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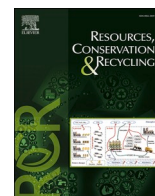
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Full length article

Life cycle assessment of LTO-rich anode waste from lithium-ion battery with a hazardous waste management approach

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ABSTRACT

Lithium titanate oxide (LTO) batteries have been under intensive research due to their stability, safety, and rapid charging characteristics. Nevertheless, uncertainties as to LTO-batteries behavior when used as a raw material in battery recycling still exist. This study provides a grave-to-gate life cycle inventory for a hydrometallurgical battery recycling process in which Li-battery waste materials nickel manganese cobalt (NMC), LTO, and graphite were used as feed. The simulation showed that NMC cathode materials and lithium from both battery waste fractions could be recovered. In contrast, the titanium present within LTO cannot be recovered by the recycling process. Nevertheless, the life cycle assessment (LCA) of the process demonstrated clear benefits of recycling battery materials, highlighted by the decrease in global warming potential, acidification, eutrophication, and ozone depletion potential. Additionally, two routes for hazardous waste management were simulated to ascertain the environmental impacts of hazardous waste management within the recycling process.

1. Introduction

Lithium-ion batteries (LiBs) are becoming increasingly important in mitigating climate change, particularly for the decarbonization of transport. Additionally, European Union regulations adopted in 2021 have set ambitious goals that require the decarbonization of at least 55 % of the energy that currently generates greenhouse emissions by 2030 (European Commission, 2021). This will require substantial increases in battery-based energy storage infrastructures, including secondary LiBs and redox flow cells, or supercapacitor technologies to ensure grid energy storage and load balancing (Schubert et al., 2023). Nevertheless, the primary materials currently required to produce LiBs include several elements that have been classified as critical or strategic raw materials (European Commission, 2023) such as cobalt, lithium, manganese, and graphite. Consequently, significant research has been dedicated to finding alternative battery electrode chemistries that either require reduced levels of critical raw materials (CRM) e.g., low/cobalt-free (Luo et al., 2022), or novel technologies that avoid the use of lithium, like sodium-ion (Kumaresan et al., 2021). For anodes in particular, several alternative CRM-free options have been studied that include biomass-derived hard carbon instead of natural graphite (Thompson et al., 2021b). Additionally, lithium titanate oxide (LTO) and silicon have also been developed as anode materials that can supplement or be

used as an alternative to graphite (Zhang et al., 2022).

In contrast to other battery types like lithium-ion phosphate (LFP), lithium-ion nickel-manganese-cobalt (NMC) and lithium manganese oxide (LMO) that typically use a combination of copper and graphite for the anode, lithium titanate (LTO) batteries utilize an alternative: $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (Yang et al., 2022). These types of LTO anodes - when combined with lithium transition metal oxide cathodes - may provide better stability, are safe to operate at lower temperatures, allow rapid charging, and generally have a long cycle life of approximately 20,000 cycles (Zhang et al., 2022). On the other hand, LTO batteries have lower energy densities when compared to those with graphite anodes. Therefore, have typically found application in uninterruptable power systems, solar-powered lighting, and as auxiliary batteries for electrical powertrains. Moreover, LTO as anode material is being increasingly used for the further electrification of buses and trains that have only short distances between rapid charging stations as well as part of car EV battery modules (e.g., in tandem with traditional NMC cells) to provide smoother acceleration (Zhang et al., 2022). As a result, it is predicted that the use of LTO as an anode material will grow at the highest compound annual growth rate (CAGR) by 2030 in comparison with the other currently available battery chemistries (Research and Markets, 2023).

Various life cycle assessments (LCA) have demonstrated that the greenhouse emissions, oil consumption, and air pollution related to

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electric vehicles (EVs) are considerably lower than internal combustion engine (ICE) vehicles during the operation/in-service stage (Girardi et al., 2015). In addition, there are also several studies that have attempted to predict the environmental impact attributable to different battery cells throughout various points of their lifetime including Cradle-to-Gate (production), Well-to-Wheel (use) and Grave-to-Cradle (recycling) (Yang et al., 2022; Yin et al., 2019; Rinne et al., 2021). Nevertheless, to have a better understanding of the environmental impacts of different battery cells, it is necessary to assess every stage of their life in detail - including industrially relevant recycling approaches - since often the study of a system is focused solely on battery use and/or manufacture (Peters, 2023). Furthermore, there is also evidence that recycled black mass from automotive batteries results in economic and environmental advantages when circulated effectively (Thompson et al., 2021). Despite recent investigations into the grave-to-gate stage, where downcycled anode material like graphite, has been explored as an absorbent for CO₂ capture (Lee et al., 2023), raw material for catalyst (Liivand et al., 2023) or to synthesize materials such as graphene oxide (Tian et al., 2024), industrial battery recycling processes primarily focus on the recycling of the economically attractive cathode materials to recover the most valuable elements like nickel and cobalt (Van Hoof et al., 2023). In such cases, leach residue—primarily composed of graphite—is either used as an energy source or a reducing agent in pyrometallurgical processes (Klemettinen et al., 2024; Van Hoof et al., 2023; Wang et al., 2021) or it ends up in waste streams (Regional State Administrative Agency for Southern Finland, 2019).

Previous research in the literature suggest that anode materials can have a marked influence on the results of the environmental impact assessments, particularly during the manufacturing stage, as generally anode - and especially LTO - production is material and energy intensive (Yang et al., 2022; Wu and Kong, 2018). However, only a few studies have focused on analyzing the environmental impacts of mining, manufacturing, and use related to LTO (Yin et al., 2019; S. Zhang et al., 2022). Similarly, there is currently only limited research related to the recycling of LTO containing batteries, for example, the work by Koh et al. (2021), in which the approach for addressing the data gaps related to life cycle inventory (LCI) of LTO recycling is to utilize the Ecoinvent database for the hydrometallurgical treatment of spent LIBs. However, this database currently only provides information related to the cathode recovery, rather than a complete determination of the environmental impacts that includes the anode materials. In contrast, Chigada et al. (2021), and Yin et al. (2019), have addressed the recycling of LTO based on a pure feed material, with the focus on a battery with an LTO anode and a NMC cathode. Nonetheless, the scenarios utilized were not representative of the expected industrial-scale recycling facilities, where graphite is likely to be encountered within the mix of feed material or black mass.

