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Comparing the environmental performance of industrial recycling routes for lithium nickel-cobalt-manganese oxide 111 vehicle batteries

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Abstract

The last decade witnessed an increasing global adoption of electric vehicles. Moreover, the nickel-cobalt-manganese battery chemistry has gained wide acceptance among manufacturers of electric vehicles in Europe. Nevertheless, the environmental impacts associated with recycling this battery technology have not been fully investigated. Therefore, a comparative life cycle assessment is presented for two hydrometallurgical and pyrometallurgical recycling routes for the highlighted battery technology. Both processes show a positive net environmental impact when considering avoided virgin material production, in particular nickel and cobalt. Environmental hotspots of battery recycling processes include extraction solvents, water emissions from the wastewater treatment process, and electricity consumption.

Keywords: life cycle assessment; electric vehicles; battery recycling

Nomenclature

EV	Electric vehicle
GWP	Global warming potential
FDP	Fossil depletion potential
FETPinf	Freshwater ecotoxicity potential
FEP	Freshwater eutrophication potential
IRP	Ionizing radiation potential
LCA	Life cycle assessment
LCIA	Life cycle impact assessment
METPinf	Marine ecotoxicity potential
TAP	Terrestrial acidification potential
eq.	equivalent

1. Introduction

The European Union (EU) is accelerating the uptake of passenger electric vehicles (EVs) in road transport, which will bring the European EV stock in use to 13 million vehicles by 2025 [1]. In April 2019, the EU enforced new CO_2 emission standards for passenger cars and vans that are responsible for almost 15% of Europe's CO_2 emissions [2]. These standards target the reduction of road transport related greenhouse gas emissions by 23% in 2030 compared to 2005. At present, lithium-ion cells with layered lithium nickel cobalt manganese oxide (NMC111) cathode, non-aqueous liquid electrolyte, and natural or synthetic graphite anode are in command of the light duty battery market [3]. However, advanced cell chemistries with superior performance are under development [4]. In addition to battery cells, an EV battery system contains other

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metal parts and electronic components that assist the safe operation of battery cells.

It is widely acknowledged that recycling technologies will play a key role in achieving environmental sustainability for EV batteries [5]. Therefore, Directive 2006/66/EC prohibits EU member states from landfilling or incinerating automotive batteries [6]. Additionally, the directive mandates recycling at least 50% of the battery mass. EV batteries contain numerous raw materials, some of which have high commodity value (i.e., nickel, and cobalt), while others are defined by the European Commission as important battery raw materials with potential risk in their supply chain (i.e., lithium and graphite) [7]. To prevent the loss of valuable raw materials, recyclers utilize hydrometallurgical and pyrometallurgical extraction methods to treat waste EV batteries. Most of these recycling processes particularly target the recovery of nickel and cobalt [8].

In this work, a baseline LCA for recycling an EV battery system of the prevalent NMC111 technology is presented. Two recycling routes are assessed that represent industrial hydrometallurgical and pyrometallurgical based processes. The study aims to identify environmental hotspots in both routes and potential impacts associated with the recycling feedstock from each process. The next sections will provide a literature review of LCA studies on EV battery recycling, a brief description of the recycling routes studied, and the actual LCA.

2. Literature review

Several LCA studies have been published on batteries from consumer electronics and EVs with the recycling stage included in the scope of the study [9, 10, 11, 12, 13, 14, 15, 16, 17, 18]. Literature review of related studies was carried out using Google and Google Scholar search engines. Search terms “LCA lithium ion battery recycling” and “LCA electric vehicles” were used. Comparative LCA studies on EVs singled out battery production and use stages as the determinants of most environmental impacts from EVs, in contrast to minor impacts from the disposal phase [9, 10, 11, 12]. Nevertheless, the study of Tagliaferri et al. suggested that potential human toxicity impacts from the disposal phase of EVs could be more than double those from internal combustion engine vehicles [12]. Naturally, this significant difference can be attributed to the recycling process of EV batteries. Additional LCA studies are available with a scope limited to the entire EV battery lifecycle [13, 14]. Similarly, LCIA results from Raugei and Winfield suggested that the environmental impacts from the recycling stage are marginal compared to those from production and use stages [14]. Furthermore, recycling process credits could be significantly influenced by the recycled lithium content [13].

