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Simulation-based life cycle assessment of secondary materials from recycling of lithium-ion batteries

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ABSTRACT

The EU Battery Regulation is aimed at minimizing negative impact of waste batteries on the environment. Recycling of lithium-ion batteries is one way to reduce those impacts. However, a lack of detailed process-level data is limiting the environmental impact assessment. In this study, the necessary data is generated using process simulation, and is used to estimate the material recovery rates and environmental impacts of a recycling nickel-manganese-cobalt-based battery. We apply and allocate the impacts of recycling to determine secondary battery material carbon footprints. The results were compared with that of primary raw materials based on mass-based and economic value-based allocation. In reference scenario, applying economic value-based allocation resulted in cobalt sulphate and nickel sulphate having 73.5 % and 57.4 % lower carbon footprint than their primary, however, lithium carbonate had a 20.8 % higher footprint. The results indicate the need to improve material recoveries for meeting EU Battery Regulation targets, while minimizing environmental impact.

1. Introduction

The emergence of energy storage technologies has the potential to mitigate transport sector-related greenhouse gas emissions but also poses a threat due to adverse environmental impacts (Pehl et al., 2017). Sustainability assessment methodologies can be used to gauge the potential and adversity of emerging technologies (Hauschild et al., 2017; Hellweg & Canals, 2014; Kara et al., 2018). However, the accuracy of these assessments relies on the availability and quality of data, which is often scarce due to factors such as low technology readiness levels, confidential process information, and limited information in the literature (Arvidsson et al., 2018; Hetherington et al., 2014; Thonemann et al., 2020). This scarcity of data restricts the usefulness of environmental sustainability assessments for emerging technologies.

Life cycle assessment (LCA) evaluates environmental impacts of complex product systems, applied to assess emerging technologies like nanomaterials, energy storage, and fuel (Gavankar et al., 2015; Griffiths et al., 2013; Hetherington et al., 2014; Liu & Rajagopal, 2019). Employing LCA early in technology development - be it in lab or pilot scale - offers crucial quantitative estimates, aiding in research prioritization (Bergerson et al., 2020; Shibasaki et al., 2007). LCA assesses environmental impacts and resource use across the entire life cycle, encompassing extraction, processing, use, and disposal stages.

Emerging recycling technologies for lithium-ion batteries (LIBs) lag behind battery production in scale and technology readiness levels (Philippot et al., 2019). Early-phase life cycle assessments (LCAs) of these technologies face challenges due to limited operational scale and lack of primary data. Addressing data scarcity can be achieved through process simulations. To assess recycling benefits, modelling a comprehensive system covering all critical LIB life cycle stages is crucial.

Numerous studies have evaluated the environmental impact and benefits of battery recycling using various methodologies (Bauer et al., 2015; Blömeke et al., 2022; Crenna et al., 2021; Dai et al., 2019; Diekmann et al., 2016; Dunn et al., 2016; Ellingsen et al., 2014; Gaines, 2014; Notter et al., 2010; Peters et al., 2016; Schmidt et al., 2016; Wang et al., 2020). While some studies rely on primary sources and stoichiometric calculations, limiting the ability to optimize life cycle inventories, others have explored process simulation-based data sources.

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However, a limited number of studies have utilized process simulation to generate necessary data for battery life cycles (Rinne et al., 2021).

Existing research provides aggregated data, lacking the granularity needed to analyse impacts at a process level. This study addresses this gap by offering detailed, process-level data on the mechanical and hydrometallurgical recycling of LIBs, as well as the production of nickel-manganese-cobalt oxide (NMC) cathode active material from both primary and recycled materials. In the context of electric vehicle (EV) batteries, NMC stands for "Lithium Nickel Manganese Cobalt Oxide", which is the chemical compound $LiNi_xMn_yCo_{1\text{-}x\text{-}y}O_2$ at the positive electrode in LIBs in which lithium ions are stored; it is commonly used in LIBs for EVs. The numbers following NMC such as 111, 622, and 811 represent the stoichiometric ratio of the nickel (x), manganese (y), and cobalt (1-x-y) in the battery cathode material, e.g. $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ for NMC622 or $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ for NMC811. The environmental effects of producing NMC compounds, alongside the benefits derived from their recycling, depends strongly on the ratios of the constituent metals. For example, global market average environmental data (Wernet et al., 2016) show that carbon-equivalent emissions associated with the production of Co substantially exceeds that of Mn and Ni, being over eightfold higher than Mn and exceeding Ni by more than 2.5 times. Consequently, NMC materials with a higher proportion of Co not only demonstrate increased carbon footprints but also highlight the enhanced environmental benefit of recycling Co to battery-grade standards, relative to Ni and Mn.

This study uniquely provides inventory data necessary to determine recovery rates and material-specific quality, essential for assessing compliance with the draft of the European Regulation on batteries (Regulation 2023/1542). Furthermore, data from simulations aid in evaluating and reducing the environmental impact of recycling processes and integrating recyclability into product design during early development stages.

The proposed EU Battery Regulation amendment sets specific targets for future battery producers, requiring the recovery of 90 % of cobalt, nickel, and copper from end-of-life batteries by 2027. Manufacturers must disclose a CO_2 footprint per kWh of energy storage and breakdown various life cycle stages. Reliable recycling data is crucial for compliance. While direct measurement is ideal, methods like process simulation can generate necessary mass and energy flow data. Our study uses process simulation models to assess LIB recycling's environmental impact and benefits for NMC production. It provides life cycle inventories, highlighting environmental hotspots, and identifies material recovery-related hotspots. This data helps determine which LIB recycling processes align with EU Battery Regulation targets.

2. Materials and methods

2.1. Simulation-based life cycle assessment

Process simulation is a valuable tool for reliably analysing the mass and energy flows needed to design and optimize industrial plants. The unit processes that constitute these plants are modelled individually and then connected through mass and energy flows to create a simulation of the entire plant. Actual product life cycles consist of, among others, several such plants connected. Therefore, it is beneficial to leverage the capabilities of process simulation to generate the life cycle inventory data needed to perform LCA. Compared to mainstream data collection methods, process simulation provides more rigorous and higher levels of detail, as it can be based on fundamental physics principles.

Process simulation-based LCA ensures a high level of physics-based detail in life cycle inventory data and ensures compliance with the laws of mass and energy conservation as well as the second law of thermodynamics to account for thermodynamic losses throughout the considered process chains (Bartie et al., 2020). This level of detailed data can support the development of integrated computational modelling based on simulation data (Baars et al., 2023; Cerdas, 2022). Furthermore, the fact that the design of new plants is implicitly predictive implies that the simulation approach is particularly suited to prospective LCA in which the availability of historical process data is limited—the fundamental basis also gives simulation models predictive capabilities. The details of the process simulation developed in this study are elaborated in Appendix 1–3.

2.2. Life cycle assessment

2.2.1. Goal, scope, and system boundaries

The functional unit of the study is defined as "The production and recycling of one NMC-based battery pack of 42.2 kWh to produce secondary battery-grade raw materials for direct re-use in battery production." The goal of our study is to evaluate the material-specific recovery rates of a recycling process and to compare results with the targets defined in the EU Battery Regulation. The study also aims to quantify the environmental benefits and impacts of recycling and identify key contributors and material flows in the battery life cycle. Finally, we intend to determine the impact of secondary materials from battery recycling using multi-output allocation procedures, and the resulting impacts are compared with primary routes.