It has been predicted by Weber et al. (2018), that LTO containing batteries - with more common cathode chemistries in their structure like NMC - will end up in the state-of-the-art recycling processes that are currently tailored to dealing with graphite-based LIB chemistries. This may cause challenges either in processing or potentially lead to the loss of lithium titanate into the process residues along with graphite. Consequently, as outlined in this paper, it is imperative that the behavior of LTO containing batteries within recycling processes also be investigated, to evaluate their recyclability in emerging metallurgical recycling processes. This may provide the potential to enhance the design of effective recycling routes that minimize associated losses and allow for anode materials recovery.

2. Materials and methods

The life cycle inventory (LCI) was obtained using the Sim module within HSC 10.1 software by Metso (2023), the parameters for the process simulation and chemical reactions for the unit operations were obtained from literature and available metallurgical information. LCA

results were obtained using OpenLCA software (GreenDelta, 2023) and the Ecoinvent 3.10 database - provided by the Swiss Centre for Life Cycle Inventories (Ecoinvent Database, 2020) - for background data.

2.1. Raw materials

The raw material used as the input feed for the process model was spent NMC811-graphite-based LIBs combined with spent NMC111 and lithium titanate oxide-based LIBs. These waste fractions were fed in equal amounts (1:1 ratio), and the composition of the mixture was calculated based on the values of Jegan-Roy et al. (2021). The material in the referred study was pre-treated via crushing and sieving to obtain a particle size of ~ 100 µm and this was further treated by drying at 60 °C to remove the electrolyte and binder.

The oxides investigated within the current study include cathode materials—lithium cobalt oxide (LiCoO₂), lithium nickel oxide (LiNiO₂), lithium manganese oxide (LiMnO₂)—as well as an anode material—lithium titanate oxide (Li₄Ti₅O₂). In addition, Al and Cu (from the current collectors) as well as Fe originating from casings are included and are considered to be in their metallic form. For simplicity, the presence of electrolyte and binder was excluded as increasingly, industrially produced black mass is pre-treated to remove such components to provide a “cleaner” matrix (Brückner et al., 2020; Latini et al., 2022). The remaining material was assumed to comprise of graphite that originates from the state-of-the-art NMC811 battery anode material. The calculated composition of the feed used in the process simulations is outlined in Table 1.

2.2. Process description

The study simulated one recycling process scenario using the SIM module, and two alternative hazardous waste management routes were evaluated using OpenLCA, as shown in Fig. 1. More detailed information about the hydrometallurgical battery recycling processes investigated is provided in supporting information (section S1).

After leaching, the resulting leach residue was determined to comprise of unreacted black mass, graphite, and titanium oxide. Consequently, this leaching residue composition was categorized as hazardous waste due to its pH, and the hazardous properties of the traces of lithium metal oxides present in the leach residue -due to incomplete dissolution during the leaching process- (Sigma-Aldrich, 2023a). Finally, two potential scenarios to deal with the hazardous waste outputs were assessed and can be outlined as follows:

SC1: 80 % of the waste generated from the leach residue is treated as hazardous waste with incineration on-site with no energy recovery. Model was obtained based on information from the Ecoinvent 3.10 database.

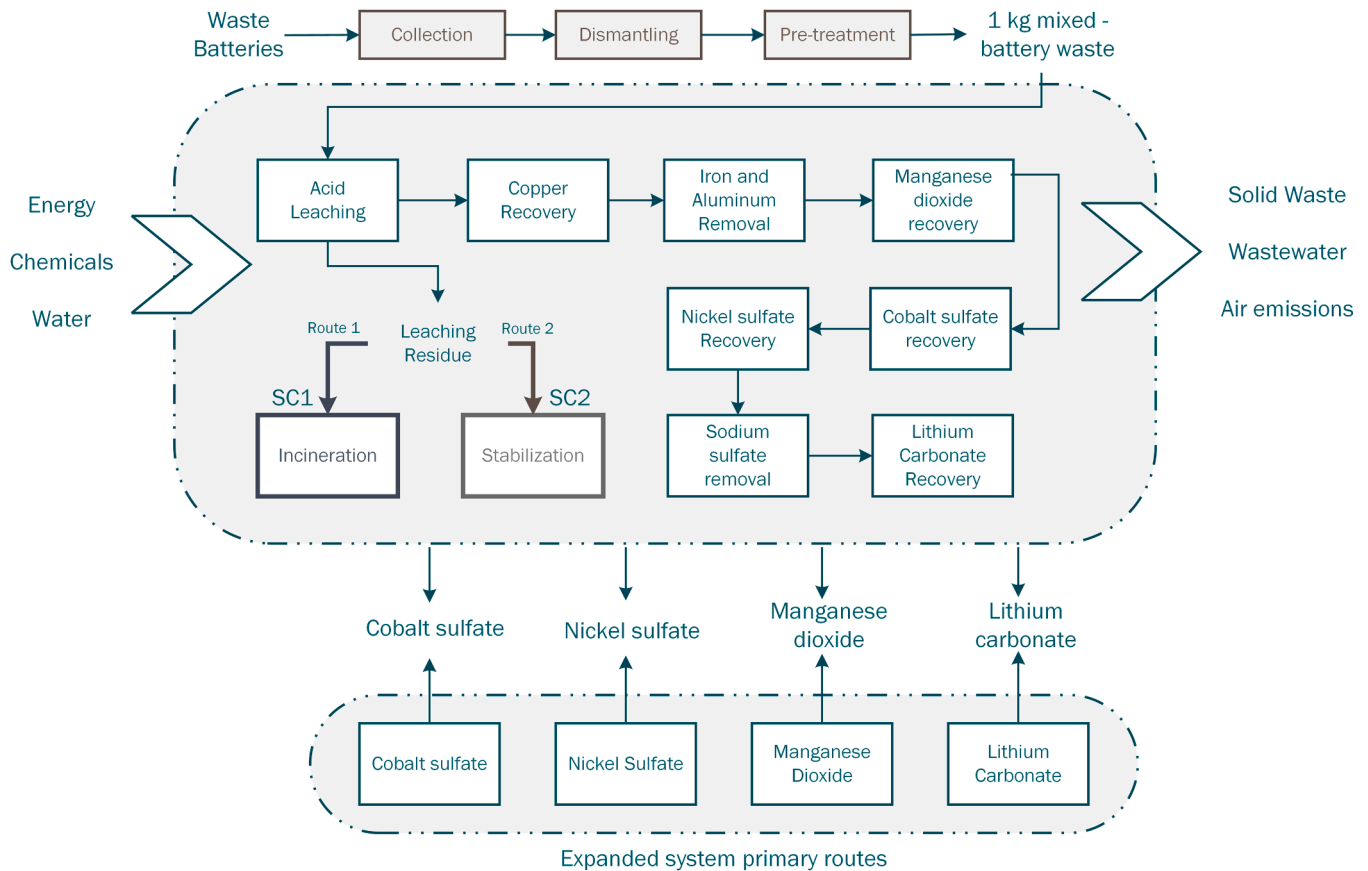
SC2: 80 % of the waste generated from the leach residue is treated as hazardous waste and is stabilized followed by on-site storage at an underground repository. Model was obtained based on information from the Ecoinvent 3.10 database.