Further LCA studies are available on exergy based resource savings and environmental impacts from hydrometallurgical and pyrometallurgical based recycling of lithium mixed metal oxide cathode batteries [15, 16, 17, 18]. For hydrometallurgical based recycling, most of the GWP impacts are generated in the hydrometallurgical treatment stage, whereas most of the avoided burdens happened in the dismantling stage where 34% of the battery mass was recovered [15]. On the basis of cumulative exergy extracted from the environment, cobalt and nickel recovery using pyrometallurgical based recycling and re-introduction to the battery production chain may halve the exergy based resource demands from the environment [16]. More recent comparative LCA studies examine the GWP from the hydrometallurgical and pyrometallurgical based recycling of the NMC111 battery [17, 18]. Both studies use the system expansion method for evaluating associated recycling credits, and concluded that the hydrometallurgical recycling process has lower direct GWP impacts and higher avoided global warming emissions from the recycled feedstock, compared to the pyrometallurgical process.

3. Materials and methods

3.1. NMC111 EV battery system inventory and recycling routes

An inventory of the NMC111 EV battery is provided by partners from Empa that will be fully disclosed in an upcoming comparative LCA study on EV battery production [19]. The supplied inventory is used as the starting point for the recycling process inventories and to apply a mass balance approach for tracking recycled content and waste streams from different process stages. While there are at least fourteen industrial battery recycling processes [20], this paper will only evaluate three generic ones termed as Process H_1, Process H_2, and Process H_3.

Process H_1 is a three stage pyrometallurgy based treatment process constructed from the Everbatt report [21]. In the first stage, the EV battery system is manually disassembled. Then, aluminium from the cooling jacket, and electronic components from the management unit are retrieved. In the second stage, cells are reduced in a shaft furnace with slag components to produce a copper based metal alloy (matte), slag, and flue dust. Based on furnace operating conditions and selected slag components, cobalt, nickel, and copper in the electrodes are concentrated in the matte. In the third stage, the matte goes through further solvent based extraction to produce high purity nickel and cobalt salts that are suitable for battery applications. Although patents exist for lithium recovery from slag [22], half of the slag produced from the shaft furnace is reused in the furnace, while the other half is landfilled. Process stages are illustrated in Figure 1.

Process H_2 is a solvent extraction based hydro treatment process that is founded on the work of Peng et al. [23]. This process requires manual disassembly, and thermal and mechanical pre-treatment to deactivate the cells and retrieve the electrode material. The thermal treatment takes place inside a vacuum furnace where the cells are pyrolized in an inert argon atmosphere. As a result, the cells are deactivated and organic content from the electrolyte, solid electrolyte interface (SEI), and electrode binder material is released. Then, the fine electrode material fraction is retrieved from inside the cells by mechanical separation activities that include shredding of deactivated cells, sieving, and air classification. Additionally, electrode foils and plastics from cell separators and housing material are separated from the fine fraction. The fourth stage involves leaching the fine electrode material in a sulfuric acid solution while maintaining a 100 g/L solid to liquid ratio [23].

