0 kg then to 15 kg, 30 kg, 45 kg, and 60 kg, most of the impact categories decreased with the presence of Fe in the BM except for freshwater ET impacts (Fig. 13). Nonetheless, no significant difference between each scenario is observed except for the fossils RU impacts of the scenario that has 60-kg Fe in the feed compared to the other four scenarios (i.e., > 10% difference).

Along with the total potential impacts decreasing with the presence of Fe, the purity and recovery of the products were also decreasing (Fig. 14). This could be due to the decrease of reagents used (see Online Resource No.2), which in turn decreased the recovery and the associated potential impacts.

3.3.2 Case study no. 2—pyrometallurgy-based recycling process

The effect of choosing modelling NMC622 cells in this study is assessed through a sensitivity analysis. For the composition of the LIBs cells, the CAM was varied from the base scenario's NMC622 ($LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$) to NMC333 ($LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$), NMC532 ($LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$), NMC721 ($LiNi_{0.7}Mn_{0.2}Co_{0.1}O_2$), and NMC811 ($LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$), which are the NMC cathodes used in EVs (Saaid et al. 2024). The sensitivity analysis showed that using NMC333 had a higher potential impact in almost all the impact categories except freshwater ecotoxicity (ETP) and non-cancerous HTP which showed NMC811 cells having the higher total impact. However, the difference is not significant as shown in Fig. 15.

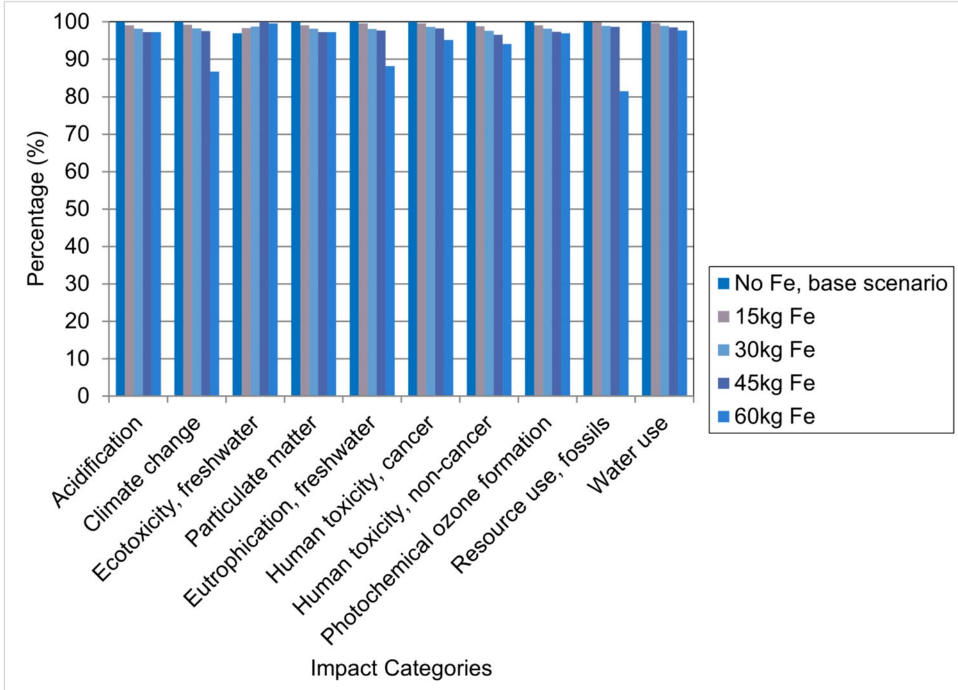


Fig. 13 A comparative bar chart for the simulated hydrometallurgy-based recycling process varying the amount of Fe in the black mass (BM) feed, represented with the most significant impact categories based on the normalization results. Environmental Footprint (EF) 3.0 Method v1.0 of Simapro v9.5 was used for the assessment

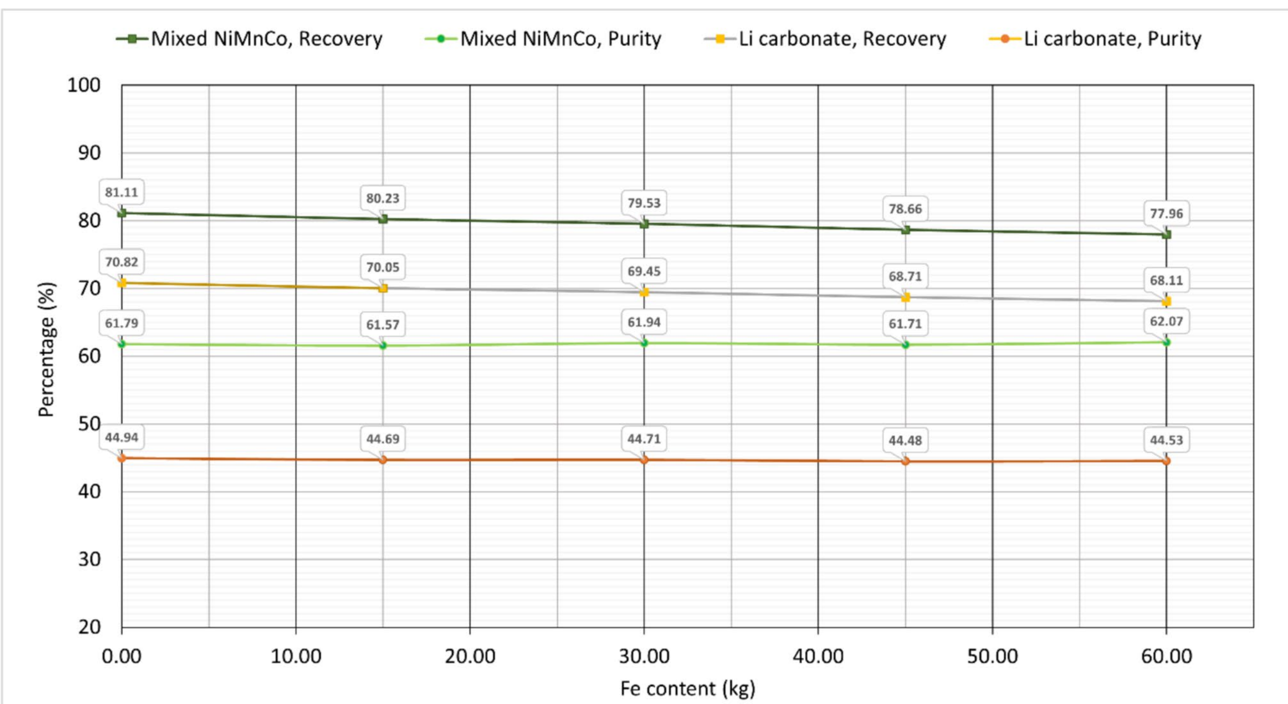


Fig. 14 A line chart showing the resulting recovery and purity, in %, of the recovered mixed NiCoMn hydroxides and lithium carbonate vs the amount of Fe present on the black mass (BM) feed based on the digital process simulation of the hydrometallurgical treatment of BM recovered from the waste Li-ion battery (LIBs) cells

