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RESEARCH AND ANALYSIS

Life cycle assessment of lithium-ion battery recycling using pyrometallurgical technologies

Mohammad Ali Rajaeifar[1,7] | **Marco Raugei**[2,7] | **Bernhard Steubing**[3] | **Anthony Hartwell**[4,7] | **Paul A. Anderson**[5,7] | **Oliver Heidrich**[1,6,7]

[1] School of Engineering, Newcastle University, Newcastle upon Tyne, UK

[2] School of Engineering, Computing and Mathematics, Oxford Brookes University, Wheatley, Oxfordshire, UK

[3] Institute of Environmental Sciences (CML), Leiden University, Leiden, The Netherlands

[4] School of Metallurgy and Materials, Birmingham University, Birmingham, UK

[5] School of Chemistry, University of Birmingham, Birmingham, UK

[6] Tyndall Centre for Climate Change, Newcastle University, Newcastle upon Tyne, UK

[7] Faraday Institution (ReLiB project), Quad One, Harwell Science and Innovation Campus, Didcot, UK

Correspondence
Mohammad Ali Rajaeifar, School of Engineering; Newcastle University; Newcastle upon Tyne, NE1 7RU, UK.
Email: Mohammad.Rajaeifar@ncl.ac.uk
Oliver Heidrich, School of Engineering; Newcastle University; Newcastle upon Tyne, NE1 7RU, UK.
Email: Oliver.Heidrich@ncl.ac.uk

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Abstract

Among existing and emerging technologies to recycle spent lithium-ion batteries (LIBs) from electric vehicles, pyrometallurgical processes are commercially used. However, very little is known about their environmental and energy impacts. In this study, three pyrometallurgical technologies are analyzed and compared in terms of global warming potential (GWP) and cumulative energy demand (CED), namely: an emerging direct current (DC) plasma smelting technology (Sc-1), the same DC plasma technology but with an additional pre-treatment stage (Sc-2), and a more commercially mature ultra-high temperature (UHT) furnace (Sc-3). The net impacts for the recovered metals are calculated using both "open-loop" and "closed-loop" recycling options. Results reveal that shifting from the UHT furnace technology (Sc-3) to the DC plasma technology could reduce the GWP of the recycling process by up to a factor of 5 (when employing pre-treatment, as is the case with Sc-2). Results also vary across factors, for example, different metal recovery rates, carbon/energy intensity of the electricity grid (in Sc-1 and Sc-2), rates of aluminum recovery (in Sc-2), and sources of coke (in Sc-3). However, the sensitivity analysis showed that these factors do not change the best option which was determined before (as Sc-2) except in a few cases for CED. Overall, the research methodology and application presented by this life cycle assessment informs future energy and environmental impact assessment studies that want to assess existing recycling processes of LIB or other emerging technologies. This article met the requirements for a gold–silver *JIE* data openness badge described at http://jie.click/badges.

KEYWORDS
electric vehicles, emerging technologies, industrial ecology, lithium-ion batteries, plasma, recycling

1 | INTRODUCTION

The scientific literature is in broad agreement in identifying ample evidence for environmental benefits of electric vehicles (EVs), when compared to conventional internal combustion engine vehicle (ICEVs). For instance, Faria et al. (2013) found that depending on the grid supply mix of the country

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where they are driven, greenhouse gas (GHG) emissions by compact EVs may be as much as 60% lower compared to conventional ICEVs of the same size segment. Raugei et al. (2018) found that under current energy grid mix conditions, the overall life-cycle demand for non-renewable primary energy of a compact BEV in the United Kingdom is 34% lower than for a similar ICEV. Such reduction may be expected to increase further under most of the future grid mix and EV penetration forecasts (Hill et al., 2019).

Lithium-ion batteries (LIBs) are a key technological component of EVs, and as such a lot of attention has understandably been paid to their environmental performance (Ellingsen et al., 2014; Harper et al., 2019; Rajaeifar et al., 2019). However, in most published environmental studies of EVs and LIBs to date, the system boundaries are drawn to exclude the end of life (EoL) phase. This is mainly due to the fact that, compared to most other industrial activities, LIB recycling is still in its infancy (Rajaeifar et al., 2020). Thus it can be classified as an emerging technology (Bergerson et al., 2020), as only a few medium-to-large scale operators exist worldwide. Research is still under way to explore recycling processes but there is no consensus which one will be the most effective and environmentally advantageous. Whilst there might be economic incentives to recover lithium (Ambrose & Kendall, 2020; Gruber et al., 2011) or cobalt (Baars et al., 2021; Ciez & Whitacre, 2019; Skeete et al., 2020), no recycling technology can produce pure enough elements that can be used in LIBs (Mohr et al., 2020). Thus, technologies do not achieve “closed-loop” recycling yet (Sommerville et al., 2021) and instead recycled lithium, for example, is used for the production of lubricants, glass, ceramics and other products (Battery University, 2019).

In general, recycling may be considered advantageous when the environmental impacts caused by the recycling processes are outweighed by the corresponding environmental credits from the recovered materials (Baars et al., 2021; Harper et al., 2019). LIB recycling entails a range of physical or chemical processes, including intermediate and direct physical processes, pyrometallurgical (thermal) treatment and various hydrometallurgical treatments, such as leaching (including bioleaching), chemical and bio-precipitation, and solvent extraction (Li et al., 2018). Combinations of the different processes are also possible, for example, leaching and precipitation processes, or a combination of thermal treatment, leaching, and precipitation processes, to maximize the recycling efficiency of valuable materials. Pyrometallurgical processes are among chemical recycling solutions that typically use high temperature to treat LIBs and separate valuable metals. One advantage of the pyrometallurgical process is that the cells or modules can be processed after a simple manual dismantling, with no pre-treatment. However, safety risks associated with electrical hazards or flammable electrolyte solvents must be considered when dismantling battery packs and handling the dismantled cells/modules. Moreover, these processes are widely used as they are rather simple, have a short-process chain, are easy to scale up and can accept various types of battery chemistries (Liu et al., 2019; Zheng et al., 2018).