The recycling route comprised dismantling and disassembly followed by pyrolysis, mechanical separation, hydrometallurgy, and product upgradation. The term secondary battery-grade raw materials refers to the materials recovered from recycling activities that are directly usable in battery production by substituting primary raw materials. To enable this direct substitution, product upgrade processes are necessary. The term battery pack comprises the cells, modules (comprised of the cells), and the pack (comprised of the modules) (Harper et al., 2019). The term battery pack would be used in a broad context unless referring to a specific section of the battery. The details of recycling routes used in our study are described in Section *2.4 Process simulations*.

To meet the goals of our study, the system boundaries consider the entire life cycle of a generic NMC-based LIB, including the production and recycling stages as indicated in Fig. 1. It is important to include recycling to determine the respective impacts of secondary material recovered from recycling. The use stage is not considered in the study. It is excluded as the process simulation models concerning recycling do not influence the impacts in the use stage, and the study intends to restrict the focus to battery production and recycling. It is assumed that the production and recycling of the LIB occur in Europe. We use process simulation models to provide data concerning the NMC production, recycling, purification, and treatment of recovered materials. The data concerning cell and battery production are based on the literature (Chordia et al., 2021; Crenna et al., 2021; Dai et al., 2019).

2.2.2. Handling of multi-functionality and allocation

Recycling processes of LIBs are typically multi-output processes since they recover different battery materials (Blömeke et al., 2022; Harper et al., 2019; Sommerville et al., 2021). Due to this multi-functionality of the processes, allocation procedures are necessary to split the environmental impacts of the process between the different materials recovered to estimate the environmental impacts associated with producing each of the secondary materials. The ISO standards suggest different approaches for allocating impacts when the subdivision of processes is not possible (ISO 2006a; ISO 2006b). Since the material recovered first in the hydrometallurgy process chain (lithium carbonate) will have all impacts related to the mechanical treatment and most of the hydrometallurgy process chain, we decided against performing allocation by subdivision. Most applied allocations in the context of recycling are either based on the mass or the economic value of the materials recovered. Both allocation factors are applied herein since they can have a significant influence on the environmental impacts allocated to the materials.

In the study, the term primary impacts of battery materials refer to the market-based dataset available in the ecoinvent database and which

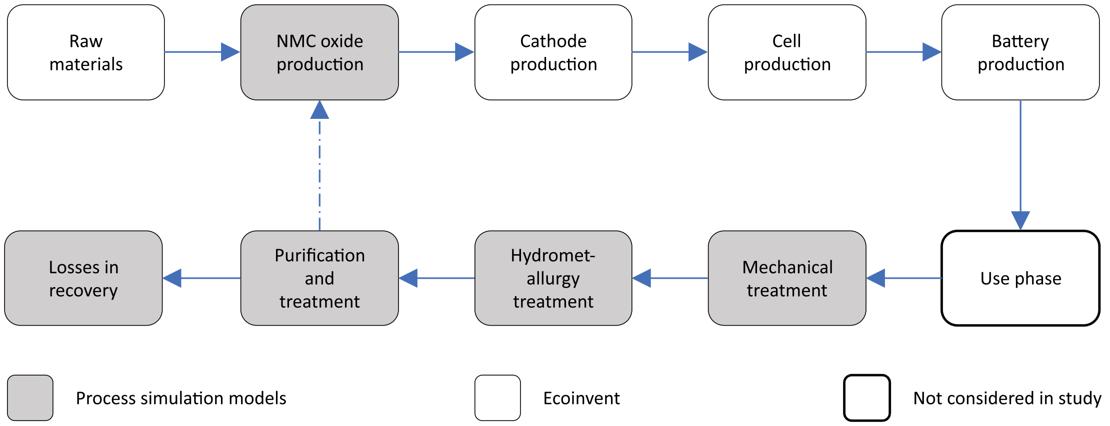


Fig. 1. Process flow diagram indicating the source of data and the system boundaries considered within the herein presented study. The two sources of data used are Ecoinvent and process simulations.

are widely used in current and previous LCA studies. The term secondary impacts refer to the environmental impact of the materials recovered from the recycling process due to application of the allocation procedures. In mass-based allocation, the environmental impact of recycling process is distributed evenly based on the mass of the material recovered. However, in the economic value-based allocation, the environmental impacts are distributed based on an allocation key, which is the product of mass of the material recovered and its economic value. This allocation procedure is preferred as it reflects the market reality of the recycling process focusing on recovery of high value products. It is also worth noting that high fluctuations exist in the prices of certain battery metals. To understand its importance on the secondary impacts, four scenarios, based on prices of battery metals, have been considered in the study. In general, the higher the price of a metal being recovered, the higher its secondary environmental impact will be due to the application of economic value-based allocation. The formula used and calculation can be seen in Appendix 4C.

2.2.3. *LCA software, database, and impact assessment method*

The analysis is carried out via Brightway2 software using background data from the *ecoinvent 3.8 cut-off* database (PRé, 2019; Wernet et al., 2016). Brightway2 is open-source python-based LCA software for conducting environmental impact assessments on products and processes (Mutel, 2017). The Life Cycle Inventory (LCI) data is generated using process simulation software, and unit processes are created as *activities* (terminology from Brightway2). The *activities* comprise of the material and energy flows necessary to represent the unit processes, and appropriate ecoinvent datasets (or *exchanges*) are selected to model them. In this manner, the LCI generated by the process simulation software is used to create unit processes and the appropriate *exchanges* are linked to the ecoinvent database. The *activities* with their corresponding *exchanges* are provided in Appendix 4B.

The life cycle impact assessment (LCIA) was done using the ILCD 2.0 2018 method. This is the method suggested by the Product Environmental Footprint Category Rules (PEFCR) guidelines. This is a set of guidelines for conducting LCA for *High Specific Energy Rechargeable Batteries for Mobile Applications*, which provides a harmonized and transparent method for calculating the environmental impact of LIBs. The following impact categories were selected based on the recommendation provided in the guideline: climate change: fossils (kg CO_2-eq); resources: fossils (MJ); resources: minerals and metals (kg Sb-eq); and human health: respiratory effects, inorganics (disease incidence).

2.3. *Process simulations*

The HSC Chemistry simulation software is used to generate the process-specific data needed to evaluate material recovery rates and to generate the data needed for environmental impact assessment (Bartie et al., 2021; Reuter et al., 2019). The HSC-Sim module includes graphical flowsheet and spreadsheet-type models, with a customizable variable list allowing the creation of diverse process models in chemistry, metallurgy, and mineralogy, where each process unit corresponds to an individual Excel file. The results of the simulation models are converted into life cycle inventory (LCI) models at a process level. These LCI models are then coupled to perform the LCA. Any process changes made in the simulation model will subsequently determine the corresponding changes in environmental impacts. This approach ensures the applicability of environmental impact assessment at a process modelling level and thus aids in evaluating the design and performance of recycling processes in the early stages of process design.