Another literature source for the composition of an NMC battery cell to compare the one used in the model was also simulated in order to further assess the sensitivity of the potential impact to the feed. The NMC cell composition noted by Mohr et al. (2020) was used to calculate the possible phases in the feed for the simulated

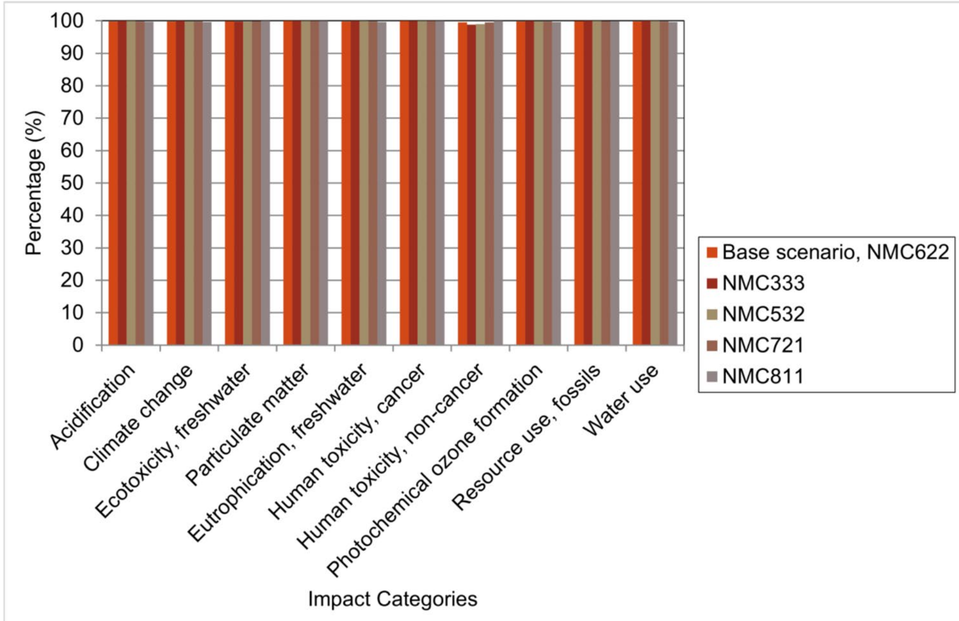


Fig. 15 A comparative bar chart for the simulated pyrometallurgical recycling process varying the cathode active material (CAM) of the waste Li-ion battery (LIBs) cells fed to the process, represented with the most significant impact categories based on the normalization of the sensitivity analysis. Environmental Footprint (EF) 3.0 Method v1.0 of Simapro v9.6 was used for the assessment

Table 8 The calculated composition of the waste NMC622 LIBs cells based on the indicated references

Assumed phases	Wang et al. (2016)	Mohr et al. (2020)
Al	8.09%	4.59%
Al2O3	0.43%	1.12%
Cu	16.31%	22.61%
C	22.46%	16.38%
LiPF6	14.66%	15.54%
PVDF	2.84%	2.14%
PPE	4.96%	2.67%
LiNi0.6Mn0.2 Co0.2O2	30.26%	34.94%
TOTAL	100.0%	100.00%

processes much like the base scenario. More details can be found in Online Resource No.2. Based on this calculation, the provided cell composition of Wang et al. (2016) (i.e., base scenario) contained more Al and C than the one based on Mohr and colleagues but a lower amount of CAM (Table 8).

Sensitivity analysis of the LCIA showed that for most of the impact categories the process that treated the LIBs cells based on Mohr and colleagues' composition showed higher total impacts except for non-cancerous HTP (Fig. 16). Significant difference (i.e., > 10%) was also observed for climate change (GWP) and fossil resource use categories. This is due to the higher amount of coke needed by the process since the cells contain less C and Al.

4 Discussions

4.1 Simulation data reliability

Recall that the objective of this study is to examine the advantages and disadvantages of using process simulation-based LCA as an eco-design tool during the conceptualization stage of recycling processes. This means that no laboratory or industry data is available yet and so the simulated process informed the LCA. However, there are several factors that prevent simulated values to fully reflect real data, and so the reliability of the simulation results needed to be verified.

Since the conceptualized hydrometallurgical treatment of waste LIBs was a combination of specific unit processes (i.e., mechanical pre-treatment, leaching, purification, and two-step precipitation), no existing process employed the exact same combination. So, it is not possible to directly compare the simulated recovery to those found in literature. Still some references used in designing the processes and setting the parameters have reported yields that could be compared to the simulated results. For example, in the leaching step, the temperature and pH used in the simulation were based on experiments by Guimarães et al. (2022) that reported 100% leaching yields for Ni, Co, and Li while 93% for Mn. In the case of the simulated process, all of these elements have 100% leaching yields (see Online Resource No.2).

For pyrometallurgical recycling treatment of batteries, general yields for Ni, Co, and Cu are reported to be > 95%