As with most, if not all, novel and emerging technologies it is difficult to compare studies and life cycle assessments (LCAs) (Bergerson et al., 2020; Moni et al., 2020). Only a small number of LCAs of LIB recycling are reported in the scientific literature and these vary significantly in their goal, scope, level of detail, LIB types and chemistries, and in the assessment methods employed. For example, Dunn et al. (2012) estimated energy savings by “closed-loop” recycling of the cathode materials and the Cu and Al current collectors for a lithium manganese Oxide (LMO) battery pack. They considered three alternative recycling routes: pyrometallurgical, hydrometallurgical, and direct physical separation, identifying potential savings from the recovery of the Al current collectors. This is at odds with the findings by Elwert et al. (2016), who analyzed hydrometallurgical recycling of a nickel manganese cobalt oxide (NMC) LIB, and found that most of the environmental benefits arose from the recovery of the outer steel casing, and, to a lesser extent, of the Co and Ni contained in the cathode. Emissions caused by the recovery of the Cu and Al electrode plates were larger than the corresponding “credits” afforded by the recovery of those metals. However, it must be highlighted that the results of Dunn et al. (2012) were theoretical estimates, which may have a larger margin of uncertainty. Hendrickson et al. (2015) also considered a theoretically “closed-loop” recycling of LMO battery packs for both pyrometallurgical and hydrometallurgical processes. In their study, most benefits appeared to accrue from lower electricity consumption in the case of the hydrometallurgical route; and pyrometallurgical recycling did not achieve significant air pollutant reductions. Cusenza et al. (2019) reported reductions in GWP and CED when recycling an LMO-NMC Li-ion battery pack using a pyro–hydro metallurgical process under closed-loop recycling. A cell-chemistry assessment was recently reported that indicates that the highest recycling benefits and lowest net impacts for NMC-type LIB are on GHG emissions (Mohr et al., 2020). Clearly many processes are used and developed to recover materials from EoL EV batteries and as Mohr et al. (2020) have shown the extent of the recycling benefit does depend on the impact categories modeled. However, there is still a large knowledge gap on environmental assessment, including pyrometallurgical processes and more specifically plasma (arc) smelting technologies. This paper aims to fill this gap, by presenting a LCA for two pyrometallurgical recycling technologies. It also reports on the environmental impacts and energy usage for an optional upgrading stage, which is a pre-treatment that reduces the size of batteries using a shredding process followed by some mechanical or thermal treatments, before LIBs are sent to the furnace.

2 | MATERIALS AND METHODS

2.1 | Goal, scope, and assumptions

LCA is a standardized and widely adopted method to compile and evaluate the potential environmental impacts of a product, process, or service from “cradle” to the “grave” (Heijungs et al., 1997; ISO 14040, 2006). The goal of this study is to conduct an LCA to compare different

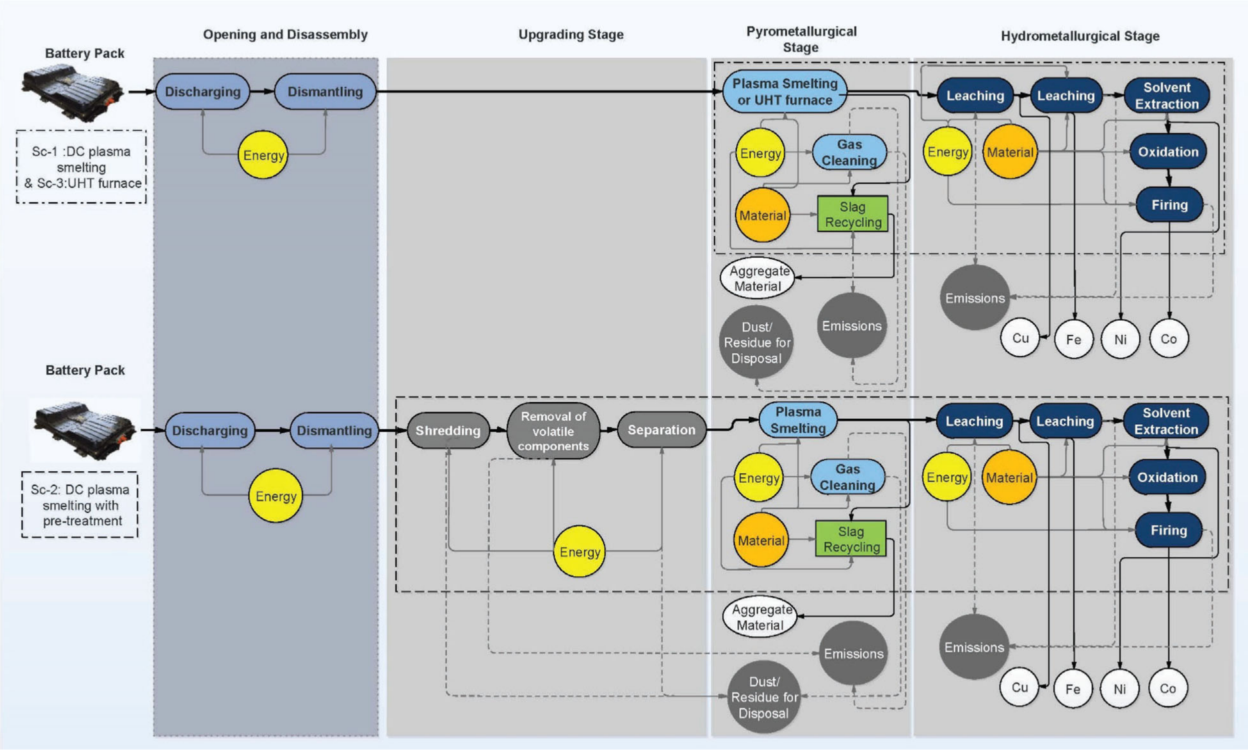

FIGURE 1 Scope for Sc-1, Sc-2, and Sc-3 (emissions presented here only include GHG emissions; background processes associated with energy and material supplies were also considered)

pyrometallurgical technologies available for the recovery of valuable metals from LIBs. Figure 1 shows the scope of the study and the system boundaries for three scenarios. Scenario 1 (Sc-1) is considering a direct current (DC) plasma smelting technology, Sc-2 is DC plasma smelting technology with pre-treatment (mechanical treatments), and Sc-3 models a commercial ultra-high temperature (UHT) furnace. All the scenarios are described in detail in Section 2.2.