The description of the process simulation models of mechanical treatment, hydrometallurgy treatment, and NMC production are presented in Appendix 1, Appendix 2, and Appendix 3 respectively. The LCI models generated from these simulation models are provided in Appendix 4A, and the Brightway2 models used for the estimation of the environmental impacts are provided in Appendix 4B

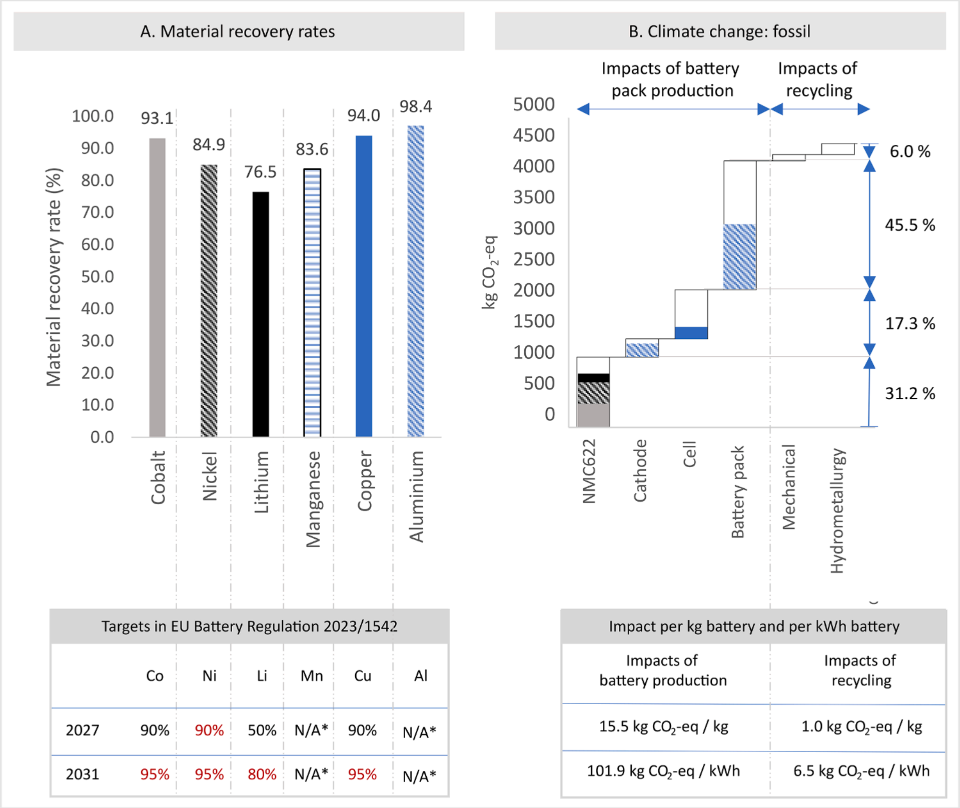
3. Results

This section provides the results to address the goal of the study. The provided results aid to evaluate the potential of the emerging recycling process to achieve the metal recovery targets set out in the EU Battery Regulation. The results also quantify the environmental benefits and impacts of recycling and identify key contributors and materials flows in the battery life cycle. Finally, the impact of secondary materials from battery recycling due to multi-output allocation procedures is indicated and compared with the impacts from primary routes.

3.1. *Evaluating recovery rates and benefits of the recycling process*

The material recovery rates for using NMC622 as cathode active material for the battery in the modelled recycling process chain are depicted in Fig. 2A–B. These results are compared with the material recovery targets specified in the EU Regulation, which provides two sets of targets: medium-level ambition for 2027 and high-level ambition for 2031 (Fig. 2A). The figure reveals that the modelled recycling process chain meets the 2027 targets for Co, Li, and Cu, but not for Ni. In 2031, the target is potentially only achieved for Li. Higher recovery rates will be required to achieve the 2031 targets for Co, Ni, and Cu.

The breakdown of the carbon footprint is as follows: fossil impacts indicate that NMC production, cell manufacturing (excluding NMC), and the battery pack contribute 31.2 %, 17.3 %, and 45.5 % of the total



Targets in EU Battery Regulation 2023/1542

	Co	Ni	Li	Mn	Cu	Al
2027	90%	90%	50%	N/A*	90%	N/A*
2031	95%	95%	80%	N/A*	95%	N/A*

Impact per kg battery and per kWh battery

Impacts of battery production	Impacts of recycling
15.5 kg CO_2-eq / kg	1.0 kg CO_2-eq / kg
101.9 kg CO_2-eq / kWh	6.5 kg CO_2-eq / kWh

Fig. 2. A-B The results of the evaluation of material-specific recovery rates of the recycling process against the targets provided in the EU Battery Regulation (Fig. 2A), and the climate change impacts of the NMC622 battery pack based on the most important life cycle stages expect usage (Fig. 2B). The material-specific recovery rates are compared with the targets for 2027 and 2031. The materials which require higher recovery rates in future are indicated in red. The impacts of the NMC622 battery pack are indicated as kg CO_2-eq per kg battery pack and per kWh energy storage capacity. The contributions of the materials recovered towards the different components of the battery pack are additionally indicated as stacked bar plots (right). The impacts are also classified as the impacts of battery production and the impacts of recycling. The contribution of individual processes in the respective classifications is provided in Fig. 3A-C. The contribution of the impacts of producing NMC for the different NMC chemistries is provided in Fig. 4A-B. The impacts of recovering secondary materials from the recycling process based on the application of multi-output allocation are provided in Fig. 5A-F. The underlying data of this figure is provided in the supporting material B. The underlying data used in the figures are available in Appendix 4C.

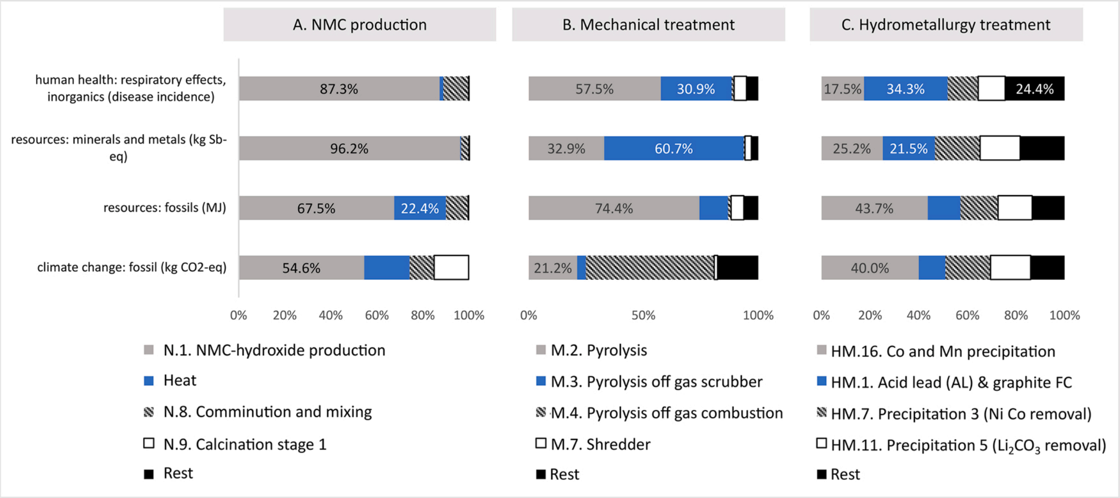


Fig. 3. A-C. The contribution analysis of the unit processes towards the four impact categories considered in the study. The contribution of the top 4 processes in respective gate-to-gate system boundaries (i. e. NMC production, mechanical treatment, and hydrometallurgical treatment) is indicated. The remaining processes are classified as 'Rest'. The unit process names are based on the process flow diagrams indicated in Figs. A1–A3. The underlying data of this figure is provided in the supporting material B. The underlying data used in the figures are available in Appendix 4C.

impacts, respectively (Fig. 2B). The impact of recycling the battery pack was found to be 275.5 kg CO_2-eq, which constitutes 6.0 % of the total impacts of the NMC622 battery pack.

Our study is based on a cut-off EoL modelling approach, i.e., the production of raw materials does have secondary inputs and no crediting is provided. For example, aluminium datasets used in the models have approximately 30 % of secondary inputs from the treatment of aluminium scrap. Therefore, it is important to not give credit for such materials, as this would result in accounting for the benefits of recycling both at the production and its EoL. However, the raw materials for the synthesis of battery electrode materials, such as nickel sulphate, cobalt sulphate, and lithium carbonate, do not have any secondary inputs. The impacts of producing secondary materials need to be determined. To determine them, the impacts of the recycling process must be allocated using multi-output allocation procedures. Results obtained for impacts allocated via both mass and economic value and their comparison with the impacts of primary sources are discussed in Section 3.4.

