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RESEARCH AND ANALYSIS

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Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes

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Abstract

On the basis of a review of existing life cycle assessment studies on lithium-ion battery recycling, we parametrize process models of state-of-the-art pyrometallurgical and hydrometallurgical recycling, enabling their application to different cell chemistries, including beyond-lithium batteries such as sodium-ion batteries. These processes are used as benchmark for evaluating an advanced hydrometallurgical recycling process, which is modeled on the basis of primary data obtained from a recycling company, quantifying the potential reduction of environmental impacts that can be achieved by the recycling of different cell chemistries. Depending on the cell chemistry, recycling can reduce significantly the potential environmental impacts of battery production. The highest benefit is obtained via advanced hydrometallurgical treatment for lithium nickel manganese cobalt oxide and lithium nickel cobalt aluminum oxide-type batteries, mainly because of the recovery of cobalt and nickel. Especially under resource depletion aspects, recycling of these cells can reduce their impact to an extent that even leads to a lower "net impact" than that of cells made from majorly abundant and cheap materials like lithium iron phosphate, which shows a more favorable performance when recycling is disregarded. For these cells, recycling does not necessarily provide benefits but can rather cause additional environmental impacts. This indicates that maximum material recovery might not always be favorable under environmental aspects and that, especially for the final hydrometallurgical treatment, the process would need to be adapted to the specific cell chemistry, if one wants to obtain maximum environmental benefit.

KEYWORDS
battery recycling, environmental impact, industrial ecology, life cycle assessment (LCA), literature review, lithium-ion batteries (LIB)

1 | INTRODUCTION

The rapidly growing demand for batteries in automotive, stationary, and mobile applications leads to increasing amounts of returned waste batteries expected in future. Lithium-ion batteries (LIB) are the currently dominating and fastest developing battery technology, except for automotive starter batteries, where lead-acid is still dominating the sector (Pillot, 2017). However, concerns associated with resource availability and environmental impacts of LIB production represent a potential limiting factor for their future deployment (Vaalma, Buchholz, Weil, & Passerini, 2018; Weil, Ziemann, & Peters, 2018). For dealing with the expected waste battery streams and for minimizing environmental impacts associated with LIB production and the corresponding potential resource limitations, recycling of waste batteries is fundamental. On the other hand, recycling of LIBs is complex and associated with significant inputs of energy and/or chemicals, raising the question about its actual environmental net benefits. Numerous life cycle assessment (LCA) studies on manufacturing and use phase of LIBs have been carried out, but their end-of-life (EoL) phase

TABLE 1 List of abbreviations

Abbreviations	Meaning
ADP	Abiotic resource depletion potential (environmental impact category)
AP	Acidification potential (environmental impact category)
CED	Cumulative energy demand (environmental impact category)
EoL	End-of-life
FU	Functional unit (unit of comparison in LCA)
GHG	Greenhouse gas
GWP	Global warming potential (environmental impact category)
HTP	human toxicity potential (environmental impact category)
LCA	Life cycle assessment
LCO	Lithium cobalt oxide (LIB chemistry)
LFP	Lithium iron phosphate (LIB chemistry)
LIB	Lithium ion battery
LMO	Lithium manganese oxide (LIB chemistry)
LTO	Lithium titanate oxide (LIB chemistry)
NCA	Lithium nickel cobalt aluminum oxide
NMC	Lithium nickel manganese cobalt oxide
SIB	Sodium ion battery

is often disregarded (Emilsson & Dahllöf, 2019; Peters, Baumann, Zimmermann, Braun, & Weil, 2017). While this corresponds with the fact that the assembly and use phase are often the ones with the largest contribution over the whole life time of the battery, a proper recycling and the corresponding reduction in demand for virgin raw materials has the potential of reducing the total environmental impacts of the battery life cycle noteworthy and can even be decisive for the environmental competitiveness of a certain battery type (Ciez & Whitacre, 2019; Weber, Peters, Baumann, & Weil, 2018). However, existing LCA studies that include recycling often use rough estimations such as an unspecified mixed battery waste stream or unspecific recycling processes. First-hand life cycle inventory (LCI) data is rarely disclosed, impeding an adaption of the underlying process models to different or novel cell chemistries. Moreover, the focus lies mainly on environmental effectiveness of recycling processes or the difference between varying processes and not on the comparison of different cell chemistries and therefore, often only one or two cell chemistries are assessed. This limits the meaningfulness of these studies, since a specific assessment can be highly relevant for the choice of a certain battery type for a given application. Creating a cell-chemistry specific model for LIB recycling also enables the assessment of emerging or future battery types like, for example, sodium-ion or magnesium-ion batteries. This study presents a model of an advanced hydrometallurgical LIB recycling process for the application to different battery compositions, using primary data from industry. Based on an overview of existing literature, existing inventory data for established recycling processes are parametrized as well, allowing their flexible use for different battery chemistries, and thus serving as a benchmark for the newly developed process. With these, the potential environmental impacts or benefits of recycling different types of LIB and of a sodium-ion battery (SIB) are calculated and analyzed. See Table 1 for a full list of the abbreviations used in this article.