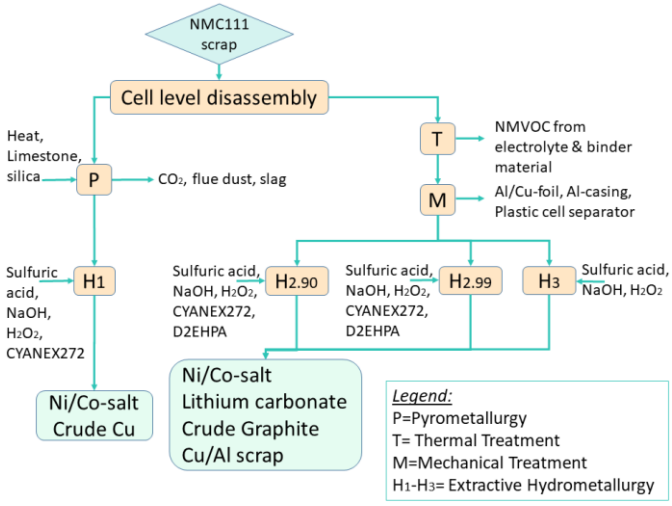

Figure 1: Process H1, Process H2, and Process H3 recycling stages

Firstly, graphite is recovered because it does not dissolve in the acid medium. Next, aluminium, and cobalt are selectively removed from the acid medium using solvents, whereas nickel and copper are removed by pH value manipulation. At a later stage, the leaching solution is concentrated and lithium is recovered by carbonation. Although solvent recycling is a common practice in extractive hydrometallurgy due to its high price, it was not disclosed in the study of Peng et al. [23]. As a result of the uncertainty in the amount of solvent that can be reused, this study suggests one scenario of Process H_2 where 90% of the solvents can be reused (Process $H_{2.90}$) and another where 99% of the solvents can be reused (Process $H_{2.99}$).

Process H_3 is a hydrometallurgical based treatment process that is founded on the work of Wang and Friedrich [24]. Process H_3 also requires thermal and mechanical treatment stages prior to hydrometallurgical extraction In the extraction process, the pH value of the leaching acid medium is adjusted with sodium hydroxide (NaOH) to co-precipitate targeted metals in the form of metal hydroxides Process H_3 has the advantage of efficient recovery of metals with no use of solvents. At first, graphite is filtered out because it does not dissolve in the acid medium. Copper is then cemented with the use of iron powder, followed by selective precipitation of aluminium. Following, nickel, cobalt and manganese are co-precipitated at higher pH values, before lithium is finally retrieved from the solution by crystallisation.

3.2. LCA: Goal and scope

The goal of this study is to identify potential environmental impacts from recycling 1 kg of NMC 111 EV battery system to avoid its landfilling or incineration in accordance with the 2006/66/EC batteries directive. Therefore, 1 kg of NMC 111 EV battery system input to recycling process is the reference flow of recycling scenarios introduced in section 3.1. In addition, the study aims to give proper credit to avoided environmental burdens from recycled feedstock in order to estimate the net environmental burden of different recycling routes of EV batteries. The scope of the study is Gate-to-Gate analysis from receiving the battery system at recycling plant to producing marketable products. LCA results are intended to be used for supporting decisions for technology scenarios, policy options, and design considerations for future battery and recycling technologies.

Environmental impacts from recycling processes are evaluated at a midpoint level using the hierarchist perspective of the ReCiPe 2016 method [25]. However, only results for the seven impact categories with highest normalized scores in all recycling scenarios will be reported. In addition, Brightway2 python framework is used to calculate LCIA scores. Process datasets are obtained from the Ecoinvent V3.6 database cutoff system model because the by product supply chain is relevant for the study and to avoid double counting when otherwise applying the system expansion approach with the allocation at point of substitution system model. If available, average European market activities are used to model background activities. An attributional modelling approach is selected for this study.

The choice of a recycling process strongly influences the quantity of recycled feedstock and its purity. For instance, Process H_1 does not recover neither the anode's graphite content because it is consumed in the shaft furnace as a reducing agent, nor the cathode's lithium content because it is mainly lost to the slag and flue dust. In contrast, Processes H_2 and H_3 can recover considerable amounts of graphite and lithium in the fine electrode powder by hydrometallurgical extraction. Thus, the system expansion approach is used to have meaningful comparisons for the environmental impacts from recycling processes and to avoid allocation of impacts or partitioning recycling processes. In other words, recycled material will avoid the production of a primary product at a certain point of substitution where both primary and secondary products display similar functions, which is purity level in this case. Table 1 details assumptions for the points of substitution of recycled materials.