It was assumed that 20 % of the leach residue could be directed to other uses such as research or as a reductant - in a manner similar to the Umicore process (Velázquez-Martínez et al., 2019). The pregnant leaching solution (PLS) was further treated in a hydrometallurgical battery recycling circuit to recover copper as metallic copper powder through cementation, whereas manganese was recovered as MnO₂ through solvent extraction (SX) and oxidative precipitation assisted by O₂/SO₂ gas (6 vol-%). Cobalt and nickel were recovered by SX and crystallization as nickel sulfate (NiSO₄•6H₂O), and cobalt sulfate (CoSO₄•7H₂O) respectively. The PLS was treated to increase the lithium concentration by Glauber salt (Na₂SO₄•10H₂O) crystallization, and the resultant solution was treated with Na₂CO₃ to precipitate the Li as lithium carbonate (Li₂CO₃). Any Al and Fe present were also removed as

Table 1

The elemental composition of the battery waste (NMC811-graphite and NMC111-LTO) used as the feed in recycling flowsheet.

Component	LiNiO ₂	LiMnO ₂	LiCoO ₂	Li ₄ Ti ₅ O ₁₂	Cu	Al	Fe	Graphite
wt%	30.71	11.33	11.81	20.86	2.82	0.62	0.97	20.85

**Fig. 1.** System boundary flowsheet with possible scenarios highlighted and system expansion.

hydroxides through the application of hydrolysis. In addition, the effluents from every unit operation within the flowsheet were combined and directed towards a neutralization step in which it was assumed that most of the remaining metallic impurities in solution would be removed at pH 7.

The feed to the system was 7000 metric tonnes (mt) of black mass per year comprised of equal parts of NMC811-graphite and NMC111-LTO, with the total amount taken referenced to the Umicore process (Velázquez-Martínez et al., 2019). Based on this input feed, approximately 1700 mt per year of lithium carbonate, 972 mt per year of manganese dioxide, 5950 mt per year of nickel sulfate, and 2250 mt per year of cobalt oxide are generated by the process. The energy demand of the most important unit operations was calculated, resulting on a total energy consumption of 1.33 MJ per kg of black mass recycled. The simulated recovery rates of the valuable battery elements were 88 % Li, 96 % Co, 93 % Ni and 93 % Mn, which indicates that Co, Ni and Mn have high recovery rates using the methods proposed in the recycling flowsheet. The lower level of lithium recovery results from losses that occur during the sodium carbonate removal stage and is expected as Lithium is recovered within the last unit process of the flow sheet, therefore there is an accumulation of minor losses in several previous unit operations like filters. Detailed information about the hydrometallurgy processes, parameters for the flowsheet simulations and energy calculations can be found in the associated supplementary information (S1).

2.3. Goal & scope

The objective of the assessment was to generate a Life Cycle Inventory (LCI) and Life Cycle Impact Analysis (LCIA) to determine the environmental impacts for waste batteries that contain LTO rich anodes in the waste feed, following the ISO standards 14040:2006 (ISO 14040, 2006) and 14044:2006 (ISO 14044, 2006). The study was conducted from grave-to-gate and the assessment was carried out with OpenLCA software that utilized the Ecoinvent 3.10 database. The functional unit selected as the basis for the calculations was 1 kg of black mass (with the composition shown in Table 1) entering the recycling process.

The system boundary presented in Fig. 1, represents a grave-to-gate analysis. The scope was to recover lithium, nickel, cobalt, and manganese through a hydrometallurgy flowsheet and to investigate lithium titanate oxide materials behavior within the emerging hydrometallurgical recycling process. Hazardous waste was considered to be treated on-site, thus no environmental burden for transportation was included. The geographical location for the study was set as Europe where possible, although due to data unavailability some geographical locations were set to either global or the closest representation of Europe, for example, Germany, Switzerland.

An avoided burden/system expansion approach was utilized to compare the impacts of metal recovery from End-of-Life (EoL) batteries to primary metal production impacts to avoid allocation (European Commission, 2010). A substitution method 1:1 calculation was made,

which considered the amount recovered by the flowsheet against the mining/harvesting and primary production of virgin metals to allow for a fair comparison. This approach has been previously employed by studies addressing metal recycling and provides a suitable base case scenario (Aromaa et al., 2022; Rinne et al., 2021).

Data sensitivity was approached by assessing the recycling system's dependency on electricity supply through variations in the levels of renewable energy sources (RES). Additionally, a sensitivity analysis for SC1 was conducted by varying the amount of the leaching residue that was sent to incineration. Uncertainty was addressed by assigning Data Quality Indicators (DQ) with expected accuracy levels (AL) to every item in the LCI to better understand and estimate the uncertainty of the simulation results. The results of the sensitivity analysis and data quality indicators are included in Annex 1.

2.4. Inventory analysis

Inventory analysis was carried out based on process simulation with HSC-SIM 10. Previously, it has been determined that process simulation is an efficient approach to compile detailed information, particularly when there is a lack of quality data available from either industrial or acceptable literature sources (Reuter, 1998; Aromaa et al., 2022; Elomaa et al., 2020; Rinne et al., 2021). The solvent extraction chemicals (D2EHPA, Cyanex 272, TBP) were not taken into consideration within the LCI. It must be noted that the organic phase used in the solvent extraction (SX) is recirculated within the system, therefore only minor additions are required to compensate for any losses that occur due to degradation - which are estimated to be around 1–5 % yearly, depending on the given system (Cheng et al., 2011). Consequently, the proposed diluent used in SX (Kerosene) was also excluded from the LCI. Moreover, as the two scenarios under comparison utilize the same amount of chemicals for the SX steps, it was decided to exclude this from both schemes to ensure a fairer comparison, therefore no significant effect on the LCA was considered. Results of the related LCI grave-to-gate simulation are presented in Table 2.