Pyrometallurgical technologies typically use dismantled batteries in the form of modules or cells as the furnace feedstock (although some technologies use pre-treated materials instead (Georgi-Maschler et al., 2012; Hu et al., 2020)). Accordingly, the functional unit (FU) for this study is "treatment of 1 tonne of LIB modules" using different pyrometallurgical technologies. This translates to 1 tonne of LIB modules entering the furnace in Sc-1 and Sc-3 (because there is no pre-treatment in these scenarios), while 1 tonne of LIB modules entering Sc-2 is firstly treated using a set of pre-treatment stages and the remaining upgraded materials ending up in the furnace, that is, 440 kg. While this FU deviates from some other studies on LIB recycling, it was deemed the most suitable FU since most of the impacts associated with the recycling process tend to scale linearly with the mass treated, irrespective of the specific chemical composition of the cathode or the energy density of the modules. LIB collection, transportation, discharging, and dismantling are outside of the boundary of analysis, because of: (1) lack of data on LIB collection and transportation; (2) lack of data on the combination of materials at pack level; (3) no robust inventories for dismantling and discharging processes; and also (4) dismantling and discharging could take place elsewhere, which would create uncertainties in transportation distances to the furnace. Furthermore, the system boundary does not include capital goods, that is, buildings and machinery, used by waste treatment facilities, as they have long lifetimes (>20 years).

This study supports decisions on the micro decision-making level rather than on the macro level, and thus it adopts an attributional modeling framework (Ellingsen et al., 2014; Rajaeifar et al., 2017). The United Kingdom was chosen as the geographical location for the study.

2.2 | Analyzed scenarios and related life cycle inventory analysis

The battery cathode type that was considered for this study was NMC111 (nickel–manganese–cobalt 1:1:1). The foreground data for the DC plasma arc technology (i.e., the main pyrometallurgical process used in Sc-1, that is DC plasma smelting, and in Sc-2, that is DC plasma smelting with pre-treatment, illustrated in Figure S1-1 in Supporting Information S1) was provided by project partners (Johnson, 2019), which quantified and modeled the "Tetronics DC plasma arc technology" in collaboration with Tetronics Company (Table S1-1 to S1-3 in Supporting Information S1 and Tables S2-1 to Tables S2-3 in Supporting Information S2).

In Sc-1 (Figure 1; and route A in Figure S1-1 in Supporting Information S1), the battery modules are removed from battery packs (but remain largely or entirely intact) and are fed directly to the furnace. As no pre-treatment or disassembly are needed, no health or safety risks are posed by any hazardous or potentially flammable materials (although a potential electricity shock risk remains due to the residual voltage in the modules). This also reduces the need for manual or automated work to handle and shred the batteries. Gases leaving the furnace are then sent to a gas cleaning unit for treatment. The furnace products consist of alloys and slag which are sent for further treatment. The metal recovery efficiencies (or so-called "recovery rates") from spent LIB modules to output alloy for Sc-1 have been set here initially on the basis of the estimates provided by a detailed thermodynamic model supplied by Tetronics, that is, 99.98% for cobalt, 99.98% for nickel, 100% for copper, and 97.8% for iron. However, given that there remains a high level of uncertainty as to whether such percentages are attainable in the real world, these recovery efficiencies have been included in a sensitivity analysis (see Section S4.1 in Supporting Information S1).

In Sc-2 (Figure 1; and Route B in Figure S1-1 in Supporting Information S1), the spent LIBs are first subjected to an upgrading stage, which entails shredding, removal of volatile components, and separation. Accordingly, 1 tonne of LIB modules entered to the recycling line leads to 440 kg of upgraded material being fed into the furnace. The LCI for the shredding process was adopted from Ecoinvent (2019) and Hischier et al. (2007), while the data for removal of volatile components was adopted from the calcination process described by the Argonne National Laboratory (ANL) GREET model (ANL, 2019a). The fuel for the removal of volatile components step was considered as UK natural gas with an energy density of 35.8 MJ/m^3 (DUKES, 2019), and the electricity is assumed as that of the national grid mix in 2019 (Table S1-5 in Supporting Information S1 and Table S2-5 in Supporting Information S2). Overall, the upgrading stage can reduce the amounts of aluminum, graphite, and organic compounds entering the plasma furnace as it removes 560 kg of materials entered to the furnace compared to Sc-1, which are responsible for producing significant amounts of less useful output materials, such as alumina-based slag and combustible exhaust gases. These outputs have disproportionate impacts on the ultimate cost of the smelting system, therefore reducing the amount of materials that are responsible for generating them prior to the smelting stage is cost-effective. However, in the absence of some of these materials (i.e., carbon containing materials), the energy consumption of the furnace goes up. Hence, an additional source of energy is needed to increase the supply of energy to the furnace, that is, electricity in DC plasma technologies. It should also be noted that in this scenario, the electrolyte (which forms part of the 560 kg/tonne that are removed) could potentially pose additional health and safety risks and must be handled carefully (Sommerville et al., 2021). The metal recovery efficiencies from spent LIB modules to output alloy for Sc-2 are determined using the same thermodynamic model as for Sc-1, and are: 99.85% for cobalt, 99.89% for nickel, 100% for copper, and 96.24% for iron. These recovery efficiencies have also been the subject of a sensitivity analysis (see Section S4.1 in Supporting Information S1). Further detailed inventory data for Sc-1 and Sc-2 are reported in Tables S1-2 and S1-3 in Supporting Information S1 and Tables S2-2 and S2-3 in Supporting Information S2.

Scenario 2 is the only process that reduces the amounts of aluminum, graphite, and organic compounds entering the plasma furnace. Graphite and organic compounds are burned-off while aluminum could be separated. Due to high uncertainty in the composition of aluminum included in spent LIBs, the separation efficiency as well as the substitution rate of the recovered aluminum when used by other market segments, aluminum at this stage was not considered as a recovered product. Instead, it has been considered that all the rejected materials (including aluminum) are sent to landfill. However, aluminum recovery has been considered as part of a sensitivity analysis in the Supporting Information S1.

Sc-3 considers an alternative industrial pyro–hydro metallurgical process using a commercial technology, in which the battery modules are sent directly to a vertical shaft UHT furnace along with coke and a slag-forming agent that typically consists of limestone, sand, and slag. This may be considered as a "benchmark" against which to judge the energy and environmental performance of Sc-1 and Sc-2. The UHT furnace can be divided into three subsequent heating zones, that is, pre-heating zone (<300°C), plastic pyrolyzing zone (~700°C), and metal smelting and reducing zone in the temperature range of 1200–1450°C (Dunn et al., 2014; Cheret & Santen, 2007). Gases leaving the smelter are burnt in a post-combustion chamber using a plasma torch (>1150°C). The foreground inventory data on the UHT furnace technology were adopted from the peer-reviewed literature (Dunn et al., 2014; Cheret & Santen, 2007) and from the EverBatt model (Dai et al., 2019) developed by ANL (2019b). The metal recovery efficiencies in Sc-3 are based on data from a commercial production facility (Cheret & Santen, 2007) with 94% for cobalt, 99% for nickel, 92.8% for copper, and 64.5% for iron (see Table S1-4 in Supporting Information S1 and Table S2-4 in Supporting Information S2 for more details). Emissions from the disposal of dust in all the scenarios were considered using the LCI for dust for disposal available in the Ecoinvent V3.6 database (Ecoinvent, 2019).