3.2. Contribution analysis to identify hotspots in the battery life cycle

Fig. 3A–C displays the relative contributions of unit processes to various impact categories in the NMC622 production (Fig. 3A), mechanical treatment (Fig. 3B), and hydrometallurgical treatment (Fig. 3C) gate-to-gate simulations. The results will be limited to discussing the climate change: fossils (kg CO_2-eq) impact.

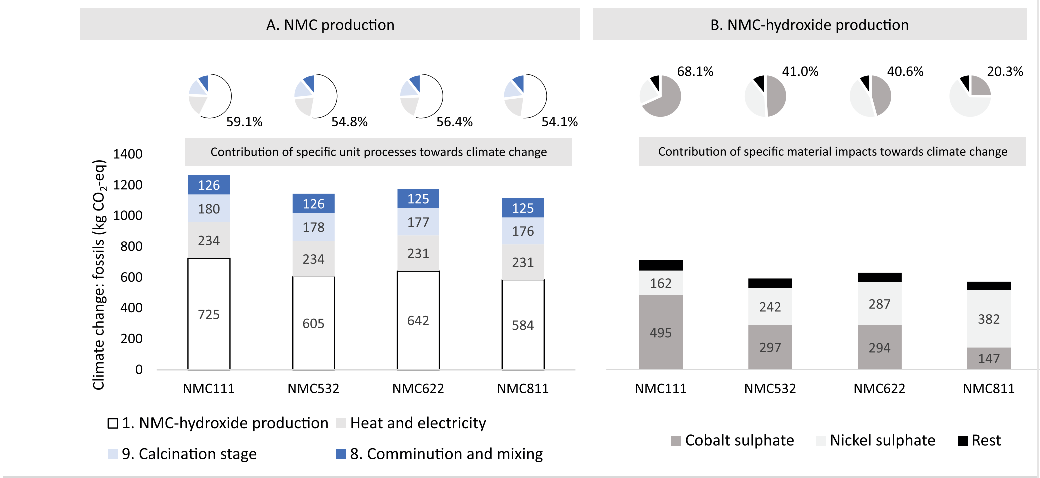
NMC622 production (Fig. 3A) is dominated by the preceding NMC in hydroxide form (hereafter referred to as NMC-hydroxide) production process, affecting all categories. A more detailed analysis of the NMC-hydroxide production process can be found in the following section. The primary driver of the climate change impact of NMC-hydroxide production is material-related impacts, specifically the production of cobalt sulphate (40.6 %) and nickel sulphate (39.6 %). Electricity and heat consumption contribute 68.1 % and 31.9 %, respectively, to these impacts. The impacts of the comminution and mixing unit process are solely due to the addition of Li_2CO_3 to form the Li-NMC hydroxide mix. The impacts of the calcination stage 1 unit process in NMC production are primarily due to direct CO_2 emissions from the calcination off-gases (9.8 kg CO_2 per battery).

The primary contributors to the climate change impact of the mechanical treatment of NMC622 batteries (Fig. 3B) are the pyrolysis, pyrolysis off-gas scrubber, and pyrolysis off-gas combustion processes. The pyrolysis process impact is due to the demand for heat (10.2 %) and electricity (89.9 %). The impacts of the pyrolysis off-gas scrubber process result from the consumption of sorption agents (56.3 %) and electricity (43.6 %). Meanwhile, the pyrolysis off-gas combustion process impacts are primarily due to the direct release of 54.7 kg CO_2 per battery into the biosphere.

The impact of hydrometallurgical treatment of NMC622 batteries is spread across the unit processes shown in Fig. 3C. The Co and Mn precipitation process impact is due to the consumption of sodium hydroxide (44.5 %) and ozone (53.3 %). The acid leach & graphite filter cake process impact is primarily driven by the consumption of hydrogen peroxide (65.3 %) and sulfuric acid (30.3 %). The Precipitation 3 (Ni-Co removal) process impact is due to the consumption of sodium hydroxide (97.2 %). The Precipitation 5 (Li_2CO_3 removal) process impact is dominated by the consumption of sodium hydroxide (97.9 %). The impacts of hydrometallurgical treatment processes are primarily due to the consumption of sodium hydroxide, which is necessary to control the pH and facilitate the precipitation of chemical compounds recovered by hydrometallurgical treatment.

3.3. Analysis of the impacts of different NMC compositions

Fig. 4A illustrates the impact of climate change: fossils with the production of different NMC for various compositions. The contribution of NMC-hydroxide production is presented to highlight the most important processes and substances. Comparing the total impacts, producing NMC for an NMC811 battery results in 12.1 % lower impacts



Total impacts (kg CO_2-eq)	1227.2	1103.8	1138.0	1079.1	725.0	605.0	642.0	584.0
Impacts per kg battery (kg CO_2-eq/kg)	4.4	4.0	4.1	4.1	2.6	2.2	2.3	2.1
Impacts per kWh battery (kg CO_2-eq/kWh)	29.1	26.2	27.0	25.6	17.2	14.3	15.2	13.8

Fig. 4. A-B. Climate change impacts of different NMC compositions. The contributions of impacts are segregated as '1. NMC-hydroxide production', 'Heat and electricity', '9. Calcination stage' and '8. Comminution and mixing.' In addition, the share of the contributions as a pie chart is indicated, especially with the share of '1. NMC-hydroxide production' is indicated as percentages in the pie chart. The contributions towards the impacts of '1. NMC-hydroxide production' are shown on the right. The respective contributions of cobalt sulphate and nickel sulphate according to their primary datasets (Wernet et al., 2016) are provided. In particular, the share of cobalt sulphate is indicated as percentages in the accompanying pie chart. In the table below, the total impacts (as kg CO_2-eq), the impacts per kg battery (as kg CO_2-eq/kg), and the impacts per kWh battery (as kg CO_2-eq/kWh) are provided. The underlying data of this figure is provided in the supporting material B. The underlying data used in the figures are available in Appendix 4C.

than for an NMC111 battery. Meanwhile, the impact of producing NMC622 is 3.1 % higher than NMC532 due to a high share of nickel sulphate and its associated impacts. It is noteworthy that only the impacts of producing NMC-hydroxide show significant variation across the battery chemistries. The contribution of hydroxide production decreases from 59.1 % in NMC 111 to 54.1 % in NMC 811 oxide, warranting further analysis of hydroxide production.

In terms of NMC-hydroxide production, the impact of producing NMC-hydroxide for an NMC 811 battery is 19.4 % lower than for an NMC 111 battery (Fig. 4B). The contribution of the materials to the impacts varies significantly across the battery chemistries. For example, the share of cobalt sulphate towards the impacts of hydroxide production decreases from 68.1 % in NMC 111 to 20.3 % in NMC 811. Thus, it can be concluded that the impacts of cobalt sulphate and nickel sulphate have a significant effect on the CO_2 emissions per kilowatt-hour of energy storage capacity.