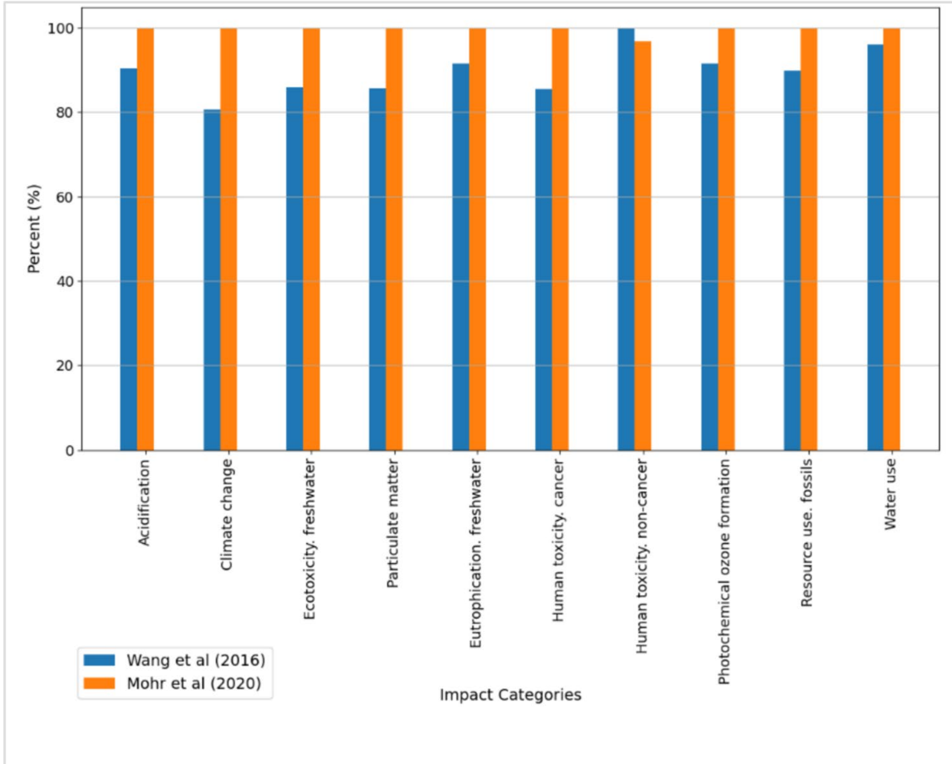


Fig. 16 A comparative bar chart for the simulated pyrometallurgical recycling process varying the reference for the composition of the waste Li-ion battery (LIBs) cells fed to the process, represented with the most significant impact categories based on the normalized results. Environmental Footprint (EF) 3.0 Method v1.0 of Simapro v9.6 was used for the assessment

(Brückner et al. 2020). This aligns with the observed recoveries, i.e., 100% in the simulation as shown in Table 6. In actual practice, there are several operational and physicochemical factors that could affect the separation of the slag and the alloys which mean that recovery would not be 100%. An example operational challenge is related to the fluidity of the melt which could cause stickiness that affects the reduction process during smelting and even losses of the metal when the furnace is tapped (Wang et al. 2024). There are other simulation approaches that incorporate methods like computational dynamic fluid (CFD), and dynamic modelling that could address this better (Huda et al. 2012; Laputka and Xie 2021) and though out of the scope of this current study, further research on this could be interesting.

Another likely source of disparity between the simulated results and in actual practice is the choice made to treat waste LIBs cells instead of modules or packs. The choice was made in order to minimize the number of impurities that could hinder the subsequent processes. However, for ease of operation, process owners in practice prefer to feed packs or modules into a furnace. Though it would likely add additional source of reductants, for example the pack casing, the other reaction gases when plastic is melted could be a source of potential impact. All these other parts could also potentially affect how the reduction reaction will go, thus affecting final recovery.

To further confirm the reliability of the simulation data, a comparison of this study to that of existing LCA for LIBs recycling, specifically those assessing hydrometallurgical and pyrometallurgical processes tested in lab, pilot, or industrial scale (i.e., primary data), was attempted. However, due to several differences in the process and assessment approach (e.g., LIB type feed, system boundaries, functional unit, and LCIA methods used), a direct correlation could not be made between the resulting potential impacts of these processes.

However, the inventory from the literature data can be used to confirm the inventory data generated in this study. For Case Study No. 1, focus is given on the amount of lixiviant used since it has the highest contribution to the potential environmental impact of the hydrometallurgical recycling process conceptualized here. In this case study, H_2SO_4 was the lixiviant used which aligned with most of the literature cited as shown in Table 9. In this table, information about the feed, lixiviant type, and dosage of LCA studies that used primary data are presented along with those that also used process simulation to generate their inventory. Compared to other studies, the amount of lixiviant (i.e., H_2SO_4) used in this study is higher compared to the average dosage used in literature even with the one from simulation. The difference in the actual process flow assessed and the feed composition possibly factored into this. Most of the processes have also been optimized to maximize recovery and efficiency. Even the previous studies that used simulated data in the assessment based their simulation on laboratory experiments of that

Table 9 A comparison of the hydrometallurgical recycling processes assessed by existing literature and the existing study

Reference	Process feed, Leaching feed	Data source	Lixiviant	Dosage
Case Study No. 1	NMC 622 Cells, BM	Process simulation	Sulfuric acid	6.6 g/g BM feed
Anwani et al. (2020)	LCO, cathode powder	Lab experiments	Sulfuric acid	2.49 g/g cathode powder
Du et al. (2022)	LIBs, not specified	Industrial, China	Sulfuric acid	2.78 g/g cathode powder
Duarte Castro et al. (2022)	NMC 111 battery packs, cathode powder	Lab experiments	Citric acid	19.23 g/g cathode powder
Mohr et al. (2020)	NMC, not specified	Industrial, Duesenfield GmbH	Sulfuric acid	0.80 g/g battery cell
Rinne et al. (2024)	NMC, BM	Process simulation	Sulfuric acid	1.88 g/g BM
Rinne et al. (2021b)	Mixed LIBs, not specified	Process simulation	Sulfuric acid	3.15 g/g mixed feed
Zhang et al. (2024)	NMC111, not specified	Industrial, Anhui Tech Company	Hydrochloric acid	0.42 g/g battery cell

specific process flow (Rinne et al. 2021b, 2024), while in this study, the process parameters were based on several references that when combined in this case was not the best available approach.

This shows the importance of simultaneously informing the simulation model with confirmatory experiments. Ideally, after the initial conceptualization and assessment, laboratory experiments will be conducted to confirm the results of the simulation and optimize the process parameters. This would allow for simultaneous assessment of the potential impacts of the process as changes are implemented to improve its efficiency while making sure that burden shifting is prevented. This is however currently out of the scope of this study but would be an interesting further study.

For the simulated inventory of the pyrometallurgical recycling process, i.e., Case Study No. 2, the same approach was used to also confirm the reliability of the results. Since some of the literature found did not use coke in the processes assessed, this made the comparison difficult. For those that used coke in their process, its ratio with the amount of feed is significantly lower than the simulated process in this study as shown in Table 10. However, it is important to note that other than a difference in the pyrometallurgical process type used, the process feed or type of LIBs being treated are also different, which in turn requires varying amount of reagents. There is therefore a huge room for improvement in terms of efficiency of the process that was conceptualized in this study. As noted previously for the hydrometallurgical simulation, the need of laboratory or other simulation modelling approach to better inform the inventory would also be needed for the pyrometallurgical process.