In all three scenarios, the alloy leaving the smelter generally contains copper, cobalt, nickel, and some iron, and is then subjected to a series of hydrometallurgical processes. However, since the main focus of this study was on the plasma smelting technology (Sc-1 and Sc-2), the subsequent hydrometallurgical step was assessed using the pre-existing GREET model (Dunn et al., 2014) along with complementary data from the literature (Dai et al., 2019; Rubin et al., 2014; Li et al., 2013). Specifically, a five-step hydrometallurgical process was considered for the recovery of copper, iron, nickel, and cobalt from the alloy. Sulfuric acid (H_2SO_4) was considered as the leaching agent in the leaching steps, HCl in the solvent extraction, H_2O_2 in oxidation and lithium carbonate (Li_2CO_3) in the firing step; all were provided as inputs from the market (i.e., none of them are produced through the process). The same hydrometallurgical assumptions were used for Scenario 3 (Sc-3).

It is worth mentioning that substitution was considered as the methodological approach for dealing with the multi-functionality problem during the study. Specifically, given the uncertainty about the purity of the recovered metal salts, and therefore their suitability to be reused directly in LIB manufacturing (Ellingsen et al., 2014), we test here two alternative hypotheses, namely closed-loop and open-loop recycling (Nordelöf et al., 2019; Richa et al., 2017). It is worth noting that the default assumption in the GREET model for the hydrometallurgical process is closed-loop

TABLE 1 "Baseline" results for GWP and CED per FU (i.e., treatment of 1 tonne of spent LIB modules)

Recycling assumptions	"Baseline" description	Impact category	Unit	Sc-1	Sc-2	Sc-3[b]	Impact category	Unit	Sc-1	Sc-2	Sc-3[b]
Closed loop	Scenario-specific recovery rates[a]	GWP	kg CO_2-eq.	−1220	−2080	−770	CED	MJ	−30,100	−33,500	−34,800
Open loop	Scenario-specific recovery rates[a]	GWP	kg CO_2-eq.	1100	−290	1410	CED	MJ	5420	−5800	−1510

[a] A global average was considered as the source of coke used in the furnace.
[b] Scenario-specific recovery rates, based on what is reported in Section 2.2.

recycling, whereby all the metals are recovered in chemical forms and at purity levels that allow them to be used in LIB production without any further processing. Such closed-loop approach is regarded here as the "best case," providing optimistically large energy and environmental credits for the system. At the opposite end of the scale, as a bare minimum the recovered metals could certainly displace equivalent quantities of "raw" metals coming from the respective primary supply chains (i.e., ore mining and beneficiation) instead of entering directly to the LIB production supply chain. This alternative open-loop recycling approach is considered here as the "worst case," in which the recovered metals, just like their respective "raw" counterparts, would have to undergo a series of refining processes before they can be used again (irrespective of the specific product systems into which they would eventually be embedded). Further elaboration of these two approaches and the considered product for displacement in either of them are provided in Section S2.1 in Supporting Information S1. The slag generated in the pyrometallurgical stage, containing lithium, manganese, aluminum, silicon, calcium, and small amounts of iron, is subjected to a series of recycling processes to be converted to aggregate materials. Although the extraction of lithium and manganese in the slag is technically possible, according to Dunn et al. (2014) this process is not economically viable nor energy efficient. Therefore, the generated slag was assumed to replace "natural aggregate" (NA) production after a series of recycling processes. Further information on slag recycling and substitution with natural aggregate may be found in Section S2.2 in Supporting Information S1.

Finally, the background inventories regarding transportation, natural gas, and material production (for use as inputs in the plasma smelting process) were adopted from Ecoinvent V3.6. Since the default LCI data for British electricity generation in Ecoinvent refers to year 2014, the British electricity mix was updated (Table S1-5 in Supporting Information S1 and Table S2-5 in Supporting Information S2). The LCA models for the different scenarios were constructed using Umberto LCA+ Software package (ifu Hamburg, 2019).

2.3 | Life cycle impact assessment

To identify and evaluate the amount and significance of the potential environmental impacts arising from the LCI, the inputs and outputs are assigned to impact categories. In this study, the categories are considered by using global warming potential (GWP) and cumulative energy demand (CED). These were chosen as: (1) they are of relevance for energy-consuming products and processes, (2) they have been widely used in the literature, and (3) the LCI for the hydrometallurgical stage adopted in this and other studies (Dunn et al., 2014; Dai et al., 2019; Rubin et al., 2014; Li et al., 2013) does not include other, non-carbon emissions. For assessing GWP, a time horizon of 100 years was chosen (ifu Hamburg, 2019). For assessing CED, the methodology proposed by Frischknecht et al. (2007) was employed, which measures the cumulative amount of energy harvested by a process, product, or service from a range of renewable and non-renewable primary energy sources (fossil fuels, uranium, biomass, wind, solar, geothermal, and hydro).

3 | RESULTS AND INTERPRETATION

Sections 3.1 and 3.2 and Table 1 report the main GWP and CED results for the three scenarios, using a set of "baseline" assumptions, which include the metal recovery rates reported in Section 2.2. Both closed-loop and open-loop results are presented and discussed. However, as uncertain substitution rates differ across products from the adopted hydrometallurgical process, the open-loop recycling results are deemed more trustworthy at this stage. Due to word limitations the CED results and analysis are only briefly mentioned in this manuscript and we refer to the detailed results that are presented in the Supporting Information files.