2.5. Impact assessments methods & indicators

ReCiPe 2016, with a hierarchist perspective (Huijbregts et al., 2017), was utilized to generate the LCIA. The impact categories for this assessment were selected following the recommendations of (Santero and Hendry, 2016) for use in LCA related to metals. The impact categories analyzed for this assessment were global warming potential (GWP, measured in kg CO₂-eq.), terrestrial acidification potential (AP, kg SO₂-eq.), freshwater eutrophication potential (F-EP, kg P-eq.), marine eutrophication potential (M-EP, kg N-eq.), ozone depletion potential (ODP, kg CFC-11-eq.), ozone formation, human health potential (HOF, kg NOx-eq.), ozone formation, terrestrial ecosystems (TOF, kg NOx-eq.), and human carcinogenic toxicity (HCT, kg 1,4-DCB).

3. Results

3.1. Impact analysis

Two separate scenarios (SC1 and SC2) were analyzed using OpenLCA with Ecoinvent 3.10. The results from SC1, SC2 and the system expanded to virgin material mining (VM) are shown in Table 3. The first scenario SC1 presents the battery recycling flowsheet that includes hazardous waste management via on-site incineration for the leach residue. This was initially modeled as "current practice" and the related environmental impacts are shown in Fig. 2a. The second scenario shown in Fig. 2b (SC2), details a battery recycling flowsheet that includes hazardous waste management through controlled underground disposal. The process flowsheet for SC1 and SC2 (displayed in Fig. 1) are otherwise identical.

It was observed in Fig. 2a that the principal contributors in the SC1

Table 2
Grave-to-gate LCI for the battery recycling flowsheet*.

Input			Output		
Leaching			Leaching		
Black mass	1	kg/kg	Leaching residue	0.405	kg/kg
Sulfuric acid	1.84	kg/kg	Oxygen gas	0.139	Nm ³ /kg
Hydrogen peroxide	0.34	kg/kg	Hydrogen	0.012	Nm ³ /kg
			Water vapor	4.208	Nm ³ /kg
Copper			Copper		
Cementation			Cementation		
Iron scrap	0.02	kg/kg	Copper	0.027	kg/kg
Iron-Aluminium			Iron-Aluminium		
Removal			Removal		
Oxygen, liquid	0.01	kg/kg	Oxygen gas	6.5 × 10 ⁻⁶	Nm ³ /kg
Caustic soda (Sodium hydroxide)	0.78	kg/kg	Iron hydroxide	0.061	kg/kg
			Aluminum hydroxide	0.017	kg/kg
Manganese			Manganese		
Recovery			Recovery		
Manganese sulfate	0.01	kg/kg	Manganese dioxide	0.139	kg/kg
Sulfuric acid	0.17	kg/kg	Oxygen gas	0.005	Nm ³ /kg
Caustic soda	0.24	kg/kg			
Oxygen, liquid	0.03	kg/kg			
Sulfur dioxide, liquid	1.7 × 10 ⁻³	kg/kg			
Cobalt			Cobalt		
Recovery			Recovery		
Cobalt Sulfate	0.01	kg/kg	Cobalt sulfate	0.32	kg/kg
Sulfuric acid	0.17	kg/kg	Water vapour	0.033	Nm ³ /kg
Caustic soda (Sodium hydroxide)	0.14	kg/kg			
Nickel			Nickel		
Recovery			Recovery		
Nickel sulfate	0.06	kg/kg	Nickel sulfate	0.849	kg/kg
Sulfuric acid	0.46	kg/kg	Water vapor	0.081	Nm ³ /kg
Caustic soda	0.37	kg/kg			
Lithium Recovery & Sodium Sulfate			Lithium Recovery & Sodium Sulfate		
Removal			Removal		
Sodium carbonate	0.34	kg/kg	Lithium carbonate	0.244	kg/kg
Caustic soda (Sodium hydroxide)	2.53X10 ⁻³	kg/kg	Sodium sulfate (anhydrous)	3.513	kg/kg
Effluent			Effluent		
Treatment			Treatment		
Lime	0.01	kg/kg	Waste water	4.78	kg/kg
			Solid Waste	0.071	kg/kg
General Energy	1.34	MJ/kg			
General Steam	4.02	MJ/kg			
General Water	8.09	kg/kg			

* The results presented are from the simulation, the scenarios were subsequently created using Open LCA.

recycling process are the use of caustic soda (sodium hydroxide), nickel sulfate, soda ash, hydroxide peroxide, the incineration of waste, and sulfuric acid. Caustic soda is used in the iron-aluminum removal, manganese recovery, cobalt recovery, nickel recovery and lithium recovery as well as in sodium sulfate removal. Thus caustic soda is responsible for a significant amount of the total environmental burdens. This is reflected in the results for SC1, with NaOH being responsible for 28 % GW, 48 % F-EP, 23 % M-EP, 41 % ODP, 33 % for H—OF and TE-OH and 30 % HCT. In contrast, nickel sulfate usage, which comes from the scrubbing step within the SX nickel recovery stage and is only utilized in that single process step, has the biggest contribution in the ODP category (15 %) as well as 14 % for ADP. The primary use of sulfuric acid is in leaching although it is also used in solvent extraction for Co, Mn, and Ni, and the used of sulfuric acid had the following environmental impacts

Table 3

LCIA results for SC1, SC2 and the mining of virgin materials (VM).

EI	Reference unit	SC1	SC2	VM
AP	kg SO ₂ -Eq	4.31×10^{-02}	4.25×10^{-02}	1.31×10^{-01}
GW	kg CO ₂ -Eq	5.06	4.09	12.81
F-EP	kg P-Eq	2.15×10^{-03}	1.87×10^{-03}	5.68×10^{-03}
M-EP	kg N-Eq	7.18×10^{-04}	6.97×10^{-04}	3.79×10^{-03}
HCT	kg 1,4-DCB-Eq	1.30	1.27	3.18
ODP	kg CFC-11-Eq	1.80×10^{-06}	1.60×10^{-06}	7.55×10^{-06}
H-OF	kg NO _x -Eq	8.43×10^{-03}	7.91×10^{-03}	3.20×10^{-02}
TE-H	kg NO _x -Eq	8.94×10^{-03}	8.36×10^{-03}	3.36×10^{-02}

contributions 61 % AP, 12 % for H—OF and the remaining of the categories below 11 %. Of the remaining significant contributors, sodium carbonate – needed for the precipitation of lithium as lithium carbonate – had its biggest impact on M-EP with 43 %, whereas the impact on the rest of the environmental categories were <10 %. Additionally, hydrogen peroxide has an 18 % contribution to the HCT category and 12 % or less in the other remaining impact categories. Technically, the use of this chemical (H₂O₂) can also be avoided, as there is evidence that peroxide-free leaching is possible (Chernyaev et al., 2021; Porvali et al., 2020) and could in future, potentially offer a more sustainable route for future battery recycling. Inclusion of residue incineration as the hazardous waste management within the SC1 scenario contributed the following additional burdens: 21 % to GW, 14 % F-EP, and 15 % to HCT.