3.4. Impacts of secondary materials

Fig. 5A–F describes the outcomes of the multi-output allocation procedures to determine the climate change: fossils impact of producing secondary materials. The results show the application of the mass and economic value-based allocation procedures. The allocated impacts of the secondary materials are compared with the impacts of the primary battery materials used in the models. Six scenarios are considered to analyse the impact of recycling impacts and the prices of battery materials, (1) reference scenario; (2) reduction in recycling impacts by 30 %; and 3–6) increase in the price of battery active materials by a factor of

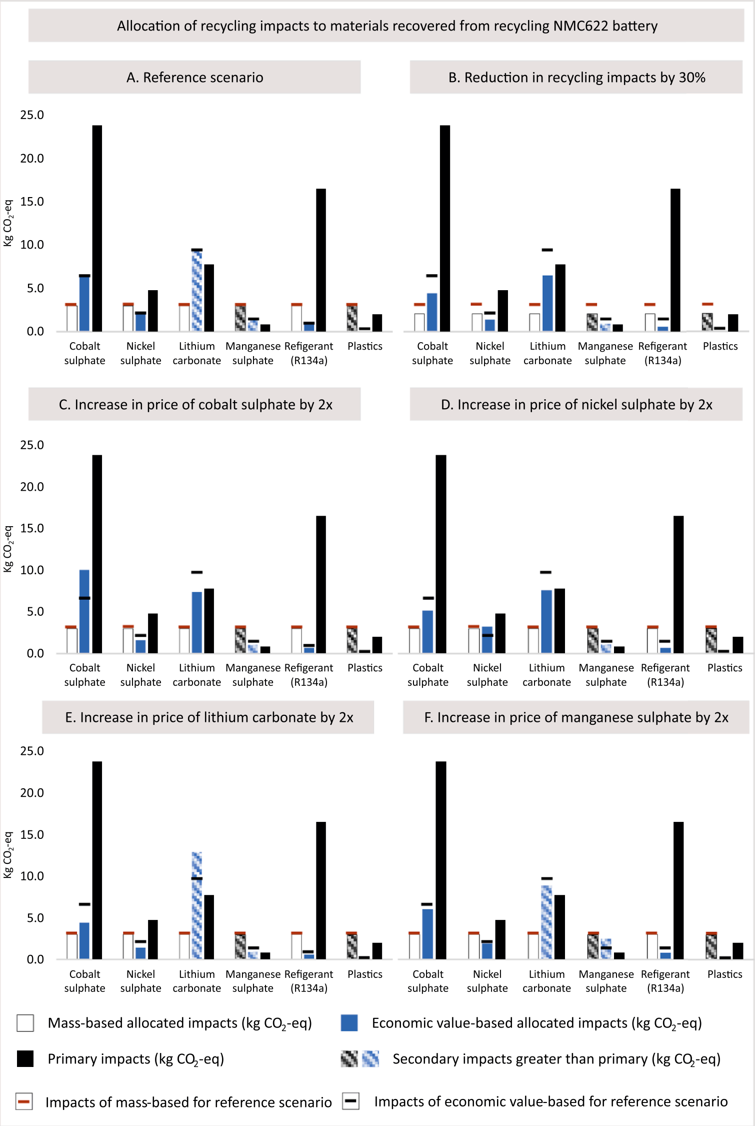


Fig. 5. A-F Impact of the secondary materials recovered from the recycling of the NMC622 battery pack on the equivalent amount of CO_2 produced. The impacts of recycling are allocated to the secondary materials based on mass and economic value allocation procedures. Three scenarios in addition to the reference scenario are considered to analyse the influence of reduction in recycling impacts and increase in the price of battery materials. In the first scenario, the impacts of recycling are directly reduced by 30 %, and the consequence of this reduction in the impacts of the secondary materials is indicated. Similarly, for demonstration purposes, an increase in the price of cobalt sulphate and nickel sulphate by a factor of 2 is considered. In all scenarios, the impacts of mass-based and economic value-based allocation are provided. The impacts due to the mass-based allocation are indicated as dotted lines. These values are compared with the impacts of primary production. In cases where the values are higher than the primary production, the bars are pattern-filled. The values of the economic value-based impacts of cobalt sulphate, nickel sulphate, and lithium carbonate are indicated. The underlying data of this figure is provided in the supporting material B. The underlying data used in the figures are available in Appendix 4C.

two. The reference scenario indicates the recycling route considered in the study as described in Appendix 1 and 2, whereas the price of battery metals is based on average market prices (indicated in Appendix 4c).

The results indicate, irrespective of the scenarios, that the secondary impacts of cobalt sulphate, nickel sulphate, and refrigerants (R134a) are lower than their primary production. The secondary impacts of lithium carbonate are also lower in all scenarios except in the reference. It is worth noting that, despite the secondary impacts of lithium carbonate being higher than its primary impacts (in the reference scenario), it is important to interpret the results in the context of the battery materials needed to produce the battery pack. For the considered NMC622 battery pack of 42.2 kWh in the study, the mass of lithium carbonate, cobalt sulphate, and nickel sulphate are 16.1 kg, 22.5 kg, and 63.1 kg respectively. The differences between the secondary and primary impacts in nickel sulphate will have a more pronounced effect toward reducing the climate change impacts compared to lithium carbonate. Furthermore, the secondary impacts of manganese sulphate and plastics (allocated based on mass) are higher than the primary impacts considered. Finally, the impacts of plastics, allocated economic value-based is lower than the primary in all scenarios.

In the reference scenario (Fig. 5A), it is evident that for mass-based allocation, all secondary materials, except manganese sulphate and plastics have a lower impact than their primary routes. Applying the economic value-based allocation all secondary materials except lithium carbonate and manganese sulphate have a lower impact than their primary routes. However, there is a large difference in the impacts of the primary battery material production, especially in nickel sulphate, cobalt sulphate, and graphite (Ali et al., 2023c; Engels et al., 2022). It is also worth noting that the impacts of secondary materials due to mass-based allocation remain the same, for example, 3 kg CO_2-eq in the case of the reference scenario. However, the impacts vary significantly due to the application of economic value-based allocation.

A target value of the reduction in recycling impacts by 30 % (Fig. 5B) subsequently reduces the impacts of secondary materials by 30 %, irrespective of the allocation procedure. Such targets can indicate the potential necessary improvements needed in battery recycling technologies, such that the impacts of secondary materials are lower than their primary production.

The subsequent scenarios concern the increases in the cost of the battery active materials (Fig. 5C–F). As indicated in Fig. 5C, a two-fold increase in the price of cobalt sulphate has increased the impacts of secondary cobalt sulphate in comparison to the reference scenario by 58.7 %. Whereas the secondary impacts of nickel sulphate and lithium carbonate have reduced by 20.0 % and 21.5 % respectively.

In Fig. 5D, the scenario concerning the two-fold price increase of nickel sulphate, the secondary impacts of nickel sulphate have increased by 60 %. However, the secondary impacts of cobalt sulphate and lithium carbonate have reduced by 19.0 % and 18.3 % respectively. Subsequently, in Fig. 5E, the secondary impacts of lithium carbonate have increased by 38.7 %, and the impacts of cobalt sulphate and nickel sulphate have reduced by 30.2 % and 30.0 % respectively.

Finally, the increase in the price of manganese sulphate, Fig. 5F, resulted in an increase in the secondary impacts of manganese sulphate by 92.3 %. However, the secondary impacts of cobalt sulphate, nickel sulphate, and lithium carbonate have only reduced by 4.8 %, 5.0 %, and 5.4 % respectively. This indicates the price of manganese sulphate has a high contribution to its secondary impacts and a marginally low contribution towards the secondary impacts of other battery materials.

4. Discussion

In this section, a discussion is provided on the advantages and challenges of using a simulation-based LCA approach, followed by the role of multi-output allocation procedures in determining the impacts of secondary materials and reflecting on the role of simulation-based LCA in the design of circular battery supply chains.

4.1. Advantages and challenges in simulation-based LCA

Process simulation is a tool commonly used to reliably calculate the balanced mass and energy flows needed to design and optimize industrial plants. These industrial plants are part of actual product life cycles and therefore, it makes sense to leverage the capabilities of process simulation to also generate the life cycle inventory data needed to perform LCA. Compared to mainstream methods, it does so more rigorously and at higher levels of detail because it is based on fundamental physics principles. The fact that the design of new plants is implicitly prospective, also implies that the simulation approach is particularly suited to prospective assessments in which the availability of historical process data is limited—the fundamental basis of simulation models also gives them predictive capabilities.