In the absence of harmonized guidelines for the interpretation phase of the LCA (Laurent et al., 2020) uncertainties are highlighted here, and more a detailed sensitivity analysis is provided in the Supporting Information where the following alternative sets of assumptions are investigated in the: (i) homogenous metal recovery rates (i.e., the same metal recovery rates as for Sc-3 across all three scenarios); (ii) different sources for coke supply for Sc-3; and (iii) aluminum recovery in the upgrading stage for Sc-2. Once combined, these alternatives are referred to as "uncertainty studies 1–7" in Tables S1-6 and S1-7 in Supporting Information S1. Finally, break-even point (BEP) analyses were performed for using electricity (in

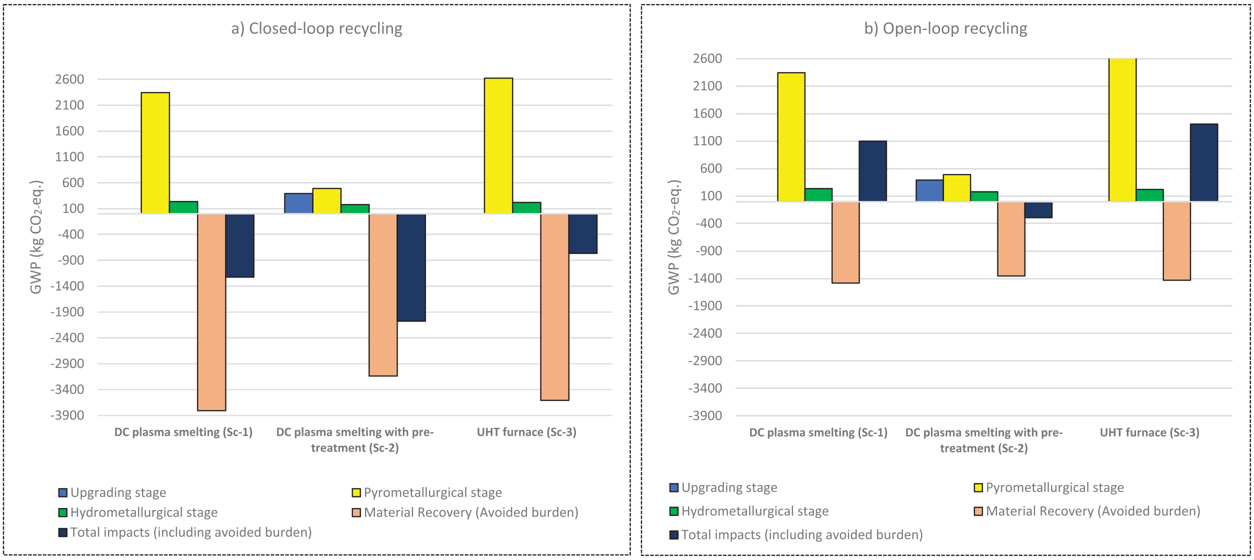

FIGURE 2 GWP of different stages for each scenario: (a) Closed-loop recycling, (b) Open-loop recycling. DC, direct current; UHT, ultra-high temperature
Note: The underlying data for this figure can be found in Supporting Information S3

the DC plasma arc furnace) instead of coke (in the UHT furnace)—that is, using Sc-1/Sc-2 versus Sc-3—as well as for employing the upgrading stage in combination with the plasma technology instead of using the common UHT furnace technology—that is, employing Sc-2 versus Sc-3 (see Chapter 5 in Supporting Information S1).

3.1 | Global warming potential

As illustrated in Figure 2, Sc-2 has the lowest total GWP, followed by Sc-1 and then Sc-3. In fact, shifting from Sc-3 as a common practice toward Sc-1 or Sc-2 could reduce GWP, irrespective of the recycling substitution assumptions. If closed-loop recycling is assumed, the reductions are 460 and 1310 kg CO_2-eq. per FU, respectively when shifting to Sc-1 and Sc-2. When instead assuming open-loop recycling, these values become 312 and 1,700 kg CO_2-eq. per FU, respectively. The larger GWP improvement for Sc-2 is mainly due to the significant reduction in the amount of input materials to the furnace (due to the upgrading stage). On the other hand, coke consumption as a reducing agent and a source of energy for the furnace in Sc-3 is a reason for comparatively worse environmental impacts of Sc-3. The results also reveal that extracting the valuable components with the highest quality for closed-loop recycling would bring significant GHG reductions. The significant reduction by closed-loop recycling is due to the fact that the recovered metal salts re-enter the LIB manufacturing chain with as little additional treatment as possible, while in the open-loop recycling the substitution of metals goes many stages back to the “raw” metal extraction (i.e., ore mining and beneficiation).

A further look at the results show that using electricity with the carbon intensity of 191 kg CO_2-eq. per MWh, that is, the British electricity mix (in Sc-1) instead of coke (in Sc-3), as a source of energy for the furnace, leads to a substantial reduction in GHG emissions from the furnace operation (−1250 kg CO_2-eq. per FU) as well as removing the upstream GHG emissions from coke production (−461 kg CO_2-eq. per FU). However, this needs a technological transition toward the DC plasma arc technology, which brings down the overall reduction in GWP from the pyrometallurgical stage to 11% (equal to −290 kg CO_2-eq. per FU), due to higher GHG emissions associated with upstream activities of input materials for the furnace operation as well as from the electricity used in Sc-1. A close look at the life cycle inventory of Sc-2 shows that less electricity and fewer input materials are needed compared to Sc-1, which results in greater environmental benefits compared to Sc-1. A further analysis showed that recovering aluminum through the upgrading stage in Sc-2 could have a major influence (Tables S1-6 and S1-7 in Supporting Information S1); therefore, uncertainty analysis was performed to quantify the aluminum recovery (see Section S4.3 in supporting Information S1).

The contribution analysis of different stages (Figure 2) shows that in all the scenarios, material recovery had the greatest role in reducing the overall GWP. Moreover, the GWP from the pyrometallurgical stage is significantly higher than that from the hydrometallurgical stage, due to impacts associated with the production of input materials as well as substantial GHG emissions from the furnace operation itself. For example, the upstream activities of material production (i.e., the production of input materials) in Sc-1 contributed to 45% of the total GWP from the pyrometallurgical stage (the total GWP is 2350 kg CO_2-eq.), followed by 39% from the furnace operation; in Sc-2, 41% of the total GWP from the pyrometallurgical stage (the total GWP is 490 kg CO_2-eq.) originating from the furnace operation (Figure 3a). In order to reduce the impact of pyrometallurgical processing, a reduction of materials containing carbon entering the furnace is essential. In another words, graphite, carbon black, plastics,