4.2 Interpretation of LCA results—case study no. 1

One difference between the simulation and Ecoinvent hydrometallurgy-based recycling of waste LIBs is the

Table 10 A comparison of the pyrometallurgical recycling processes assessed by existing literature and the existing study

Reference	Process feed	Process type and temperature	Data source	Main reductant	Coke:feed ratio
Case Study No. 2	NMC 622 cells	Smelting, 1300 °C	Process simulation	Coke	63.92 kg/kg cell
Lu and Wang (2024)	LCO electrodes	Smelting, n/a	Lab experiments	Coke	0.33 kg/kg electrode
Liu et al. (2024)	NMC 532 cathode	Pyrolysis, 650 °C	Literature	Coke	0.04 kg/kg CAM
Blömeke et al. (2022)	NMC 622 packs	Smelting, n/a	Literature	n/a	n/a
Feng et al. (2022)	NCM cells	Smelting, n/a	Literature	Coke	0.13 kg/kg cells
Kallitsis et al. (2022)	NMC 333 cells	Smelting, n/a	Literature	n/a	n/a
Quan et al. (2022)	NCM	Smelting, n/a	Literature	n/a	n/a

treatment of the Cu and Al scrap. For the simulated model, the Cu and Al contained in the LIBs cells were not separated out of the coarse fraction after screening and were considered recyclable. This is commonly done in industrial practice, i.e., scraps are set aside until there is enough for further processing. On the other hand, it is considered waste in the Ecoinvent dataset (Hischier 2012) which resulted to their treatment contributing to the over-all potential impacts of the Ecoinvent recycling process.

Also, the simulated recycling process is assessed under the assumption that all the process water used are reused and not discharged thus resulting in only air emissions, while the Ecoinvent have both water and air emissions. Nonetheless, the results of the analysis for the Ecoinvent dataset showed insignificant contributions from the process emissions. Note that due to the software limitation, there are other factors that affect the reactions in these processes, like agitation, which is not considered in the simulation. Since only thermodynamic requirements for the reactions to be possible is being considered and not the kinetics, this could lead to a higher consumption of reagents in the simulation compared to what is actually consumed in a pilot or industrial scale process. This could also be the reason why the total potential impacts for the simulated process is generally higher than the Ecoinvent dataset, e.g., 0.23 kg H_2SO_4/kg of treated waste LIBs based on the Ecoinvent data while 1.61 kg H_2SO_4 was used in the simulated process per kg of waste LIBs treated (see Online Resource No.2).

With regards to the main contributor, both the simulated process and Ecoinvent showed H_2SO_4 as one of the highest contributors to most impact categories. It should however be noted that in the Ecoinvent dataset, the actual inorganic chemicals used in the process are not specified and instead grouped together. This included caustic soda and 20 other different chemicals from different regional markets. Sulfuric acid, the lixiviant for leaching, and soda ash, the pH regulator and precipitant for Li_2CO_3, are also included in this "inorganic chemicals" list which likely resulted to double counting.

Though assessing the process using avoided burden approach showed significant benefits due to the "avoided" primary products, the results should be carefully presented in order avoid misrepresenting the actual potential impacts of the process. This is especially important when eco-designing the process. The cut-off approach or others that focuses more on the impacts without taking the credits of the products would be able to give a more holistic perspective that will fit the objectives of the conceptualization and eco-design phase of any process.

4.3 Interpretation of LCA results—case study no. 2

For the pyrometallurgy-based recycling of waste LIBs, since coke had the highest contribution to most of the impact categories, minimizing its use should be considered to help lower its share to the total impact of the process. This result also confirms the concern for using coke in smelting processes. In reducing the coke consumption, it means that other carbon sources or reductants, with lower potential impacts, should be fed in the process to assist or even fully replace it. The waste LIBs itself have C-containing components as well as Al, which acted as reductant (Makuza et al. 2021). Industrial practice sometimes feed scrap metals to the process to improve the efficiency. However, due to the criticality of Al metals, other types of scrap are better used for this purpose.

With regards to the varied CAM chemistries of the waste NMC LIBs treated being changed, recall that no significant difference to the impact categories were observed. It has been noted though that pyrometallurgical recycling of LIBs is more economical for LIBs containing more Ni and Co (Gaines 2014) which means that mixing various NMC cells as feed to a pyrometallurgical treatment could be an option. The possibility to treat several types of battery chemistry is commonly cited as an advantage in pyrometallurgical recycling processes.

The recovered alloy in pyrometallurgical recycling however will require further treatment, commonly hydrometallurgy which is currently practiced industrially (Assefi et al. 2020; Makuza et al. 2021), in order to recover the Ni, Co, and Cu for economical use. For Li, which is one of the CRMs found in LIBs but lost in the slag in this case, recall that more than 30% of it is observed to go to the flue gases (Table 7). This is also observed in real practice (Stallmeister and Friedrich 2023) which is why in other cases, instead of the slag, the flue dusts are subjected to hydrometallurgical process to recover the Li (Hu et al. 2021).

The loss of the critical Li material and the use of coke (i.e., highest potential contributor to the process' impacts) are subjects of concern in the current pyrometallurgical conceptualized here. This means that other pyrometallurgical processes will need to be tested or even be combined with other approaches to optimize the recycling of waste LIBs.

4.4 Strengths and limitations of process simulation-based LCA

Based on the results and observations made in conducting this study, several advantages and limitations of applying process-simulation based LCA have been observed. One advantage of using digital simulations to gather data for the inventory, especially during the conceptualization stage of a process, is the identification of the possible process air and water emissions that are thermodynamically possible with the given process inputs. This information on the possible composition of the emissions could serve as a guide on choosing the treatment approach that will be required to meet discharge quality standards and, in turn, decrease potential environmental impacts of the process.

Note that in this assessment, the treatment of the process emissions prior to discharge was not included in the system boundary, i.e., they are recycled back into the process for reuse. If included, the resources required for the discharge treatment will also have a burden share on the over-all potential impacts of the recycling process. The downside to this is the need for deep knowledge and understanding of the process chemistries and reactions so that the process inputs indicated in the simulation are as close as possible to reality. This emphasizes the need for close collaboration between process engineers and LCA practitioners when designing processes, not just for recycling.

Another advantage is that the thermodynamic feasibility of the processes being designed can be confirmed through these simulations. However, there are several parameters during actual operation that could affect efficiency and yield, which cannot be accommodated in the digital simulation depending on the software being used. As observed from the simulations, recovery was highly affected by the amount of reagents used. This is not necessarily true in actual practice due to other factors that could be adjusted without adjusting the amount of input (e.g., agitation) and still achieve the same recovery due to the improved reaction rates. This will clearly affect the resulting data for the LCI, which could either over- or underestimate consumption and emissions and, in turn, the resulting potential impacts. Validating how changing the amount of reactants participating in the process reaction will affect the recovery, as well as the potential impacts of the process, could be assessed in future studies.