Despite the above-mentioned advantages, several challenges exist while taking a simulation-based LCA approach. The primary challenges tend to be complexity and model limitations. Developing a simulation model requires detailed knowledge and an understanding of the processes involved. The complexity of the model can make it difficult for some stakeholders to understand the results. Process simulations are also limited by the information available and may not accurately reflect existing processes. To improve accuracy and reliability, and to confirm the validity of assumptions, validation with experimental or industrial data is essential.

4.2. Role of multi-output allocation on the impact of secondary materials

The results indicate the influence of the impacts of the recycling process and the price of battery materials on the secondary impacts of the materials recovered. The scenarios considered in the study demonstrate the critical aspects determining the impacts of the secondary battery materials. For example, as seen in Fig. 5A, the approach considered in the study can be used to set targets for recycling processes that aim to produce secondary materials with impacts lower than their primary. In the example, the 30 % reduction in the impacts of the recycling process was sufficient to reduce the secondary impacts of lithium carbonate.

The results also clearly indicated the secondary impacts of manganese sulphate are always higher than their primary production irrespective of the scenario considered. This finding indicates that using secondary manganese sulphate in battery production does not provide any additional advantages in terms of climate change. However, there are recycling processes that aim to co-precipitate NMC in a single step rather than as separate sulphates. In such cases, the economic value of the respective NMC should be considered before determining the potential reduction in climate change impacts.

It is worth noting that the increase in the price of lithium carbonate has the highest influence on the secondary impacts of cobalt sulphate and nickel sulphate (Fig. 5E). This increase is since the total economic value of the lithium carbonate is the highest amongst all the battery active materials recovered. The total economic value is the product of the mass of material recovered and its price per kg. Despite the mass of nickel sulphate recovered, at 31.3 kg, being the highest, the total economic value, due to the high price of lithium carbonate (at $40 per kg lithium carbonate) results in lithium carbonate having the highest total economic value. This relation demonstrates the importance of combining both the mass of material recovered and the economic value before attributing the impacts of recycling to the secondary materials.

In addition to secondary impacts, the climate change impacts of the different battery chemistries are indicated in Fig. 4A-B. This result demonstrates the relationship due to the changing molar ratios of the battery metals and distinguishes this from the contribution of production related impacts. The climate change impacts due to the production of the NMC do not vary across chemistries, however, noticeable differences are only due to the contribution of the battery metals.

4.3. Role of simulation-based LCA in the design of circular battery supply chains

Simulation-based LCA can help in the design of circular battery supply chains by providing a comprehensive evaluation of the environmental impacts of different supply chain configurations. This type of analysis can consider various factors such as the materials used, the production processes involved, the transportation of materials and products, and the EoL treatment of batteries. By considering all of these factors, simulation-based LCA can help to identify the most environmentally sustainable supply chain configurations and design options, taking into account the trade-offs between environmental performance, economic viability, and technical feasibility. However, this would necessitate a comprehensive techno-economic assessment.

Moreover, simulation-based LCA can help to optimize the design of circular battery supply chains by exploring different scenarios and assessing the impact of changes in materials used and their sourcing, technologies, production processes, EoL management, and other factors. This approach allows designers and decision-makers to consider the full range of trade-offs and make informed decisions about the best course of action. For instance, a simulation-based LCA can help to identify opportunities for reducing emissions, improving energy efficiency, or increasing the use of recycled materials in the supply chain from early in the design process, so enabling design-for-recycling.

4.4. Limitations of the study

The simulation models must be confirmed with experimental or industrial data. Despite the pending validation, the existing models are based on validated findings from literature for similar processes (Wang & Friedrich, 2015). Further assumptions are based on standard theoretical tools, such as using Pourbaix (Eh-pH) diagrams to confirm the thermodynamically stable species present in aqueous solutions at different values of pH. Results validated using our experimental work will be the subject of a future publication.

As with most LCI data collection, the development of process simulation models is a time-consuming task. It requires an in-depth understanding of the chemical and metallurgical processes being modelled to ensure that the resulting inventory data are based on the actual physics governing the process, rather than aggregated data sets.

4.5. Outlook

The EU Battery Regulation proposes carbon footprint thresholds for the batteries placed in the market. These thresholds will be based on the relative distribution of the carbon footprint values of the batteries on the market. However, such thresholds must be re-examined based on principles of absolute sustainability (Ali & Ryberg, 2023). Absolute sustainability assessments aim to measure and compare impacts against predetermined planetary thresholds, while relative assessments such as LCAs compare impacts between alternatives without a predefined sustainability threshold (Steffen et al., 2015).

It is also necessary to ensure such thresholds limit impacts of mobility within the planetary boundaries. This will ensure that the batteries produced from upcoming giga factories do not place an additional burden on the planetary boundaries. However, there is a general lack of methodology that can fairly allocate the planetary boundaries to giga-scale battery production. Incorporating the principles of absolute sustainability in the planning of battery giga factories will aid in the successful development of technology roadmaps for electric vehicles (Ali et al., 2023b).

5. Conclusion

In this study, the material-specific recovery rates for the recycling of NMC622 battery packs were compared with the targets established in the EU Battery Regulation. The results show that the modelled recycling process met the targets for three out of the four materials outlined in the regulation in 2027. However, the results indicate the need to achieve even higher recovery rates to meet the high-level ambition targets set for 2031. The breakdown of climate change: fossil impact of the NMC622 battery pack indicated that the NMC production, cell manufacturing (excluding NMC), and the battery pack contribute 31.2 %, 17.3 %, and 45.5 % of the total impacts respectively. The impact of recycling the battery pack was found to be 6.0 % of the total impact of climate change impact.

Multi-output allocation procedures were used to determine the climate change impact of producing secondary materials and compared it to the impacts of the primary battery materials. In reference scenario, applying economic value-based allocation resulted in cobalt sulphate and nickel sulphate having 73.5 % and 57.4 % lower carbon footprint than their primary, however, lithium carbonate had a 20.8 % higher footprint. The results also indicate that the price of lithium carbonate had the highest influence on the secondary impacts of cobalt sulphate and nickel sulphate.

Overall, the results of this study highlight the importance of efficient recycling processes for reducing the environmental impact of battery production. The need for higher material recovery rates at purities suitable for battery production, especially for nickel, to achieve the high-level ambition targets by 2031 was also emphasized. The results of this study can serve as a useful reference for future studies and decision-makers in the field of battery production and recycling. Additionally, the results can serve as a guide for policymakers and stakeholders in the battery industry to prioritize their efforts in reducing the environmental impact of battery production and recycling. The process simulation-based approach demonstrated in this study advances the field of prospective LCA of emerging battery recycling technologies. This study has provided meaningful data and insights needed for a comprehensive assessment of emerging technologies.

CRediT authorship contribution statement

Abdur-Rahman Ali: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing – original draft. **Neill Bartie:** Data curation, Methodology, Software, Writing – original draft. **Jana Husmann:** Writing – original draft, Investigation. **Felipe Cerdas:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Daniel Schröder:** Writing – review & editing, Project administration, Funding acquisition. **Christoph Herrmann:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

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

Data availability

The necessary data for the environmental impact assessment and the graphs are provided in the SI.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2023.107384.

Appendix 4A-C. Supplementary material

Appendix 4A: An Excel sheet which provides the life cycle inventory for the simulation models.
Appendix 4B: An Excel sheet which provides the Brightway2 models used in the estimation of the life cycle impact assessment scores.
Appendix 4C: An Excel sheet with provides the underlying data used in the figures.