Table 1: Points of substitution assumed for recycled feedstock from different recycling routes

	Process H_1	Process H_2	Process H_3
Lithium	N/A	Lithium carbonate	Lithium carbonate
Nickel	Nickel sulfate salt	Nickel sulfate salt	Nickel sulfate salt
Cobalt	Cobalt sulfate salt	Cobalt sulfate salt	Cobalt sulfate salt
Graphite	N/A	Graphite (below battery grade)	Graphite (below battery grade)
Aluminium	N/A	Aluminium oxide, metallurgical	Aluminium oxide, metallurgical
Copper	Copper concentrate	Copper concentrate	Copper concentrate

3.3. LCA: Inventory

A modular bottom-up approach is adopted in developing the LCA inventory of this study. In addition, pyrolysis process data

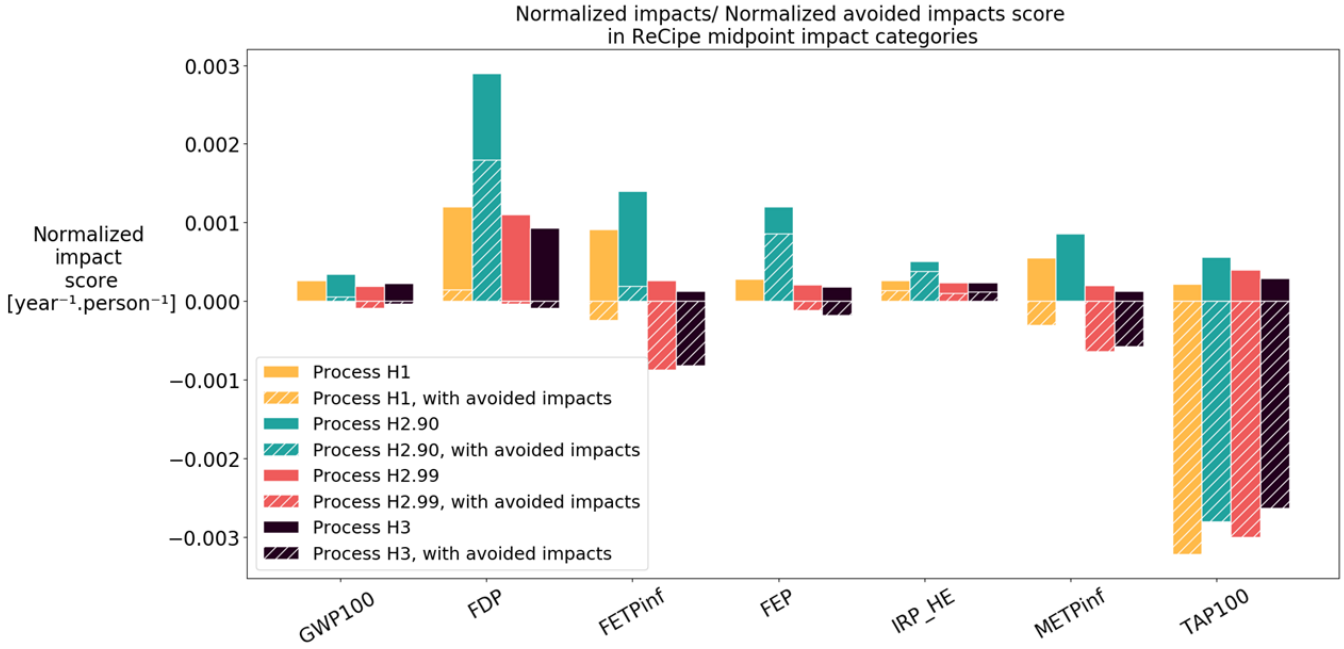

Figure 2: Normalized LCA midpoint impact category scores with and without recycling feedstock credits

are based on previous work from Diaz et al. on evaluating gases from thermal processing [26]. Estimates for electricity consumption in the mechanical treatment stage are derived from datasheets of commercial waste electrical and electronic equipment (WEEE) processing plants [27]. The inventory of the battery furnace reduction process is based on data published in the EverBatt report [21]. Additionally, the inventory of solvent based extractive hydrometallurgy is adapted from the work of Peng et al. [23]. Finally, the inventory of the chemical co-precipitation extractive hydrometallurgy is adapted from the work of Wang & Friedrich [24].