2 | METHODS

2.1 | Literature review

2.1.1 | Methodology

In order to identify all published studies that include quantitative data on the environmental impact of LIB recycling processes, a literature review is carried out in Google Scholar, Scopus, and ScienceDirect, using the search strings "LCA," "life cycle assessment," "environmental impact," "environmental assessment" in combination with "Li-Ion," "Lithium-Ion," "Lithium-Ion recycling," and "battery recycling." Only studies that provide quantitative results and that separately indicate the environmental impact of the recycling phase are considered. The underlying recycling process must be specified and results for the specific type of treatment must be indicated. Studies related to products in which LIBs are embedded, such as electric vehicles, are included if quantitative results for the recycling phase are given explicitly for the battery or battery cell.

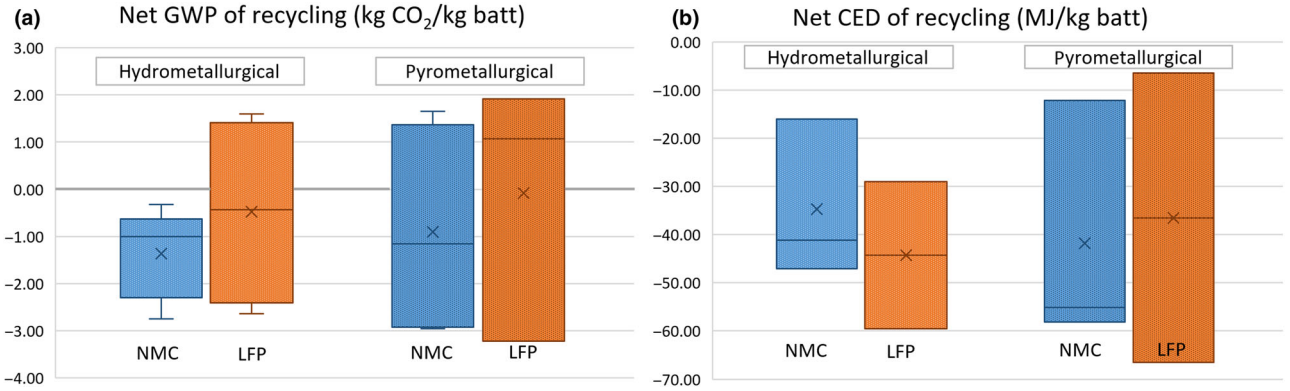

FIGURE 1 Range of global warming potential (GWP) and cumulative energy demand (CED) of battery recycling from literature: (a) GWP in kg CO_2-equivalents per kg of battery cell, (b) CED in MJ per kg of battery cell. Negative values indicate a net benefit of recycling, positive values additional impacts caused by recycling. Underlying data used to create this figure can be found in Table S2.6 in Supporting Information S2

2.1.2 | Review results

Key parameters of studies that fulfil these prerequisites are extracted and displayed in Table S3.1 in Supporting Information S3. Several studies provide results for a wide range of impact categories. However, for better overview and comparability, the impact categories shown are limited to the ones that are the most widely used within all studies (cumulative energy demand [CED], global warming potential [GWP], abiotic resource depletion potential [ADP], acidification potential [AP]). A total number of 27 studies that meet the defined criteria are identified, of which 16 explicitly express the impact of recycling as their analysis goal. The remaining 11 consider the whole life cycle of LIBs and include recycling as part of the life cycle. However, there are a few examples of these full LCA studies that show a very detailed analysis of the included recycling stage (Dunn, Gaines, Barnes, Sullivan, & Wang, 2014; Hawkins, Singh, Majeau-Bettez, & Strømman, 2013; Olofsson & Romare, 2013).