The principal contributors to the second recycling scenario (SC2) showed identical overall chemical usage to that of SC1. Results from the environmental impact assessment generated for SC2 are shown in Fig. 2b. These findings showed that the use of sodium hydroxide, with 35 % of GW, 56 % F-EP, 24 % M-EP, 45 % ODP, 36 % H—OF, 35 % TE-OF and 31 % HCT was one of the major contributors, along with sodium carbonate (46 % for M-EP). In addition, sulfuric acid use in SC2 was determined to have a similar level of impact on SC1 with 62 % for AP and 20 % and 12 % for HCT, H—OF and TE-OF respectively and additional other contributions of <8 %. The overall contribution of sodium carbonate was also found to be of an almost equivalent level in SC2 (vs. SC1) of 44 % for M-EP and <11 % across the rest of the categories. The contribution of hydrogen peroxide demonstrated higher levels for both HCT (19 %) and GW (15 %), and around ~ (5–9 %) in the rest of the categories except AP with 2 %. The results might be associated to the relatively intensive impacts of HCT and GW emissions from background processes. In contrast, the AP of the background processes are less intensive, therefore generating minimal kgSO₂-Eq/kg emissions. Nevertheless, the most noticeable change calculated for the environmental impacts resulted from the shift in hazardous waste management

methodology from incineration (2.4 % to GW and 12 % to HCT) to underground disposal (≤ 3 % in all categories).

Fig. 3 provides a comparison of the environmental impacts in the system expansion of SC1 and SC2 vs. the mining of virgin materials for cathode manufacture. The 100 % indicated in Fig. 3 represents the environmental impacts of virgin cobalt, nickel sulfate, manganese dioxide, and lithium carbonate mining, processing, and final purification. When comparing SC1 vs. SC2 it is observed that SC2 has a smaller EI contribution than SC1 in every impact category. In particular, there are reductions observed in both GWP (10 %) and F-EP (6.9 %), with the remaining impact categories all showing some margin of decrease (≤ 6 %). The difference between the two scenarios results from the inclusion of combustion by rotary kiln in SC1 to minimize the total amount of hazardous waste as ashes (Dempsey and Oppelt, 1993). Even though SC2 has smaller EI contribution than SC1 in every impact category, due to the nature of the data which was obtained from laboratory scale findings and that the difference is not highly significant, this study cannot definitively demonstrate that SC2 is more environmentally friendly than SC1.

On the other hand, the use of underground deposits for disposal is a viable alternative option for hazardous waste management, especially when the available technology to deal with the hazardous waste generated is limited (Visvanathan, 1996). Nevertheless, according to Zhu et al. (2023), incineration is presently the most popular hazardous waste management practice to deal with the graphite-containing waste that is generated from the battery recycling processes. Therefore SC1 can be considered as being currently more representative of the industrial operations used in battery recycling. For SC1, this battery recycling approach reduces GW by 45 %, AP by 50 %, 47 % for F-EP, 49 % M-EP, 47 % ODP, 48.5 % H—OF, 48.5 % TE-OF, 49.5 % HCT, when compared to the primary production of the materials recovered from the battery recycling flowsheet. In contrast SC2, although minimal, provides a further reduction of the associated environmental impacts: 55 % for GW, 51 % AP, 54.5 % F-EP, 51 % M-EP, 53 % ODP, 52 % H—OF, 52 % TE-OF and 51 % HCT. - Previous battery recycling studies reported GW as 2.07 (pyro-hydro) and 2.64 (thermomechanical-hydro flowsheets) kgCO₂-Eq, respectively (Van Hoof et al., 2023). It is worth noting that the environmental impacts of the flowsheet presented in this study are not exactly aligned with previous research due to the differences in flowsheet, feeds, and material recovered. For instance, although Van Hoof et al. (2023) included leach residue incineration, the environmental burden was not considered due to the assumption of energy recovery. Consequently, the present study demonstrates - by comparing both battery recycling scenarios - the hypothesis that even though the system boundaries include two different hazardous waste management

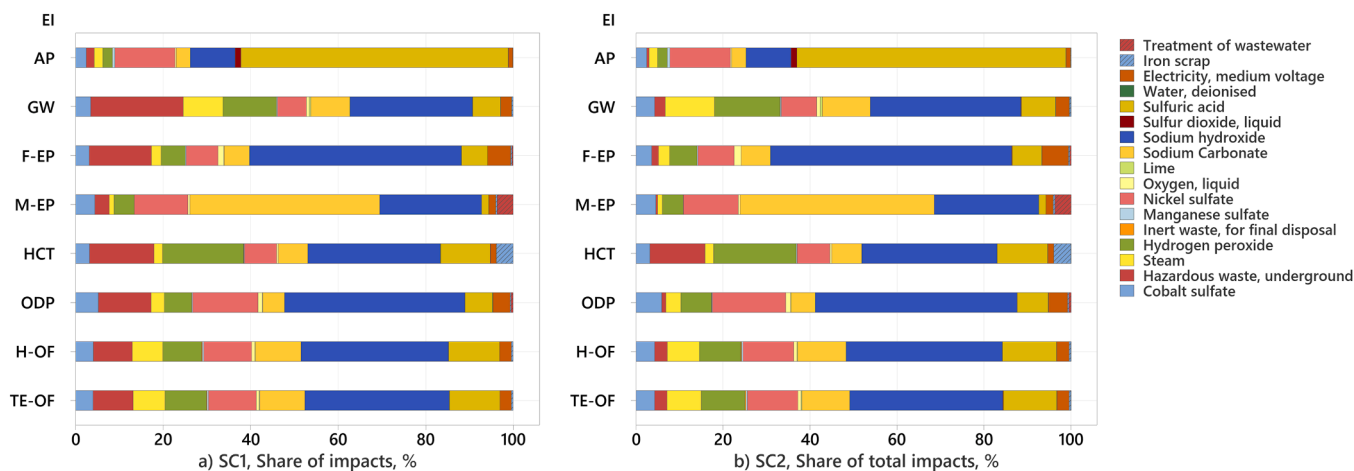


Fig. 2. a) Calculated environmental impacts related to the waste incineration route, SC1, and b) Calculated environmental impacts related to the safe underground disposal route, SC2.