4. Results and discussion

4.1. LCA: Impact assessment

This study evaluates the potential environmental impacts from considered process paths but does not provide an insight into economic aspects of recycling processes. LCIA results suggest comparable environmental impacts from the recycling routes of processes H_1, $H_{2.99}$, and H_3. On the contrary, Process $H_{2.90}$, with 90% of the solvents being recycled, is the least favorable route. However, most studies reviewed earlier do not report solvents in their LCA inventory. Moreover, the solvents considered in this study are organophosphorus compounds that can be associated with significant environmental impacts from the production process of the solvent and the consequent phosphorous emissions to water during wastewater treatment.

The environmental impacts from extractive pyrometallurgy are on a level with hydrometallurgy ones. The GWP of 1 kg of EV battery system from Process H_1 is 2.1 kg CO_2 eq., mainly from shaft furnace electricity consumption and direct CO_2 emissions from subsequent post combustion gas treatment. The electricity demand of the shaft furnace drives the score of the IRP category to 0.15 kg U235 eq. The matte hydrotreatment to recover cobalt sulfate and nickel sulfate salts demands kerosene, CYANEX272, and NaOH. As a result, the FDP impact score is 0.68 kg oil eq., FEP impact score is 1.82E-04 kg phosphorous eq., and TAP impact score is 9E-03 kg SO_2 eq. The normalized scores are illustrated in Figure 2.

Previous studies assume either the incineration of non recoverable products [18], or high points of substution for the feedstock [17, 15]. These assumptions have major influence on estimating net burdens, or benefits, of battery recycling. In contrast to previous work, the net GWP burden of Process H_1, after subtracting the credits of the recycling feedstock, is -2.3E-02 kg CO_2 eq., compared to -3.3 from [17] and 1.5 from [18]. Moreover, [17] estimates the net FEP burden at -2.8E-02 kg phosphorous eq. compared to 1.4E-06 from this study, while the net IRP burden reported in [17] is 7 times lower than the 8.0E-02 kg U235 eq. estimate from this study.

Extractive hydrometallurgy recycling routes demonstrate superior environmental performance with limited solvent demand, as in Process $H_{2.99}$, and best with no solvent demand, as in Process H_3. GWP is the only impact category for which the score of Process H_3 (1.8 kg CO_2 eq.) is 17% more than $H_{2.99}$. This is attributed to higher demand for NaOH by the chemical co-precipitation stage in order to achieve a final pH value of 10 in the leaching medium. Particularly, the upstream chlor-alkali electrolysis process generates significant GWP impacts.

In general, estimates for the net environmental burdens of extractive hydrometallurgy recycling routes are higher than those reported in previous work. Higher estimates are attributed to assumptions for recycling efficiency, chemicals used in the extraction process, and points of substitution assumed for the recycled feedstock. The net GWP burden of Process H_3 is -0.3 kg CO_2 eq., compared to -3.9 from [17], -2.7 from [15], and -0.3 from [18]. Moreover, [17] estimates the net FEP burden at -2.8E-02 kg phosphorous eq. compared to -4.9E-03 from [15] -1.2E-04 from this study. Furthermore, the net IRP burden reported in [17] is -0.2 kg U235 eq., whereas this study evaluates net IRP burden at 6.7E-02 kg U235 eq.

The recycled feedstock from Process $H_{2.99}$ shows the highest avoided environmental impacts, followed by the feedstock from Process H_3. Extractive hydrometallurgy has the advantage of recovering more materials from the cells, unlike extractive pyrometallurgy which losses the majority of the lithium, graphite, and aluminium content to the co-products of the shaft furnace. The main driver of the avoided environmental impacts is the recovery of nickel and cobalt in a high purity sulfate salt form, followed by the recovery of copper and aluminum, and at a lower level the recovery of lithium and graphite.