Among the studies that use own inventory data for the considered recycling processes, only seven disclose these data for verification and further use (Ciez & Whitacre, 2019; Dewulf et al., 2010; Dunn et al., 2014; Fisher, Wallén, & Paul, 2006; Hao, Qiao, Liu, & Zhao, 2017; Hawkins et al., 2013; Hendrickson, Kavvada, Shah, Sathre, & Scown, 2015). Dewulf et al. (2010) compare the cumulative energy requirements of virgin material for LIB production with that of recycled material and base production data for recycled material on obtained information from several Umicore facilities. Dunn et al. (2014) conduct a material and energy flow analysis for four different recycling processes, using, for example, average energy values for industrial process steps. The LCI by Fisher et al. (2006) is based on industry data (Recupyl, Batrec Industrie AG) and also represents the basis for Hischier, Classen, Lehmann, and Scharnhorst (2007) whose work is used for the respective processes in the widely used Ecoinvent database (Wernet et al., 2016). Hao et al. (2017) refer to industry data not further specified, while Hawkins et al. (2013) model their own process (shredding and cryogenic treatment) with auxiliary processes from Ecoinvent. Hendrickson et al. (2015) base their dataset on the Umicore Patent No. US 7,169,206 B2 (2007), and (Ciez & Whitacre, 2019) reuse data from previous studies (Dunn et al., 2014) and Argonne National Laboratory's GREET database (Dai, Kelly, Dunn, & Benavides, 2018). The remaining studies either use inventories from these studies or are based on own obtained industrial data, but do not disclose the inventories for further use.

As can be observed in Table S3.1 in Supporting Information S3, existing LCAs on LIB recycling use varying system boundaries and functional units (FU) like energy, driven distance, or recovered content, with either absolute or relative results. Moreover, the authors use different ways of quantifying the benefit, or generally the impact of recycling. Some indicate the total recycling impact, including process impacts and benefits of material recovery, while others do not account for credits of recovered material outputs and thus obtain environmental burdens for the recycling processes. At this point, the study by Ciez and Whitacre (2019) must be highlighted as they present results for three different cell chemistries (lithium nickel manganese cobalt oxide [NMC], lithium nickel cobalt aluminum oxide [NCA], lithium iron phosphate [LFP]) each in pyrometallurgical, hydrometallurgical, and direct recycling.

The very heterogeneous approaches impede the comparison of results for common existing LIB chemistries such as NMC, NCA, LFP, or lithium titanate oxide (LTO) across studies. Despite the difficulties in comparing the results, a few studies that are based on the same FU and system boundaries and assess the same impact categories can be contrasted. NMC and LFP in hydrometallurgical and pyrometallurgical recycling are identified to be the most frequently assessed combination, mostly assessed on a battery mass basis. Interestingly, this correlates only partially with the current relevance of LIB chemistries in terms of market share, where NMC is by far the most common chemistry, but followed by NCA (Panasonic/Tesla), while LFP and lithium maganese oxide (LMO) only play minor roles (van Sande, 2015). Figure 1 shows the range of results for GWP and CED obtained from the review (as far as results can be referred to a common unit of comparison, i.e., 1 kg of battery cells). It becomes obvious that, despite the identical FUs, the results still differ to a large extent. Especially in terms of GWP, the net benefit of pyrometallurgical recycling seems to be surprisingly small.

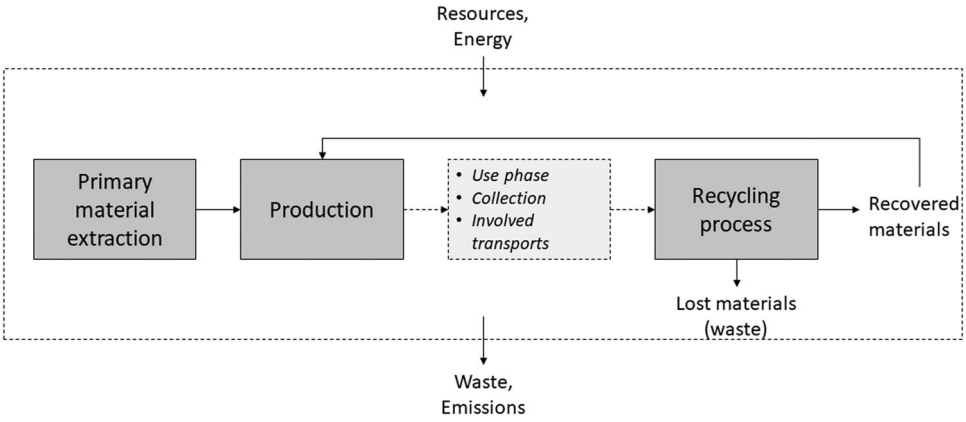

FIGURE 2 System boundaries

To analyze the different recycling results for different cell chemistries in more detail, and especially with a comparable scope and base, a recycling model is required that relates the recycling outputs to the individual cell composition. This would allow drawing more reliable conclusions on the environmental effectiveness of current and future battery recycling and the influence of different battery chemistries within.