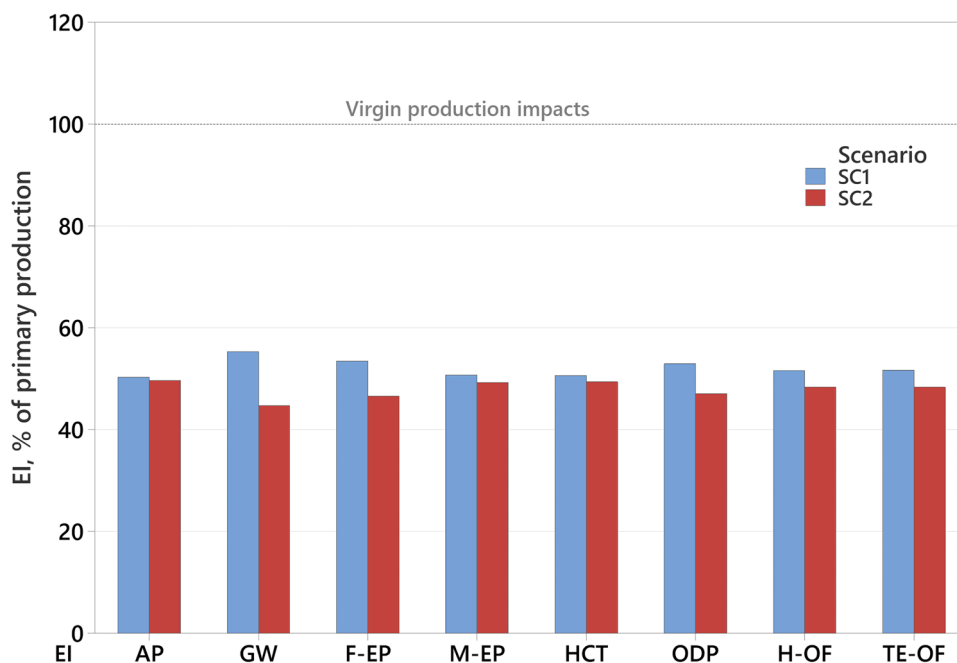


Fig. 3. Benefits comparison of the environmental impacts of SC1 and SC2 vs. virgin materials (defined as = 100 % EI Contribution).

protocols, there are still significant environmental benefits to be gained versus the production of the equivalent virgin materials.

4. Discussions

A hydrometallurgical battery recycling flowsheet for NMC-graphite and NMC-LTO rich black mass was studied in the current paper. The models focus on a hydrometallurgical approach, therefore the mechanical pretreatment steps of the waste batteries such as dismantling, crushing, and sieving were excluded from the system boundaries. Additionally, the potential effects due to the presence of residual electrolyte and binder materials were also considered to be outside of the scope of work of this study, thus the work focused solely on cathode, anode and conductor materials. This can be highlighted as being a weak point in the existing experimental literature – as scientific papers rarely report the behavior or impact of electrolyte and binder materials in the metallurgical recycling and unit processes. Data obtained from literature sources for the flowsheet simulation are typically comprised of laboratory batch experiments which might not wholly resemble the industrial scale. It could be expected that continuous operations that aim at higher efficiencies will affect process retention times and therefore, the various elemental concentrations within the final PLS.

The mass percentage of the LCO, LNO, and LMO was changed to stoichiometrically match that of a NMC811 composition, since the original data correspond to industrial black mass that was of the type 532 based on calculations. Additionally, it was assumed that the Al, Cu and Fe content in LTO material were the same as that from NMC due to data gaps. Cobalt sulfate and nickel sulfate data quality relies on the estimation of data obtained from SX based on NMC 111, however the feed for the system is a mix of NMC 811/111 cathodes. Solvent extraction chemicals (D2EHPA, Cyanex 272, TBP) were excluded from the LCI owing to a relatively low level of yearly losses in the SX system. Furthermore, in some cases of data unavailability, it was necessary to utilize either a global geographical location or the closest representation to Europe e.g., Germany, Switzerland etc. to attain reasonable estimates from the database.

There remains a degree of uncertainty related to the LCI (detailed in Table 1) that must be considered when comparing the results with other battery recycling LCAs. It was determined from the data quality

indicators that uncertainty arises from lack of appropriated data for the simulation related to the parameters for the battery recycling process, the resemblance from batch experiments to the industrial scale, and the absence of thermodynamic information for the system. Hence, the data quality used in the simulation might have a considerable impact on the model accuracy, reliability, and validity of the results.

Even though the effluent from the wastewater was treated, it still contained approximately 0.5 % (~1.2 g/L) of SO_4^{2-} according to the HSC-sim simulation, which suggests that further wastewater treatment would be required (Chatla et al., 2023), which was beyond the scope of this study. As mixed waste salts are illegal to dispose of in landfill due to the complications that are introduced when treating landfill leachate, the production of salts as by-products is considered an environmental concern (Tuovinen et al., 2021; Adesina and Rodrigue Kaze, 2021; Annamalai et al., 2022). Although sodium sulfate ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) was also obtained as a by-product from the simulation of the battery recycling process the associated environmental impacts of this material were also deemed to be beyond the scope of this study. $\text{Fe}(\text{OH})_3$, and $\text{Al}(\text{OH})_3$ were categorized as non-hazardous waste according to their corresponding SDS (Sigma Aldrich, 2023b; Chemical, 2024), however further laboratory validation should be carried out to ensure their properties are not altered by the battery recycling process. Additionally, the environmental impacts of the non-hazardous waste were also excluded as it was considered that their waste management would not represent a significant increase in the environmental impacts and that they could be used for co-processing in the manufacturing of other applications (Song et al., 2017).

Regardless of the level of uncertainty and sensitivity of the process, (see Annex 1) it was evident that there was a decrease of the EI when compared to the mining and refining of similar virgin materials. Hence, this study confirms the hypothesis that recycling materials from spent batteries can be more environmentally friendly than virgin materials production. Nonetheless, when a comparison between incineration and underground disposal is undertaken, the overall difference cannot be considered as significant therefore it is currently unclear whether the use of underground deposits for disposal results in a reduced environmental burden. LCA practitioners that would like to exploit the results detailed here in the future must exercise caution and aim for relevance at the functional unit, system expansion, goal and scope of the study, data

uncertainty and all the assumptions made in order to carry out this study.

The sensitivity analysis, included in Annex 1, conducted for different feeds—NMC811/graphite and NMC111/LTO in a 50/50 ratio and NMC811/graphite only—revealed no significant differences in yields, except for lithium, which increased from 88 % to 92 %. The LCA results showed only a small difference in their environmental impact contributions (approximately ± 6 %). Nonetheless, as a direct evaluation of the two systems is challenging due to the difference in outputs, further research on scalable graphite and titanium oxide recovery methods is needed to enable a more precise comparison.