4.2. LCA: Interpretation

Pyrometallurgical extraction has the advantage of rapidly concentrating valuable elements within a metal alloy. Moreover, avoided environmental impacts from the recycled feedstock are comparable to those from hydrometallurgical extractions. In addition to potential environmental impacts from the shaft furnace electricity consumption and gas emissions, the recycling process is burdened with impacts from the production of chemicals, wastewater treatment, and emissions to water.

The greater environmental impacts from solvent based extractive hydrometallurgy recycling routes are generated during the production of CYANEX272 solvent, in particular during the production of barium hydroxide and phosphorous white precursor material. Additional environmental impacts are generated during the production of kerosene, which is used as the organic phase of extraction solvents. Therefore, more oil and gas production, more barium and phosphorous emissions to water, and more sulfate emissions to air will give rise to more environmental impacts. In Figure 3, FDP and TAP impact categories, with high normalized scores for all recycling scenarios, as well as GWP are chosen as representative impact categories to highlight recycling activities that generate most environmental impacts.

Extractive hydrometallurgy recycling routes have many more opportunities for improvement. Firstly, the solid to liquid ratio which will decide the amount of acid, neutralizing agents, and solvents is probably the first parameter to consider when upgrading or even developing new recycling processes. Secondly, organophosphorus based solvents require very high reuse rates to neutralize the potential environmental impacts from their production and even the subsequent wastewater treatment as has been shown with the CYANEX272. Thirdly, a good trade-off between solvent use and co-precipitation can avoid a high final pH value of leaching medium and the resulting demand for neutralizing agents and its subsequent environmental impacts. Finally, the choice of leaching acid, i.e. mineral, organic, or by product, can significantly influence potential acidification impacts.

5. Conclusion and future work

In this work, an LCA study on recycling EV batteries is presented. In addition, the study reports a detailed modular process inventory that considers all inputs and outputs to the process. For the system expansion approach, proper points of substitution are assigned to recycled feedstock to perform meaningful comparisons between treatment processes with different co-products.

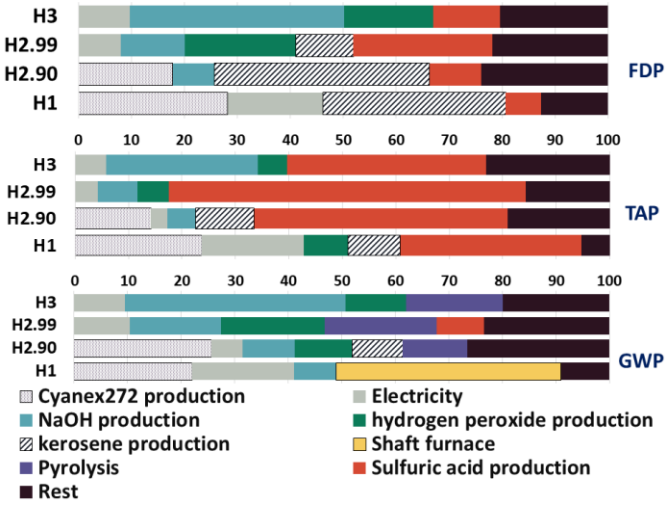

Figure 3: Contribution of different processes in battery recycling to the environmental impacts from FDP, TAP, and GWP impact categories

The LCA technique is useful in defining environmental hotspots in pyrometallurgical and hydrometallurgical based treatment of EV batteries. LCA results show potential environmental impacts from the shaft furnace gas emissions, and the chemicals used in the leaching process. Furthermore, the purity of recovered nickel and cobalt has the strongest influence on the magnitude of environmental credit to the recycled feedstock.

For future work, the impacts from additional state of the art battery chemistries as well as novel solid based lithium ion batteries will be considered. Further recycling processes that combine both extractive pyrometallurgy and hydrometallurgy will be evaluated. In addition, the consideration of product purity and recycling efficiency in the system expansion approach within the LCA study may guide future practices for defining cut-off values for solvents and acceptable recycling efficiencies and product quality. Finally, the outcomes of the LCA study will also be used to guide the developments for new recycling technologies, with high focus on lithium recovery and the quantification of environmental benefits.

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