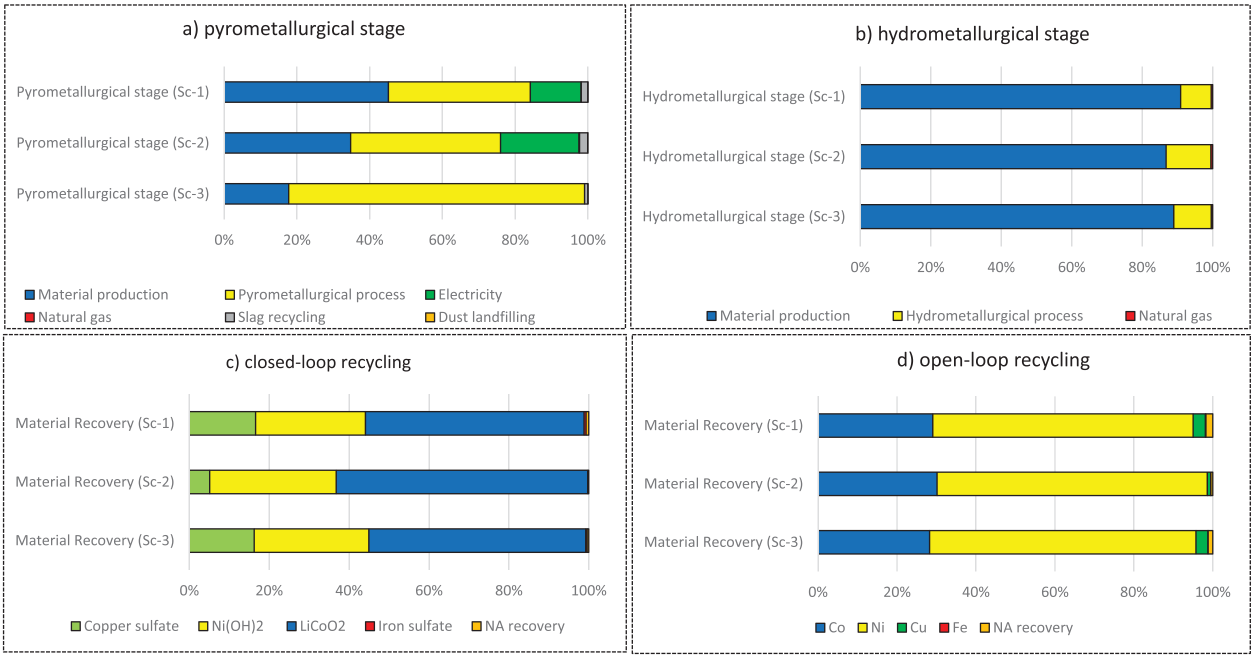

FIGURE 3 Contribution of different sources from total GWP (kg CO_2-eq.) in each major stage involved in each scenario. (a) Pyrometallurgical stage, (b) hydrometallurgical stage, (c) recovery stage assuming closed-loop recycling, and (d) recovery stage assuming open-loop recycling stage. It should be noted that GHG emissions from the pyrometallurgical process itself are higher than those from each input material. Here, aggregated amounts of GWP for the material production are illustrated for the sake of simplicity in illustration
Note: The underlying data for this figure can be found in Supporting Information S3

polyethylene terephthalate, polypropylene, polyethylene, ethylene carbonate, dimethyl carbonate, and PVDF are among the carbon containing materials that exist in EV batteries. This might not be easily possible for Sc-3 as these materials are burnt as a source of energy for the pyrometallurgical process (Hendrickson et al., 2015). However, alternative energy sources could make this happen for UHT furnace technology.

Upgrading is an opportunity to divert some carbon from the furnace and avoid fully burning it off. However, in the absence of these materials, the energy consumption of the furnace goes up. Hence, an additional source of energy is needed to increase the supply of energy to the furnace, that is, electricity in DC plasma technologies. In Sc-2, 27% of the carbon content in the feed material is burned-off before entering the furnace in the "removal of volatile compounds" stage (see Figure 1 for the stages involved in the upgrading stage). Then, a part of the remaining carbon containing materials (~51%) was assumed to end up in a landfill after the "separation stage," while ultimately 22% of the carbon content in the batteries ends-up in the furnace. Accordingly, the GWP from the pyrometallurgical stage reduced significantly (to almost one fifth compared to Sc-1 and Sc-3). However, based on the BEP analysis (See Chapter 5 in Supporting Information S1), if carbon separation does not happen and all the separated carbon is burned-off (i.e., ~450 kg additional GHG emissions), Sc-2 still has a large GHG reduction capacity (see also Table 1 and compare GHG emissions of Sc-2 with the others). The possible reduction in material consumption for the furnace operation is not considered herein as it is a pure metallurgical issue that is beyond the scope of the current study. However, possible optimization would be favorable to reduce GWP, especially a reduction in quicklime consumption in Sc-1 and 2, as well as a reduction in coke consumption in Sc-3. Moreover, use of different electricity mix, for example, using more renewable fuels can reduce GWP.

Figure 3b also shows that most of the impact (>86%) in the hydrometallurgical stage comes from material production, that is, the upstream activities of the material consumed as input. Accordingly, the upstream activities of Li_2CO_3 and H_2SO_4 consumption are major sources of GHG emissions within the hydrometallurgical stage in Sc-1 (>43% for Li_2CO_3 and >27% for H_2SO_4) and Sc-3 (>44% for Li_2CO_3 and >25% for H_2SO_4), while in Sc-2 the upstream activities of lithium carbonate (>55%) and hydrogen peroxide (H_2O_2) (>16%) consumption are the major contributors. The lower amount of alloy (due to lower amount of feed material sent to the furnace) in Sc-2 reduced the H_2SO_4 consumption and its associated upstream GHG emissions. Using organic acid leachants could reduce the secondary pollution (i.e., emissions from use phase such as toxic gas emission) caused by using inorganic ones without sacrificing the leaching efficiency (Li et al., 2018). However, raw materials for organic acids are critical factors (Gaines & Dunn, 2012) in choosing the least energy-intensive and environmentally low-impact acid for leaching. Alternatively, biomaterials for bioleaching could be investigated as they can reduce secondary pollution and leave non-hazardous material as residue that meets environmental limitations for safe disposal (Heydarian et al., 2018; Sommerville et al., 2021).