This is still another advantage of using digital process simulation-based LCA. Through simulation, one is able to test different parameters which sometimes are not easily possible in a laboratory or even in a pilot-scale setting. This is commonly the case for high-temperature processes, which could be hazardous, or those using reagents that are too reactive or expensive. This digital "experiments" like the other insights gained from the simulations would need to be supplemented by real-world data (i.e., pilot or industrial-scale data) to further inform and optimize the inventory of the process and the subsequent LCIA.

5 Conclusions

In this study, the potential environmental impacts of different recycling processes for waste Li-ion batteries were assessed using an approach that utilized digital process simulation to supplement the inventory phase of the LCA, also referred as process simulation-based LCA. Identifying the advantages and disadvantages of this approach as an eco-design tool was the main goal of this assessment especially when used on processes that are still on its conceptualization phase, i.e., no laboratory, field, or real-time data yet.

From the results, it can be concluded that the use of digital process simulation-based LCA can be an effective approach in designing processes especially in considering its potential environmental impacts. The simulations allow the confirmation of the thermodynamic feasibility of the process being designed as well as vary several process parameters that would normally take hours or days if conducted as a laboratory experiment. The assessment of the potential environmental impacts of these processes can also be done simultaneously with these digital experiments, ensuring that the potential burdens of any process changes are monitored and considered for the final process design.

There is, however, a clear need for collaboration between process engineers and LCA practitioners to make sure that a well-informed digital simulation is used for the assessment. Since the simulation is mainly focused on the thermodynamic feasibility of the process, the effects of other factors that could affect the reaction kinetics, like agitation and particle size, need to be confirmed through laboratory experiments. There have been other studies that combined several other simulation approaches and software to generate a more informed material inventory data, and applying the same approach in combination of an environmental analysis is a possible course of action to further improve simulation-based LCA approach for the purpose of process conceptualization and design.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s11367-025-02478-z.

Funding No research grant solely intended to conduct this research was received. This study was conducted as part of CISTEMEEC, one of the projects under the Reverse Metallurgy + portfolio which is co-funded by the European Union NextGeneration and the Region of Wallonia.

Data availability Process simulation.

Process simulation models used to generate the inventory for the simulated recycling processes and used to generate the data presented in Figs. 7 and 14 are available upon request. The same simulations were used to generate the inventory used in the LCA that resulted to the LCIA results shown in Figs. 9, 10, 11, 12, 13, 15, and 16.

LCIA results.

LCI results used for the LCIA of the simulated recycling processes are available upon request as well as the detailed values for the LCIA results presented. For the LCIA results presented in Fig. 9b the data are based on existing datasets on Ecoinvent, specifically (Hischier 2012).

Declarations

Competing interests The authors declare no competing interests.

References

Anwani S, Methekar R, Ramadesigan V (2020) Resynthesizing of lithium cobalt oxide from spent lithium-ion batteries using an environmentally benign and economically viable recycling process. Hydrometallurgy 197:105430. https://doi.org/10.1016/j.hydromet.2020.105430

Aromaa R, Rinne M, Lundström M (2022) Comparative life cycle assessment of hardmetal chemical recycling routes. ACS Sustain Chem Eng 10:10234–10242. https://doi.org/10.1021/acssuschemeng.2c01969

AsadiDalini E, Karimi Gh, Zandevakili S, Goodarzi M (2021) A review on environmental, economic and hydrometallurgical processes of recycling spent lithium-ion batteries. Miner Process Extr Metall Rev 42:451–472. https://doi.org/10.1080/08827508.2020.1781628

Assefi M, Maroufi S, Yamauchi Y, Sahajwalla V (2020) Pyrometallurgical recycling of Li-ion, Ni–Cd and Ni–MH batteries: a minireview. Curr Opin Green Sustain Chem 24:26–31. https://doi.org/10.1016/j.cogsc.2020.01.005

Bartie N, Cobos-Becerra L, Frohling M et al (2021) Process simulation and digitalization for comprehensive life-cycle sustainability assessment of Silicon photovoltaic systems. In: 2021 IEEE 48th Photovoltaic Specialists Conference (PVSC). IEEE, Fort Lauderdale, FL, USA, pp 1244–1249. https://doi.org/10.1109/PVSC43889.2021.9518984

Battaglia G, Berkemeyer L, Cipollina A et al (2022) Recovery of lithium carbonate from dilute Li-rich brine via homogenous and heterogeneous precipitation. Ind Eng Chem Res 61:13589–13602. https://doi.org/10.1021/acs.iecr.2c01397

Bernstein WZ, Tamayo CD, Lechevalier D, Brundage MP (2019) Incorporating unit manufacturing process models into life cycle assessment workflows. Proc CIRP 80:364–369. https://doi.org/10.1016/j.procir.2019.01.019

Blömeke S, Scheller C, Cerdas F et al (2022) Material and energy flow analysis for environmental and economic impact assessment of industrial recycling routes for lithium-ion traction batteries. J Clean Prod 377:134344. https://doi.org/10.1016/j.jclepro.2022.134344

Brückner L, Frank J, Elwert T (2020) Industrial recycling of lithium-ion batteries—a critical review of metallurgical process routes. Metals 10:1107. https://doi.org/10.3390/met10081107

Cao Y, Li L, Zhang Y et al (2023) Co-products recovery does not necessarily mitigate environmental and economic tradeoffs in lithium-ion battery recycling. Resour Conserv Recycl 188:106689. https://doi.org/10.1016/j.resconrec.2022.106689

Chen Q, Lai X, Hou Y et al (2023) Investigating the environmental impacts of different direct material recycling and battery remanufacturing technologies on two types of retired lithium-ion batteries from electric vehicles in China. Sep Purif Technol 308:122966. https://doi.org/10.1016/j.seppur.2022.122966

Cosate De Andrade MF, Souza PMS, Cavalett O, Morales AR (2016) Life cycle assessment of poly(lactic acid) (PLA): comparison between chemical recycling, mechanical recycling and composting. J Polym Environ 24:372–384. https://doi.org/10.1007/s10924-016-0787-2

Du S, Gao F, Nie Z et al (2022) Life cycle assessment of recycled NiCoMn ternary cathode materials prepared by hydrometallurgical technology for power batteries in China. J Clean Prod 340:130798. https://doi.org/10.1016/j.jclepro.2022.130798