Appendix 1. Mechanical treatment of EoL batteries

This section describes the process simulation of the mechanical treatment of EoL batteries. The process flow diagram of the mechanical treatment is depicted in Fig. A1. First, the EoL batteries are discharged and dismantled (*M.1. Dismantling)*. This step removes module-level cabling and electronic components, the refrigerant, and aluminium, steel and plastic casing components (*Streams 2–6*). After this, the intact cell packs are transferred to the pyrolysis process (*M.2. Pyrolysis*). Pyrolysis is assumed to be conducted in the absence of oxygen at 740 °C (Kaminsky, 1993; Huang et al., 2018). During pyrolysis, electrolytes are evaporated and polymer materials, e.g. polypropylene (PP), polyethene (PE), polyethene terephthalate (PET), membranes, and binders are converted into gaseous, oil, and solid phases consisting of various organic compound combinations (Kaminsky, 1993; Yoshioka et al., 2004).

The solid product from the pyrolysis process contains the residue from polymer conversion, as well as Al foils from cell casings and the cathode, Cu foil and graphite from the anode, and the Li-NMC active material. This material mixture is shredded (*M.7. Shredder*) under an inert nitrogen atmosphere, after which a screen (*M.8. Screen*) is used to separate the shredded mixture into coarse and fine fractions. The course fraction primarily consists of the Al and Cu foils, but also any other larger particles not removed in the previous steps.

Although previous studies have shown that pyrolysis largely separates foils and active materials (Schwich et al., 2021), the possibility of active material and graphite carry-over to the coarse fraction via foils (and vice versa) is always present. Adopting a conservative approach, it is assumed that a small amount of the fine fraction is transferred to the coarse fraction and that a small amount of Al and Cu foils exits the screen via the fine fraction. Air separation (*M.9. Zig zag*) is employed to further classify the coarse fraction into light and heavy fractions, the latter containing the recovered foils and any other heavier materials, e.g., steel particles if present in EoL cell packs. To separate these in preparation for further external recycling, a magnetic separation (*M.10. Magnetic separator*) step removes any steel particles from the Al/Cu foil mixture, followed by electrostatic separation (*M.11. Electrostatic separator)* of the Al and Cu foils.

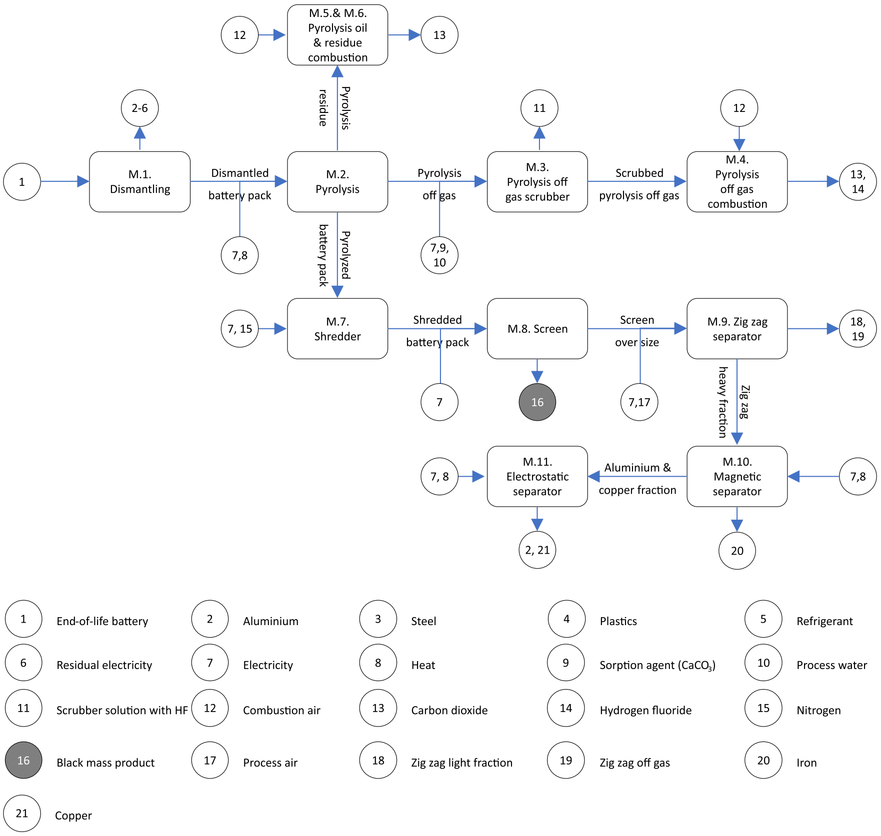

Fig. A1. The process flow diagram of the mechanical treatment process of used LIB. The main product of this process is the Black mass (Stream 16). The black mass product is subsequently transferred to the hydrometallurgical treatment process, as indicated in Fig. A2.

Appendix 2. Hydrometallurgical treatment of black mass

The process flow diagram for the hydrometallurgical treatment process is depicted in Fig. A2. The fine fraction from the screen (unit M.8 in Fig. A1), the so-called ‘black mass’, is transferred to the hydrometallurgical recycling process, which is largely based on the process proposed by Wang and Friedrich (2015) for the recovery of graphite, Cu powder, Li_2CO_3, and cathode metal salts. The authors conducted several leaching and selective precipitation experiments to optimize the designed process to maximize recoveries. In the first step, the black mass is leached in sulphuric acid. Except for graphite, most active material compounds in the pyrolysis residue dissolve in this step. Graphite and any undissolved species are removed from the aqueous solution in the following filtration step for further treatment. Next, Cu is removed from the solution using cementation with Fe, during which Fe enters the aqueous solution and elemental Cu precipitates for removal in the filtration step that follows. Al, Fe, Co, Ni, Mn, and Li are then removed from the solution using selective precipitation through stepwise increases in pH achieved via the addition of sodium hydroxide (NaOH). Al and Fe hydroxides ($Al(OH)_3$ and FeO•OH, respectively) are precipitated first (at pH ~2.5) followed by Ni as $Ni(OH)_2$ and Co as $Co(OH)_2$ at pH ~8, and Mn as $Mn(OH)_2$ at pH ~10, so removing these metals from solution. In the final precipitation step, Li is precipitated as lithium carbonate (Li_2CO_3), which is achieved through the addition of sodium carbonate (Na_2CO_3). Process simulation software (HSC Chemistry, Metso Corporation) is used to develop and validate the simulation model based on the process description and results, respectively, reported by Wang and Friedrich (2015).

As described above, Ni and Co are initially precipitated as a mixture of their hydroxides. In the selected recycling process, Ni, Co, and Mn are recycled in separate closed loops so that the chemistry of newly produced NMC active material can be selected freely in our simulations. Therefore, Ni and Co need to be separated and purified in preparation for their return to the active material production process. Furthermore, filter cakes produced after each of the selective precipitation steps are impure due to, among others, incomplete dissolution and precipitation, co-precipitation of undesired compounds, inefficient filtering, and cross-contamination of the precipitates and filtrates across filter media. For this paper, Co and Ni are separated using oxidative selective precipitation to remove Co and a small amount of carried-over Mn to produce a Ni-rich solution. The simulation model described above was expanded and validated based on the process design and experimental results reported by Ichlas et al. (2020). Ni is then precipitated in hydroxide form using NaOH. For the purification of the Mn hydroxide filter cake, a simple dissolution and reprecipitation process is assumed. In the simulation model, the recovered $Ni(OH)_2$, $Co(OH)_2$, and $Mn(OH)_2$ are recycled to the NMC in hydroxide form, where they are assumed to displace stoichiometric amounts of virgin Ni, Co, and Mn, respectively. Similarly, recovered Li_2CO_3 is recycled to the NMC production step. It should be noted that 1:1 displacement of virgin materials would only be possible in a completely integrated supply chain, and when demand for recycled materials exists, which is assumed in this paper.