The contribution analysis also reveals that under closed-loop recycling assumptions, most of the avoided burdens due to metals recovery in all the scenarios come from $LiCoO_2$ followed by $Ni(OH)_2$ production (Figure 3c), while under open-loop recycling assumptions, avoided nickel production

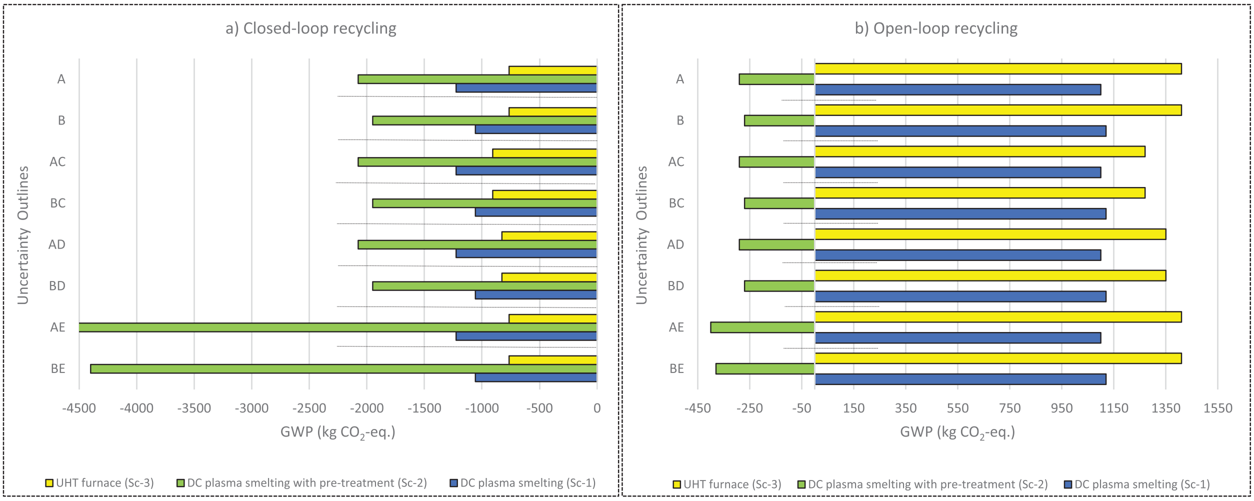

FIGURE 4 The overall Sensitivity results for GWP. (a) Scenario-specific recovery rates and coke from GLO supply (in Sc-3); (b) Homogenous recovery rates (same metal recovery efficiencies for all the scenarios) and coke from GLO supply (in Sc-3). AC, scenario-specific recovery rates and coke from Germany (in Sc-3); BC, homogenous recovery rates (same metal recovery efficiencies for all the scenarios) and coke from Germany (in Sc-3); AD, scenario-specific recovery rates and coke from USA (in Sc-3); BD, homogenous recovery rates (same efficiencies for all the scenarios) and coke from USA (in Sc-3); AE, scenario-specific recovery rates and coke from GLO supply (in Sc-3) and aluminium recovery (in Sc-2); BE, homogenous recovery rates (same metal recovery efficiencies for all the scenarios) and coke from GLO supply (in Sc-3) and aluminium recovery (in Sc-2). DC, direct current; UHT, ultra-high temperature
Note: The underlying data for this figure can be found in Supporting Information S3

("nickel mine operation, sulfidic ore'') followed by credits from avoided cobalt production ("cobalt production from reduction of grey and black cobalt oxide") are the main contributors (Figure 3d).

The sensitivity analysis (Chapter S4 in Supporting Information S1) depicts that the considered sources of uncertainty do not change the best option determined before (as Sc-2) except in a few cases for CED (see Figure 4 for GWP and Tables S1-6 and S1-7 in Supporting Information S1 for CED). The results also reveal that although GHG emissions in Sc-3 significantly depend on the source of the coke used, this scenario does exhibit the highest GWP (the lowest environmental performance) under different assumptions. Moreover, if both pyrometallurgical technologies achieve homogenous recovery rates, the GHG reduction does drop for Sc-1 and Sc-2 but this does not change the order of the scenarios and still Sc-3 and Sc-2 are the worst and best options, respectively. Aluminum (Al) recovery in Sc-2 could significantly increase the GHG reduction rate by up to 118–125% (closed-loop assumptions) and 37—40% (open-loop assumptions). This shows that recovering the Al components significantly helps to offset energy and GHG emission figures from the pyrometallurgical process. Although the Al content in the first generation NMC111 batteries considered in this study seems high (26.8% at module level, due to more inclusion of Al in cell housing and module periphery), the bills of materials for NMC622 and NMC811 batteries exhibit a 13.5% Al content at module level and a 24.5% Al content at pack level (Dai et al., 2018), which still could bring considerable benefits if recovered. Steel is another material in EV batteries that can bring some GHG benefits if recycled. For example, an analysis conducted herein using A2Mac1 bills of materials shows that Nissan Leaf Tekna 2018 and Nissan Leaf SV 2017 have around 27% steel at pack level (A2Mac1, 2019).

Considering that the new NMC generation batteries include more nickel content, the avoided GWP from metal recovery in open-loop recycling could be much more than for NMC111 (even if they contain less cobalt). This is due to significantly higher GHG emissions associated with "nickel mine operation," compared to the mining of the other recovered metals. The impact of different NMC compositions on GWP of different examined recycling scenarios were assessed, that is, a shift from the examined NMC111 chemistry herein to new and emerging NMC chemistries, that is, NMC532, NMC622, and NMC811 (Figure S1-9 in the supplementary material S1). NMC111 chemistry was used in BMW i3, Daimler EQC, Ford Focus-E, Hyundai Ioniq, Kia Soul, Smart ForFour and ForTwo, Volkswagen E-up!, and E-golf models (Baars et al., 2021), while NMC532 chemistry has already been used in the 2018 Nissan Leaf Tekna as well as the 2017 Geely Emgrand (A2Mac1, 2019). NMC622 is already being used in the Chevrolet Bolt, Opel Ampera-E, Renault Kangoo, and Zoe models (Baars et al., 2021). Envision AESC which is the cell provider for Nissan cars in the United Kingdom is also aiming to use NMC811 from 2020 (Envision AESC, 2020). This chemistry is also expected to be used in the new BMW i5 in 2021 (Ding 2019 et al., 2019). According to Figure S1-9 in Supporting Information S1, even when shifting toward new chemistries with lower cobalt contents, Sc-2 could outperform the others. This obviously highlights the importance of waste stream improvement through an upgrading stage.

Shifting toward new chemistries would lead to a moderate decrease in GHG reduction benefits of Sc-1 and Sc-2 under closed-loop recycling (except for using NMC532 for which reductions are more intense). This is the same for Sc-3 under closed-loop recycling, however, using NMC811 would increase the GHG reduction benefits of this scenario compared to the other chemistries. The situation would be different in open-loop recycling in which all the scenarios show a promising GHG reduction when shifting toward high nickel content chemistries.