Duarte Castro F, Mehner E, Cutaia L, Vaccari M (2022) Life cycle assessment of an innovative lithium-ion battery recycling route: a feasibility study. J Clean Prod 368:133130. https://doi.org/10.1016/j.jclepro.2022.133130

Ekvall T, Björklund A, Sandin G, Lage J (2020) Modeling recycling in life cycle assessment. Report Number: 2020:05. https://www.lifecyclecenter.se/publications/modeling-of-recycling-in-life-cycle-assessment/

Elia Group (2023) Belgium's 2022 electricity mix: the increase in renewable energy and availability of nuclear power plants kept exports high. https://www.elia.be/en/press/2023/01/20230106_energymix2022#

Ellingsen LA-W, Majeau-Bettez G, Singh B et al (2014) Life cycle assessment of a lithium-ion battery vehicle pack. J Ind Ecol 18:113–124. https://doi.org/10.1111/jiec.12072

Elomaa H, Rintala L, Aromaa J, Lundström M (2020a) Process simulation based life cycle assessment of cyanide-free refractory gold concentrate processing–case study: cupric chloride leaching. Miner Eng 157:106559. https://doi.org/10.1016/j.mineng.2020.106559

Elomaa H, Sinisalo P, Rintala L et al (2020b) Process simulation and gate-to-gate life cycle assessment of hydrometallurgical refractory gold concentrate processing. Int J Life Cycle Assess 25:456–477. https://doi.org/10.1007/s11367-019-01723-6

Elwert T, Römer F, Schneider K et al (2018) Recycling of batteries from electric vehicles. In: Pistoia G, Liaw B (eds) Behaviour of lithium-ion batteries in electric vehicles. Springer International Publishing, Cham, pp 289–321

EU Urban Mobility Observatory (2022) Is there life after death for Europe's lithium-ion batteries? In: EU urban mobility observatory. https://urban-mobility-observatory.transport.ec.europa.eu/news-events/news/there-life-after-death-europes-lithium-ion-batteries-2022-06-30_en. Accessed 17 Jul 2024

European Parliament, Council of the European Union (2006) Directive 2006/66/EC of the European Parliament and of the Council of 6 September 2006 on batteries and accumulators and waste batteries and accumulators and repealing Directive 91/157/EEC. https://eur-lex.europa.eu/eli/dir/2006/66/oj/eng

European Parliament, Council of the European Union (2008) Directive 2008/98/EC. https://eur-lex.europa.eu/eli/dir/2008/98/oj/eng

European Parliament, Council of the European Union (2023) Regulation (EU) 2023/1542 of the European Parliament and of the Council of 12 July 2023 concerning batteries and waste batteries, amending Directive 2008/98/EC and Regulation (EU) 2019/1020 and repealing Directive 2006/66/EC. https://eur-lex.europa.eu/eli/reg/2023/1542/oj/eng

Feng T, Guo W, Li Q et al (2022) Life cycle assessment of lithium nickel cobalt manganese oxide batteries and lithium iron phosphate batteries for electric vehicles in China. J Energy Storage 52:104767. https://doi.org/10.1016/j.est.2022.104767

Fisher K, Wallén E, Laenen PP, Collins M (2006) Battery waste management life cycle assessment: final report. Environmental Resources Management Ltd. https://www.epbaeurope.net/assets/resources/090607_2006_Oct.pdf

Flash Battery SrL (2022) Which chemistry is most suitable for the electrification of your vehicle? Let's discover the different types of batteries. https://www.flashbattery.tech/en/types-of-lithium-batteries-which-chemistry-use/

Gaines L (2014) The future of automotive lithium-ion battery recycling: charting a sustainable course. Sustain Mater Technol 1–2:2–7. https://doi.org/10.1016/j.susmat.2014.10.001

U.S. Geological Survey (2023) Mineral commodity summaries 2023: U.S. Geological Survey. https://doi.org/10.3133/mcs2023

Geoscience Australia (2022) Critical minerals at Geoscience Australia. In: www.ga.gov.au. https://www.ga.gov.au/scientific-topics/minerals/critical-minerals

Gnansounou E, Vaskan P, Pachón ER (2015) Comparative techno-economic assessment and LCA of selected integrated sugarcane-based biorefineries. Biores Technol 196:364–375. https://doi.org/10.1016/j.biortech.2015.07.072

Government of Canada (2022) Critical minerals: an opportunity for Canada. In: www.canada.ca. https://www.canada.ca/en/campaign/critical-minerals-in-canada/critical-minerals-an-opportunity-for-canada.html

Grohol M, Veeh C (2023) Directorate-general for internal market, industry, entrepreneurship and SMEs (European Commission) European Commission, study on the critical raw materials for the EU 2023-final report. European Commission. https://op.europa.eu/en/publication-detail/-/publication/57318397-fdd4-11ed-a05c-01aa75ed71a1

Guimarães LF, Botelho Junior AB, Espinosa DCR (2022) Sulfuric acid leaching of metals from waste Li-ion batteries without using reducing agent. Miner Eng 183:107597. https://doi.org/10.1016/j.mineng.2022.107597

Gupta V, Appleberry M, Li W, Chen Z (2024) Direct recycling industrialization of Li-ion batteries: the pre-processing barricade. Next Energy 2:100091. https://doi.org/10.1016/j.nxener.2023.100091

Han B, Anwar UiHaq R, Louhi-Kultanen M (2020) Lithium carbonate precipitation by homogeneous and heterogeneous reactive crystallization. Hydrometallurgy 195:105386. https://doi.org/10.1016/j.hydromet.2020.105386

Harper G, Sommerville R, Kendrick E et al (2019) Recycling lithium-ion batteries from electric vehicles. Nature 575:75–86. https://doi.org/10.1038/s41586-019-1682-5

Heeren N, Mutel CL, Steubing B et al (2015) Environmental impact of buildings—what matters? Environ Sci Technol 49:9832–9841. https://doi.org/10.1021/acs.est.5b01735

Hischier R (2012) Treatment of used Li-ion battery, hydrometallurgical treatment, GLO. Ecoinvent dataset.