2.2 | Life Cycle Assessment (LCA)

2.2.1 | LCA framework

The system boundaries established for the assessment are depicted schematically in Figure 2. They include cell production, the EoL phase (i.e., recycling processes including emissions and disposal of waste streams), and all corresponding upstream processes (i.e., mining and resource extraction and raw material provision, generation of energy and synthesis of required chemical agents and materials). The FU is defined as 1 kWh of storage capacity provided by the battery cell, calculated on the basis of given energy densities (Wh/kg) of the different cell chemistries (see Table S1.3 in Supporting Information S1). All batteries are assumed to be assembled in identical pouch cell housings. Due to the specific focus on the EoL phase, the use phase is excluded (depicted in grey in Figure 2), thus highlighting the potential of recycling for reducing the impacts associated with battery production. Excluding the use phase is not a common approach for LCA of batteries, since it disregards highly relevant parameters like efficiency or lifetime. However, it allows focusing more explicitly on the recycling and its potential benefits by evaluating the potential of recycling for mitigating the impact related with battery cell production, independently of the possible use of the battery. A full life cycle approach would add significant differences in terms of efficiency and lifetime. These are highly relevant parameters for the total life-cycle impact of batteries, but would conceal to a significant share the differences between the battery types in terms of recyclability. Also, the focus is on evaluating the potential performance of the different recycling pathways when treating different cell chemistries, and less on comparing cell chemistries in terms of net environmental impact.

Four different cell chemistries are compared: LIB types lithium nickel cobalt aluminum oxide (NCA), lithium nickel manganese cobalt oxide with stoichiometrically equivalent shares of Ni, Mn, and Co (NMC_{111}; in the following simply referred to as NMC), and lithium iron phosphate (LFP), plus the emerging technology SIB. The layered oxides NCA and NMC offer high working voltages and thus high energy densities which makes them favorable for the use in electric powertrains. Yet, they contain scarce metals like cobalt and nickel that are also bound to high prices. LFP, although offering a lower energy density, represents a cheaper and safer alternative, requiring no critical materials (Woehrle, 2018). SIBs are chosen as an example for future cell technologies. The compositions of the different LIB cells are retrieved from Peters and Weil (2018) and cross-checked against Dai et al. (2018). The SIB is based on the work by Peters, Buchholz, Passerini, and Weil (2016), but adjusted to the same cell layout like the LIB as proposed by Peters and Weil (2018). These inventory data are based on the most recent and most complete LCI data available currently. For the SIB, a general lack of detailed LCA considering different cathode active materials can be identified, and the one used here reflects the current state of the art in the field. However, due to the low technology readiness level of the SIB, their results are associated with major uncertainties, as typical for prospective assessments of not-yet established technologies. The corresponding bills of material are provided in Tables S1.1 and S2.2 in Supporting Information S1.

ILCD midpoint is applied as impact assessment method, considering two impact categories: global warming potential with a 100 year time perspective (GWP) and abiotic depletion potential (ADP; i.e, the depletion of mineral non-renewable resources) (EC-JRC 2011; Peters & Weil, 2016). Global warming, that is, climate change, is considered to be of special interest due to its high presence in public debate and due to the ongoing discussion about the energy intensity and greenhouse gas emissions associated with LIB production processes and correspondingly long amortization times (Emilsson & Dahllöf, 2019; Schrader, 2017). Resource depletion is the other scientific battlefield, with intensive discussions going on about the finiteness of strategic key materials for LIB, their outreach and corresponding limits of electrification (Vaalma et al., 2018; Weil et al., 2018).

Other environmental impacts, while potentially equally relevant, are much less within the focus of the debate (Peters et al., 2017). We therefore limit our discussion to these two impact categories for the sake of readability (considering all potential impact categories equally often ends up in lengthy discussions that are of little interest to the majority of the readers). However, the results for the remaining impact categories are calculated and provided fully in Supporting Information S2 to any interested reader. OpenLCA 1.7.4 in combination with underlying process data from Ecoinvent 3.4 (Wernet et al., 2016) is used as a software for the implementation and calculation of results.