The analysis of the environmental impacts of raw metal extraction for Co, Ni, Cu, and Fe has shown that in addition to GWP as a major concern, other impact categories are also critical, for example, eutrophication, ecotoxicity, ozone layer depletion, acidification, particulate matter, and human toxicity (Farjana et al., 2019). In fact, the impacts in some of these categories could even be considered to be relatively more severe than carbon emissions, for example, eutrophication in cobalt mining (Farjana et al., 2019). These impacts could be avoided when recovering materials through pyrometallurgical technologies. More interestingly, nickel mine operations show greater environmental impacts (per kg of Ni extracted) in all the above-mentioned impact categories compared to cobalt (per kg of Co extracted) and this could be promising for nickel-rich batteries. These environmental impacts could also be avoided when achieving high purity metal products in a closed-loop recycling further to the benefits arising from skipping material processing for the battery supply chain (Mistry et al., 2016). Therefore, recycling of LIBs through pyrometallurgical technologies could have wider impacts on the environment and thus considering the other impact categories is essential. However, this needs a more detailed LCI for pyrometallurgical and hydrometallurgical steps that could pass the completeness criteria for including the above-mentioned impact categories in the calculation by ensuring that all the emissions are considered, and thus biased conclusions are prevented.

Finally, the BEP analysis (see Chapter 5 in Supporting Information S1) showed a few points, first that when replacing coke with electricity in Sc-1, a careful consideration of the electricity grid is required since some of the resulting values may be lower than the carbon intensity of the electricity mix in certain countries. Such limitations are comparatively more severe when considering open-loop recycling. Second when considering a potential shift from Sc-3 to Sc-2, the results of the BEP analysis are more encouraging, since (i) the resulting GWP limits are significantly higher, and importantly, higher than those of the grid mix in most countries, and (ii) as discussed in Section S4.3 in Supporting Information S1, recovering aluminum in the preliminary upgrading stage could bring about further benefits (Al recovery was kept at zero across all options considered in BEP analysis). It is also worth mentioning that based on the BEP results (Chapter S5 and Table S1-16 in Supporting Information S1), alternative upgrading stages could generate even more carbon emissions than that of modeled in this study (up to 1430 kg CO_2-eq per 1 tonne of battery treated which is significantly higher than that of modeled in this study, that is, 392 kg CO_2-eq per 1 tonne of battery treated in Sc-2).

4 | CONCLUSIONS

Evaluating emerging technologies to address the carbon reduction efforts at global and local scale requires detailed studies to inform business and decision-makers. The use of electric vehicles is expected to grow exponentially in the coming years, and so will EoL LIBs albeit with a time lag of 10—15 years depending on the length of use in EVs and an eventual second life in, for example, energy storage applications. The expected material demand for EV batteries will thus be enormous and appropriate recycling technologies are crucial to make EV batteries sustainable. These technologies are still emerging although some are more mature than others. Environmental analyses are key to identify and guide promising technologies and inform business and decision-makers to guide investment decisions as well as incentives. LIBs from recycled battery materials would be a prime example of a circular economy (Baars et al., 2021) but achieving this at low environmental and economic cost is challenging and requires collaboration of LIB producers, consumers, and recyclers (Sommerville et al., 2021), as well as sustainability analysts.

This study conducted an LCA to compare two different pyrometallurgical technologies for the recovery of valuable metals from LIBs, a direct current (DC) plasma smelting technology (Sc-1 and Sc-2) and UHT furnace (Sc-3). Our research also assessed the influence of an upgrading stage (as a pre-treatment for LIBs) before sending the battery materials to the DC plasma arc furnace (Sc-2). Comparing the LCA findings with results reported in the literature is neither easy nor straightforward (Moni et al., 2020), due to the fact that a range of factors can differ across the studies: battery chemistries considered, modeling approach used for the pyrometallurgical and hydrometallurgical process, LCI data used and LCIA methods employed, and, perhaps most crucially, recycling assumptions. However, our study shows that shifting from UHT furnace technology (Sc-3) toward DC plasma arc technology (Sc-1 and Sc-2) can reduce the total GWP of the whole recycling process. At the same time, though, this technology shift could increase the CED per functional unit, except when shifting to Sc-2 under open-loop recycling.

In fact, possibly the most striking result of all is that, under open-loop recycling assumptions (which, as explained above, are probably the most realistic ones), only in Sc-2 do the carbon emission and energy credits afforded by the recovered metals overcompensate for the emissions and energy required to carry out the battery recycling process, resulting in negative overall values for both GWP and CED. On the contrary, both Sc-3 and Sc-1 result in a net positive GWP values under open-loop recycling, meaning that more GHGs are emitted overall when recycling LIBs than would be when sourcing the same "raw" metals from the respective primary supply chains. If on the one hand these could be taken as discouraging results, on the other hand, they point to two clear positive take-home messages: (i) the DC plasma arc technology, when coupled with the upgrading pre-treatment of the incoming LIBs (Sc-2), already provides a very robust improvement over the industry-standard UHT furnace technology (Sc-3), which also implies the importance of battery waste improvement to make recycling a game changer; and (ii) it is imperative that further technological development be directed at improving the follow-up hydrometallurgical process to achieve the best possible degree of "recycling loop closure," so as to enable the recovered metal salts to re-enter the LIB manufacturing chain with as little additional treatment as possible, thereby nudging the overall GWP and CED results of the whole recycling chain toward the "best case" results reported here under "closed-loop" assumptions. It is worth noting that other recycling alternatives to pyrometallurgical technologies like hydro or direct recycling might also be beneficial, but these require more investigation, specifically at industrial scale.

It is recognized that LIB recycling could be more beneficial in other impact categories which were not considered in this study, such as human and ecological toxicity, and abiotic resource depletion. It should also be noted that additional recycling benefits could accrue if some of the outer materials at the pack level, for example, casing, enclosures, are recovered through dismantling and separation prior to the modules entering the pyrometallurgical stage. For instance, the sensitivity analysis presented in this study shows that if the aluminum is removed from the battery modules and recovered in the upgrading stage (Sc-2), the GWP and CED would further decrease as a result of aluminum recovery (see Section S4.3 in Supporting Information S1). This diverts aluminum from the furnace which can make less useful output materials, such as alumina-based slag.

Overall, the results of this study shed new lights on the importance of the upgrading stage for the recycling process specifically. The research demonstrated that commercial upgrading processes that are economically profitable and achieve the efficient removal of many materials from LIBs prior to sending them to the furnace should be pursued. The research methodology, design, and application of the LCA study does inform future LCA studies of other emerging technologies that go beyond recycling in general, or EVs and LIBs specifically.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

ORCID

Mohammad Ali Rajaeifar https://orcid.org/0000-0002-0880-0614
Marco Raugei https://orcid.org/0000-0001-5026-8556
Bernhard Steubing https://orcid.org/0000-0002-1307-6376
Paul A. Anderson https://orcid.org/0000-0002-0613-7281
Oliver Heidrich https://orcid.org/0000-0002-6581-5572

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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