Hu X, Mousa E, Ye G (2021) Recovery of Co, Ni, Mn, and Li from Li-ion batteries by smelting reduction-part II: a pilot-scale demonstration. J Power Sources 483:229089. https://doi.org/10.1016/j.jpowsour.2020.229089

Huda N, Naser J, Brooks G et al (2012) Computational fluid dynamic modeling of zinc slag fuming process in top-submerged lance smelting furnace. Metall Mater Trans B 43:39–55. https://doi.org/10.1007/s11663-011-9558-6

IEA (2022) Energy system of Belgium. https://www.iea.org/countries/belgium

ISO 14040 (2006) Environmental management—life cycle assessment—principles and framework

Jolliet O, Saadé-Sbeith M, Shaked S et al (2016) Expert judgment and default uncertainty estimates. In: Environmental life cycle assessment. CRC Press, Taylor & Francis Group, Boca Raton, p 169

Jung J, Zhang J (2022) Current commercial hydrometallurgical recycling process. Hydrometallurgical recycling of lithium-ion battery materials, 1st edn. CRC Press, Boca Raton, pp 40–73

Jung JC-Y, Sui P-C, Zhang J (2021) A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments. J Energy Storage 35:102217. https://doi.org/10.1016/j.est.2020.102217

Jung J, Sui P-C, Zhang J (2023) Hydrometallurgical recycling of lithium-ion battery materials, 1st edn. CRC Press, Boca Raton

Kallitsis E, Korre A, Kelsall GH (2022) Life cycle assessment of recycling options for automotive Li-ion battery packs. J Clean Prod 371:133636. https://doi.org/10.1016/j.jclepro.2022.133636

Kim M, Kim J, Kim J et al (2024) Modeling framework for optimization of the life cycle sustainability of lithium-ion batteries by nickel manganese cobalt recycling process. In: Computer Aided Chemical Engineering. Elsevier, pp 1105–1110. https://yonsei.elsevierpure.com/en/publications/modeling-framework-for-optimization-of-the-life-cycle-sustainabil

Kniel GE, Delmarco K, Petrie JG (1996) Life cycle assessment applied to process design: environmental and economic analysis and optimization of a nitric acid plant. Environ Prog 15:221–228. https://doi.org/10.1002/ep.670150410

Koroma MS, Costa D, Philippot M et al (2022) Life cycle assessment of battery electric vehicles: implications of future electricity mix and different battery end-of-life management. Sci Total Environ 831:154859. https://doi.org/10.1016/j.scitotenv.2022.154859

Korthauer R (ed) (2019) Lithium-ion batteries: basics and applications. Springer-Verlag GmbH Germany. https://doi.org/10.1007/978-3-662-53071-9

Laputka M, Xie W (2021) A review of recent advances in pyrometallurgical process measurement and modeling, and their applications to process improvement. Min Metall Explor 38:1135–1165. https://doi.org/10.1007/s42461-021-00386-y

Liu Z, Sederholm JG, Lan K-W et al (2023) Life cycle assessment of hydrometallurgical recycling for cathode active materials. J Power Sources 580:233345. https://doi.org/10.1016/j.jpowsour.2023.233345

Liu K, Xu Z, Wang M et al (2024) Mechanisms of thermal decomposition in spent NCM lithium-ion battery cathode materials with carbon defects and oxygen vacancies. Environ Sci Technol 58:21362–21373. https://doi.org/10.1021/acs.est.4c06562

Lu Y, Wang J (2024) Life cycle assessment for spent lithium-ion batteries' recycling process: environmental impact, energy consumption, and sensitivity analysis. ACS Sustainable Chem Eng 12:12966–12975. https://doi.org/10.1021/acssuschemeng.4c04541

Makuza B, Tian Q, Guo X et al (2021) Pyrometallurgical options for recycling spent lithium-ion batteries: a comprehensive review. J Power Sources 491:229622. https://doi.org/10.1016/j.jpowsour.2021.229622

Malmgren S, Ciosek K, Lindblad R et al (2013) Consequences of air exposure on the lithiated graphite SEI. Electrochim Acta 105:83–91. https://doi.org/10.1016/j.electacta.2013.04.118

Meshram P, Pandey BD, Mankhand TR (2015) Hydrometallurgical processing of spent lithium ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching. Chem Eng J 281:418–427. https://doi.org/10.1016/j.cej.2015.06.071

Mohr M, Peters JF, Baumann M, Weil M (2020) Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes. J Ind Ecol 24:1310–1322. https://doi.org/10.1111/jiec.13021

Mousa E, Hu X, Ånnhagen L et al (2022) Characterization and thermal treatment of the black mass from spent lithium-ion batteries. Sustainability 15:15. https://doi.org/10.3390/su15010015

Mousavinezhad S, Kadivar S, Vahidi E (2023) Comparative life cycle analysis of critical materials recovery from spent Li-ion batteries. J Environ Manag 339:117887. https://doi.org/10.1016/j.jenvman.2023.117887

Mutel C (2017) Brightway: an open source framework for life cycle assessment. JOSS 2:236. https://doi.org/10.21105/joss.00236

Ng ZW, Gan HX, Putranto A et al (2023) Process design and life cycle assessment of furfural and glucose co-production derived from palm oil empty fruit bunches. Environ Dev Sustain 25:13937–13958. https://doi.org/10.1007/s10668-022-02633-8

Nordelöf A, Poulikidou S, Chordia M et al (2019) Methodological approaches to end-of-life modelling in life cycle assessments of lithium-ion batteries. Batteries 5:51. https://doi.org/10.3390/batteries5030051

Pavón S, Kaiser D, Mende R, Bertau M (2021) The cool-process—a selective approach for recycling lithium batteries. Metals 11:259. https://doi.org/10.3390/met11020259

Pell R, Wall F, Yan X et al (2019) Mineral processing simulation based-environmental life cycle assessment for rare earth project development: a case study on the Songwe Hill project. J Environ Manage 249:109353. https://doi.org/10.1016/j.jenvman.2019.109353

Premathilake DS, Ambaye TG, Botelho Junior AB et al (2024) Comparative environmental and economic assessment of emerging hydrometallurgical recycling technologies for Li-ion battery cathodes. Sustain Prod Consum 51:327–344. https://doi.org/10.1016/j.spc.2024.09.015

Quan J, Zhao S, Song D et al (2022) Comparative life cycle assessment of LFP and NCM batteries including the secondary use and different recycling technologies. Sci Total Environ 819:153105. https://doi.org/10.1016/j.scitotenv.2022.153105

Rada S, Cuibus D, Vermesan H et al (2018) Structural and electrochemical properties of recycled active electrodes from spent lead acid battery and modified with different manganese dioxide contents. Electrochim Acta 268:332–339. https://doi.org/10.1016/j.electacta.2018.02.135

Reuter MA (1998) The simulation of industrial ecosystems. Miner Eng 11:891–918. https://doi.org/10.1016/S0892-6875(98)00078-8