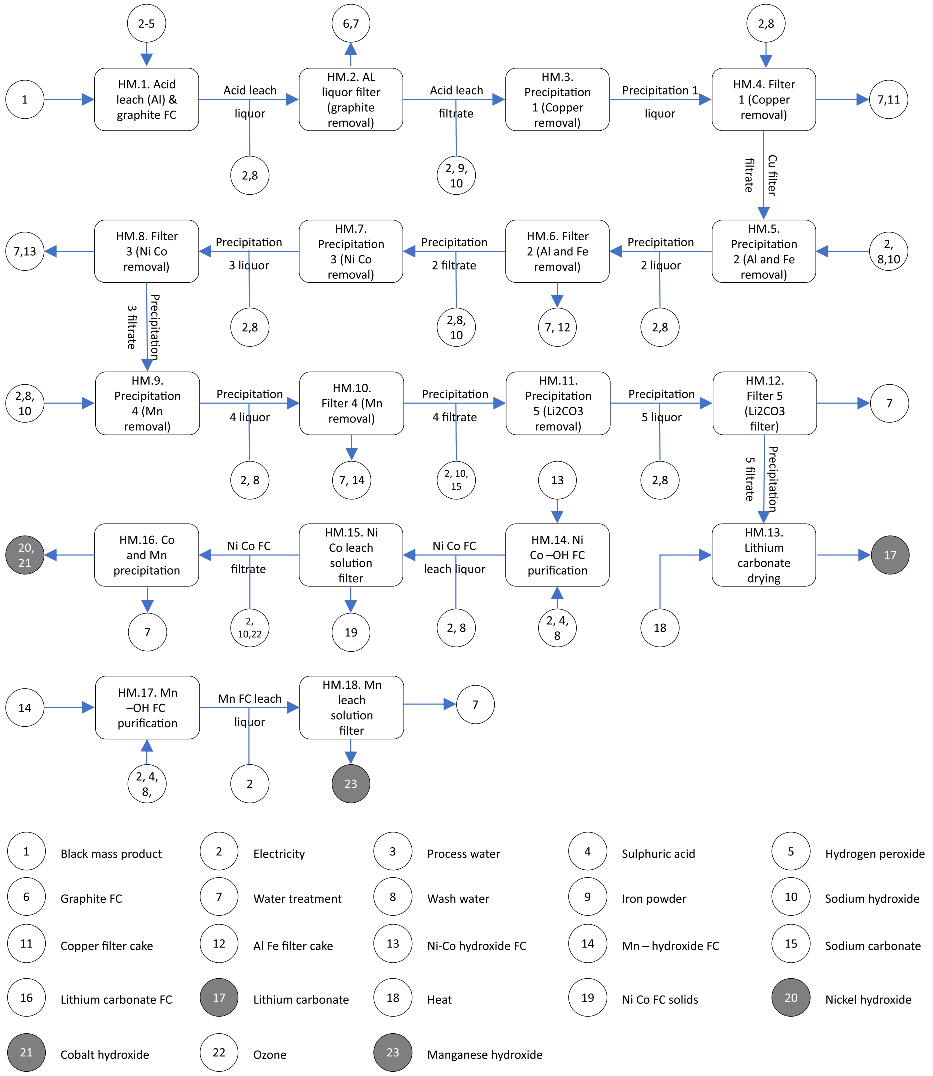

Fig. A2. The process flow diagram of the hydrometallurgical treatment process. The input to this process is black mass production (from Fig. A1). The hydrometallurgical treatment of the black mass results in the recovery of Ni, Mn, and Co in hydroxide form and lithium as lithium carbonate. The metal hydroxides are subsequently purified in the upgrade steps. The purified hydroxides are subsequently returned to the NMC active material production step to be re-used in cell production (as indicated in Fig. A3).

Appendix 3. Production of NMC

The simulated NMC active material production process is largely based on the descriptions and parameters provided by Heimes et al. (2019). The first step in this process, as indicated in Fig. A3, is the synthesis of the NMC-hydroxide [$Ni_xMn_yCo_{1-x-y}(OH)_2$] active material precursor where the three metals enter the process in sulphate form. To facilitate closed-loop recycling, the Ni, Co, and Mn hydroxides recovered during recycling re-enter the process here. In the next step, the NMC-hydroxide is converted to the NMC-oxide active material ($LiNi_xMn_yCo_{1-x-y}O_2$) through the addition of Li (as Li_2CO_3) and calcination. Recovered Li_2CO_3 re-enters the production process here. It is assumed that the purities achieved in the recycling process meet the specifications for NMC cathode active material production. The simulated production process includes further steps to recover ammonia from the NMC-hydroxide production step (Kim et al., 2021). The ammonium sulphate [$(NH_4)_2SO_4$] product can be used as a fertilizer.

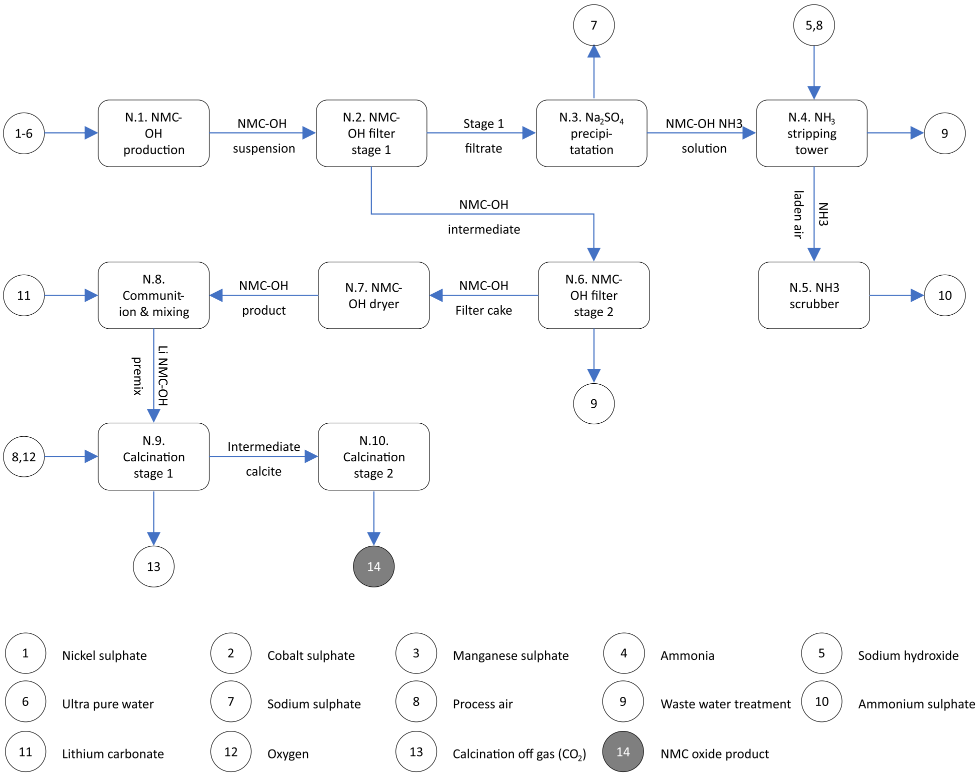

Fig. A3. The process flow diagram to produce nickel-manganese-cobalt (NMC) oxide. The materials recovered from hydrometallurgical treatment (from Figure A2) are used in closed-loop recycling (Streams 1,2,3, and 11). The main product of this process is NMC, which is used in the production of cathodes, as indicated in Fig. 1.

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