Despite the need for additional improvement to the simulated flowsheet due to data gaps, this research provides a credible and justifiable appraisal of what could be considered current practice in an industrial-scale battery recycling process. Furthermore, the findings are consistent with other LCA/LCI process modelling driven assessments of similar focus. Nevertheless, it is suggested that further research must focus on improving the data quality through experiments related to battery recycling processes, researching routes to minimize waste, typically from the anode material, to deliver an optimal battery recycling process, which efficiently recovers battery-grade materials that aim for environmental friendliness to successfully achieve sustainability.

5. Conclusions

In the current research, a hydrometallurgical battery recycling flowsheet was simulated to study the recycling of battery waste materials (NMC-811/graphite) together with recently commercialized LTO-based battery materials (NMC111/LTO). The results demonstrated that for the studied recycling process lithium, manganese, nickel, and cobalt could be recovered, whereas graphite and titanium oxide remained in the leach residue. The recovery rates of the process were simulated to be 88 % Li, 96 % Co, 93 % Ni and 93 % Mn, values that exceed the EU battery regulation recycling targets set for 2027 (European Parliament, 2023) (50 % Li, 90 % Co, and Ni). Nonetheless, it is worth noting that such recovery levels will not fulfill the 2031 recycling target for nickel (95 %), highlighting the need for further research related to nickel extraction and recovery.

Additionally, a grave-to-gate LCIA was carried out - using the life cycle inventory results obtained from the process simulation - to quantify the environmental impacts of the recycling process and assess two hazardous waste management routes. Outputs of the LCIA showed that the use of caustic soda, hydroxide peroxide, nickel sulfate, sodium carbonate, and sulfuric acid within the recycling process were the principal contributors to the environmental impacts. Thus, innovations in the background processes for these chemicals and optimized use could further help in decreasing the environmental impacts. Furthermore, the associated sensitivity analysis undertaken indicated that the environmental impacts of the battery recycling process are not highly dependent on the power generation mix from which the energy is supplied.

In the current state-of-the-art scientific literature, the environmental impacts of battery recycling waste management have been mostly overlooked. Therefore, two hazardous waste disposal approaches were investigated for the treatment of the recycling process leach residue. The methods studied were incineration (SC1) and stabilization and underground storage (SC2), available in the Ecoinvent 3.10 database. The findings suggested that hazardous waste management has an important contribution to the environmental impact of the recycling process. The treatment of hazardous waste (80 % of the leach residue) was found to contribute between ~ 2 to 21 % (SC1) or ~ 1 to 12 % (SC2) of the different environmental impacts, within the boundaries of the studied

system. This highlights the potential that leach residue reprocessing e.g., via graphite or TiO_2 valorization may provide advantages in the future. In contrast, a comparison of the environmental impacts due to the final disposal method found that incineration resulted in slightly higher (0.6–10 %-units) effects when compared to underground storage.

This study identified that battery recycling process waste with LTO-rich anode waste may result in increased waste from leach residue when compared to graphite-rich anodes. However, when comparing any of the studied scenarios (including hazardous waste management within the boundaries), clear benefits were found for battery recycling in contrast to the corresponding environmental impacts associated with producing these materials from virgin minerals.

This work outlines some of the challenges that may arise in EoL battery recycling when new and emerging chemical compositions are integrated into current battery recycling processes. Further investigation is necessary to corroborate the interaction of NMC, graphite, and LTO in the leaching stage. Simulated data must be ideally tested at the pilot plant scale to confirm the certainty of the model related to yields and purity of the recycled materials obtained through process simulation as lower final yields would result in reduced environmental benefits. In case that the purities of the recycled materials would be lower than the battery grade purities specified by industry, recycled materials would require re-crystallization or further purification stages that could lead to an increase in the predicted environmental impacts from this study. Additional experiments would be also required to construct a more reliable process simulation as the current study relies on the estimation of data obtained from SX experiments, which are based on lower nickel concentrations (NMC 111 waste). Overall, the study highlights the importance of experimental and industrial-scale data availability for recycling, particularly for emerging battery chemistries like LTO.

Supplementary Information

Supporting information: Detailed process descriptions, flowsheets, simulation design parameters can be found in the online version (PDF)


CRediT authorship contribution statement

Diana Arellano-Sanchez: Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Conceptualization. **Marja Rinne:** Writing – review & editing, Supervision, Software, Methodology, Investigation, Conceptualization. **Benjamin P. Wilson:** Writing – review & editing, Supervision, Project administration, Conceptualization. **Mari Lundström:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization.

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.

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Supplementary materials

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Appendix 1

A1.1. Sensitivity and uncertainty analysis

Several researchers have attempted to predict different renewable energy sources (RES) scenarios that fulfill the required energy demand, whilst simultaneously meeting the European Union-mandated decarbonization goals to reach carbon neutrality by 2050 (European Commission, 2021; Knopf et al., 2015; Li and Leung, 2021). As a result, one of the largest factors of uncertainty is the electricity generation from where the system will be supplied and how it will affect the environmental impacts of the studied system. Hence, an analysis was carried out to understand the dependency of the recycling process on energy. Results of the sensitivity analysis are presented in Fig. A1 (a) based on the different scenarios of energy supply outlined by Knopf et al. (2015). The assessment showed that the recycling processes is not highly dependent on energy meaning that the model it is not overly sensitive to the electricity mix and the results should be valid regardless of future EU energy market developments. Nonetheless, it should be noted that the analysis detailed here only includes the main recycling methods and excludes upstream processes like waste battery collection, dismantling etc. (See Fig. 1). According to the results of this assessment it is expected that if the same methodology would be applied to the operations further upstream, there would be a more noticeable change in the calculated impacts due to the more energy intensive nature of the mechanical pretreatment steps that can include large crushing, magnetic separation and sieving (Peng et al., 2018; Pudas et al., 2011).