Reuter MA, van Schaik A (2015) Product-centric simulation-based design for recycling: case of LED lamp recycling. J Sustain Metall 1:4–28. https://doi.org/10.1007/s40831-014-0006-0

Rinne M, Elomaa H, Lundström M (2021a) Life cycle assessment and process simulation of prospective battery-grade cobalt sulfate production from Co-Au ores in Finland. Int J Life Cycle Assess 26:2127–2142. https://doi.org/10.1007/s11367-021-01965-3

Rinne M, Elomaa H, Porvali A, Lundström M (2021b) Simulation-based life cycle assessment for hydrometallurgical recycling of mixed LIB and NiMH waste. Resour Conserv Recycl 170:105586. https://doi.org/10.1016/j.resconrec.2021.105586

Rinne M, Aromaa-Stubb R, Elomaa H et al (2024) Evaluation of hydrometallurgical black mass recycling with simulation-based life cycle assessment. Int J Life Cycle Assess 29:1582–1597. https://doi.org/10.1007/s11367-024-02304-y

Saaid FI, Kasim MF, Winie T et al (2024) Ni-rich lithium nickel manganese cobalt oxide cathode materials: a review on the synthesis methods and their electrochemical performances. Heliyon 10:e23968. https://doi.org/10.1016/j.heliyon.2023.e23968

Salgado RM, Danzi F, Oliveira JE et al (2021) The latest trends in electric vehicles batteries. Molecules 26:3188. https://doi.org/10.3390/molecules26113188

Segura-Salazar J, Lima FM, Tavares LM (2019) Life Cycle Assessment in the minerals industry: current practice, harmonization efforts, and potential improvement through the integration with process simulation. J Clean Prod 232:174–192. https://doi.org/10.1016/j.jclepro.2019.05.318

Spengler T, Geldermann J, Hähre S et al (1998) Development of a multiple criteria based decision support system for environmental assessment of recycling measures in the iron and steel making industry. J Clean Prod 6:37–52. https://doi.org/10.1016/S0959-6526(97)00048-6

Stallmeister C, Friedrich B (2023) Efficient lithium recovery from end-of-life batteries in pyrometallurgical recycling processes by early-stage separation from black mass. In: Metallurgy and materials society of the Canadian institute of mining metallurgy and petroleum (CIM) (ed) proceedings of the 62nd conference of metallurgists, COM 2023. Springer Nature Switzerland, Cham, pp 727–737. https://doi.org/10.1007/978-3-031-38141-6_97

Tas G, Klemettinen A, Serna-Guerrero R (2024) Circular and sustainable: evaluating lithium-ion battery recycling using a combined statistical entropy and life cycle assessment methodology. ChemSusChem 17:e202400376. https://doi.org/10.1002/cssc.202400376

Thompson DL, Hartley JM, Lambert SM et al (2020) The importance of design in lithium ion battery recycling–a critical review. Green Chem 22:7585–7603. https://doi.org/10.1039/D0GC02745F

Toro L, Moscardini E, Baldassari L et al (2023) A systematic review of battery recycling technologies: advances, challenges, and future prospects. Energies 16:6571. https://doi.org/10.3390/en16186571

Treyer K (2023) Market for electricity, medium voltage. Ecoinvent dataset.

UNEP (2001) Eco-design. In: European Environment Agency. https://www.eea.europa.eu/help/glossary/eea-glossary/eco-design

Velázquez-Martínez O, Valio J, Santasalo-Aarnio A et al (2019) A critical review of lithium-ion battery recycling processes from a circular economy perspective. Batteries 5:68. https://doi.org/10.3390/batteries5040068

Verrecht B (2023) Towards sustainable battery recycling–an industrial perspective (*Powerpoint presentation*).

Wagner-Wenz R, van Zuilichem A-J, Göllner-Völker L et al (2023) Recycling routes of lithium-ion batteries: a critical review of the development status, the process performance, and life-cycle environmental impacts. MRS Energy Sustain 10:1–34. https://doi.org/10.1557/s43581-022-00053-9

Wang X, Gaustad G, Babbitt CW (2016) Targeting high value metals in lithium-ion battery recycling via shredding and size-based separation. Waste Manage 51:204–213. https://doi.org/10.1016/j.wasman.2015.10.026

Wang R, Purohit S, Paymooni K, Honeyands T (2024) Sticking in shaft furnace and fluidized bed ironmaking processes: a comprehensive review focusing on the effect of coating materials. Metall Mater Trans B 55:2977–3006. https://doi.org/10.1007/s11663-024-03188-x

Williams E, Eikenaar S (2022) Finding your way in multifunctional processes and recycling. https://pre-sustainability.com/articles/finding-your-way-in-allocation-methods-multifunctional-processes-recycling/

Woeste R, Drude E-S, Vrucak D et al (2024) A techno-economic assessment of two recycling processes for black mass from end-of-life lithium-ion batteries. Appl Energy 361:122921. https://doi.org/10.1016/j.apenergy.2024.122921

Wu F, Li L, Crandon L et al (2022) Environmental hotspots and greenhouse gas reduction potential for different lithium-ion battery recovery strategies. J Clean Prod 339:130697. https://doi.org/10.1016/j.jclepro.2022.130697

Xu Z, Zhiyuan L, Wenjun M, Qinxin Z (2023) Pretreatment options for the recycling of spent lithium-ion batteries: a comprehensive review. J Energy Storage 72:108691. https://doi.org/10.1016/j.est.2023.108691

Zanoletti A, Carena E, Ferrara C, Bontempi E (2024) A review of lithium-ion battery recycling: technologies, sustainability, and open issues. Batteries 10:38. https://doi.org/10.3390/batteries10010038

Zhang Z, Ramadass P (2012) Lithium-ion battery systems and technology. In: Meyers RA (ed) Encyclopedia of Sustainability Science and Technology. Springer, New York, New York, NY, pp 6122–6149

Zhang X, Li L, Fan E et al (2018) Toward sustainable and systematic recycling of spent rechargeable batteries. Chem Soc Rev 47:7239–7302. https://doi.org/10.1039/C8CS00297E

Zhang G, Shi M, Hu X et al (2024) Life cycle assessment of methods for recycling retired ternary lithium batteries. J Energy Storage 89:111815. https://doi.org/10.1016/j.est.2024.111815

Zou H, Gratz E, Apelian D, Wang Y (2013) A novel method to recycle mixed cathode materials for lithium ion batteries. Green Chem 15:1183. https://doi.org/10.1039/c3gc40182k