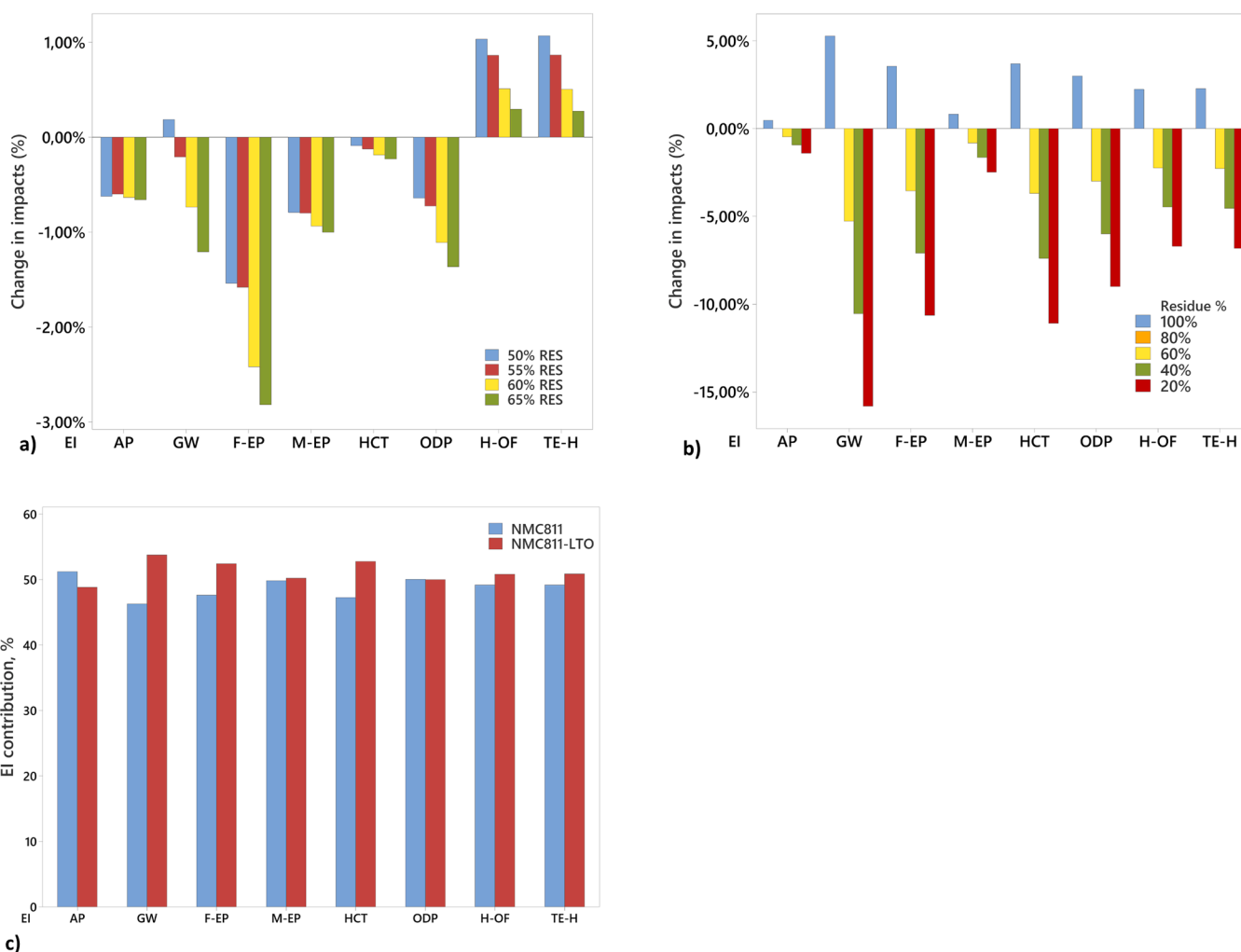


Fig. A1. a) Change (%) of the environmental impacts concerning the energy supply (based on data from Knopf et al. (2015)). b) Changes (%) in the environmental impacts of SC1 due to the amount of leaching residue waste stream sent for incineration. c) Comparison of the impacts resulting from a NMC811/graphite feed vs. NMC811/graphite (50 %) and NMC111/LTO (50 %). Percent is calculated within the levels of every impact category.

It was observed that the amount of leaching residue (LR) created was one of the primary contributors to the environmental impacts related to SC1 - for the calculations it was estimated that only 80 % of the LR ended up in the hazardous waste stream for incineration. In future scenarios, it is expected that the mass of LR (which is composed of TiO_2 , graphite and unreacted electrode active material remnants) can be reduced if a specific novel route is developed for the recycling of graphite or TiO_2 that remains after NMC-graphite and NMC-LTO battery leaching. Nevertheless, as such processes are still only at the development stage (Liivand et al., 2023; Zhu et al., 2023), a sensitivity analysis for SC1 was carried out based on the assumption that different percentages of the LR were subjected to incineration. As shown in Fig. A1(b) the environmental impacts are reduced when

the total amount of incinerated LR is decreased. The assessment suggests that reducing the amounts of leaching residue created by recycling and developing novel valorization methods, notable environmental advantages and improved sustainability for the battery recycling system may be achieved. However, it must be noted that such novel valorization routes—like that proposed by Liivand et al. (2023)—may require the increased use of chemicals like H_2SO_4 , $NaNO_3$ and $KMnO_4$, amongst others. Therefore, in order to make full comparison, also these novel valorization processes should be modelled in detail to better determine their environmental impacts.

A sensitivity analysis was carried out for the recycling process in which the feed was limited to NMC811 and graphite (further details about this simulation outlined in table S4). Results from the LCA - shown in Fig. A1(c)- suggest that the difference in their environmental impact contributions is minimal ($\pm 6\%$), as the overall difference is not considered significant, this study cannot ensure that a certain material feed is more environmentally friendly than any other. Consequently, as the simulation was constructed only with data from laboratory scale sources, further research that provides higher quality and more reliable input information is needed to allow a definitive conclusion.

Data Quality Indicators (DQ) with expected accuracy levels (AL) were given to every item in the LCI to estimate and understand the uncertainty of the results. DQ1 (AL -10 to +15 %) and DQ2 (AL-15 to +25 %) are normally assigned to measured and reference data respectively. Consequently, none of the data in the LCI was assigned to these levels due to the lack of direct measures from either industrial or pilot plant operations and literature data related to similar processes. Since most of the data was obtained by simulation, estimation, or upscaled data, most of the items in the LCI are classified as DQ3 (AL-20 to +30 %), whereas electricity, steam, cobalt sulfate, and nickel sulfate are allocated as DQ4 (AL-30+50 %). Energy sourced from electricity was calculated only for the most important unit operations following a methodology previously outlined by Elomaa et al. (2020), with steam data utilized to provide the energy balance in the process simulation, however, no value for heat capacity (cp) for the lithium metal oxides were able to be referenced for the black mass to obtain the most accurate heat balance. Additionally, cobalt sulfate and nickel sulfate data quality relies on the estimation of data obtained from SX based on a NMC 111 feedstock, although the input for the modelled system was the more nickel rich NMC 811. DQ5 (AL -50+100 %) which refers to estimation was not assigned to any item of the LCI in this study. The detailed assessment of the data quality can be found in Table S7.

Data availability

Data will be made available on request.

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