2.2.2 | Inventory

Current industrial recycling processes for LIB cells usually involve either pyrometallurgical (high temperature) or hydrometallurgical (chemical) separation methods for the contained metals. Therefore, the current state of the art of LCA models for LIB recycling processes is represented by a pyrometallurgical process and a basic hydrometallurgical process based on secondary inventory data from Fisher et al. (2006). In order to consider future developments, first-hand data for an advanced hydrometallurgical process is obtained from industry (Duesenfeld GmbH, personal communication, July 2014). Hence, three different recycling processes are considered:

(i) Current pyrometallurgical treatment.
(ii) Current hydrometallurgical treatment.
(iii) Advanced hydrometallurgical treatment.

The datasets for (i) and (ii) in Fisher et al. (2006) also form the basis for respective processes in Ecoinvent (Hischier et al., 2007) but do not differentiate between different LIB cell types or chemistries, that is, only provide generic outcomes. Therefore, the provided inventory data is reviewed and adapted to distinct cell chemistries: The aggregated inventory for the *current pyrometallurgical process* (i) is based on LIB treatment by the company Batrec. The precise process flow is not disclosed, it is only known that the process involves a crushing step before neutralization and further processing (Fisher et al., 2006). Further details on the exact pyrometallurgical treatment are not revealed. A difference to hydrometallurgy is the loss of lithium to the slag during typical pyrometallurgical treatment. Moreover, components that can possibly be recycled in future, such as electrolyte, are burnt in the smelter (Rothermel, Winter, & Nowak, 2018). The inventory data as provided by Fisher et al. still includes several ambiguities. In known pyrometallurgical processes, such as the one by Umicore, lithium, aluminum, and all plastic parts are lost after thermal treatment. However, the dataset provided by Fisher et al. still contains plastic output. This may result from an initial disassembling step before the waste batteries enter the furnace. Moreover, the corresponding Ecoinvent dataset assumes that the non-ferrous metal output still contains aluminum which is then lost in the further treatment of non-Fe-Co-metals. Despite these inconsistencies, the pyrometallurgical process is not modified further, following the way it is modeled in the existing Ecoinvent dataset. A detailed description of the parametrization and example calculations are provided in Section 4.1. of the Supporting Information S1.

Aggregated process data for *current hydrometallurgy* (ii) originates from the Valibat process by the company Recupyl, representing their recycling activities in 2004. In this process, waste batteries are first shredded under inert gas and then chemically treated. A more detailed process description can be found in Ekberg and Petranikova (2015). Resulting process outputs are the metal constituents contained in the cathode material (lithium salts and respective other metals) as well as separated parts of the cell housing (aluminum, copper, and plastic). Supporting Information S1 (Section 4.2) provides details on the modeling procedure.

In addition to the product outputs of current hydrometallurgical processes, the *advanced hydrometallurgical process* (iii) from Duesenfeld GmbH includes the recovery of electrolyte and graphite at battery grade. A previous mechanical treatment comprises initial crushing, air classification, and sieving. The off-gas emerging from this step is cleaned via condensing and an activated carbon filter which needs to be replaced and reprocessed periodically. The subsequent hydrometallurgical treatment includes leaching, solvent extraction, and precipitation. Inventory data is available separately for mechanical and hydrometallurgical treatment. The company emphasizes that the data provided for their hydrometallurgy does not represent a suitable treatment for LFP or SIB cells. In the modeled process, the hydrometallurgical step is nevertheless applied to all cell types and the effect on LFP and SIB is particularly analyzed, see Supporting Information S1 (Section 4.3) for details on the underlying model.

For every dataset, the available inventory data is parametrized for cell-specific assessment. This is done based on recovery rates for different cell components that are retrieved from the given process data and the cell-chemistry specific compositions of the respective waste batteries. Recovered material from recycling is considered avoided primary material and thus gives environmental credits that are rested from the overall impact, assuming closed-loop recycling. Thus, the recovered product quality must comply with battery grade requirements and for each battery input, the recovered material is qualitatively equivalent to the virgin material needed to produce the respective battery cell. For the implementation in OpenLCA, the datasets are modeled with the quantitative reference of 1 kg treated battery cells to provide datasets for broader applications. The results of the calculations are finally adjusted according to the above introduced energy densities to obtain results for the defined FU of 1 kWh. Figure 3 shows the three process flows, indicating all considered recovered products. A detailed description of the parametrization approach, underlying assumptions for recovered material, and resulting LCI tables for all three processes are provided as MS-Excel sheet in Supporting Information S2.

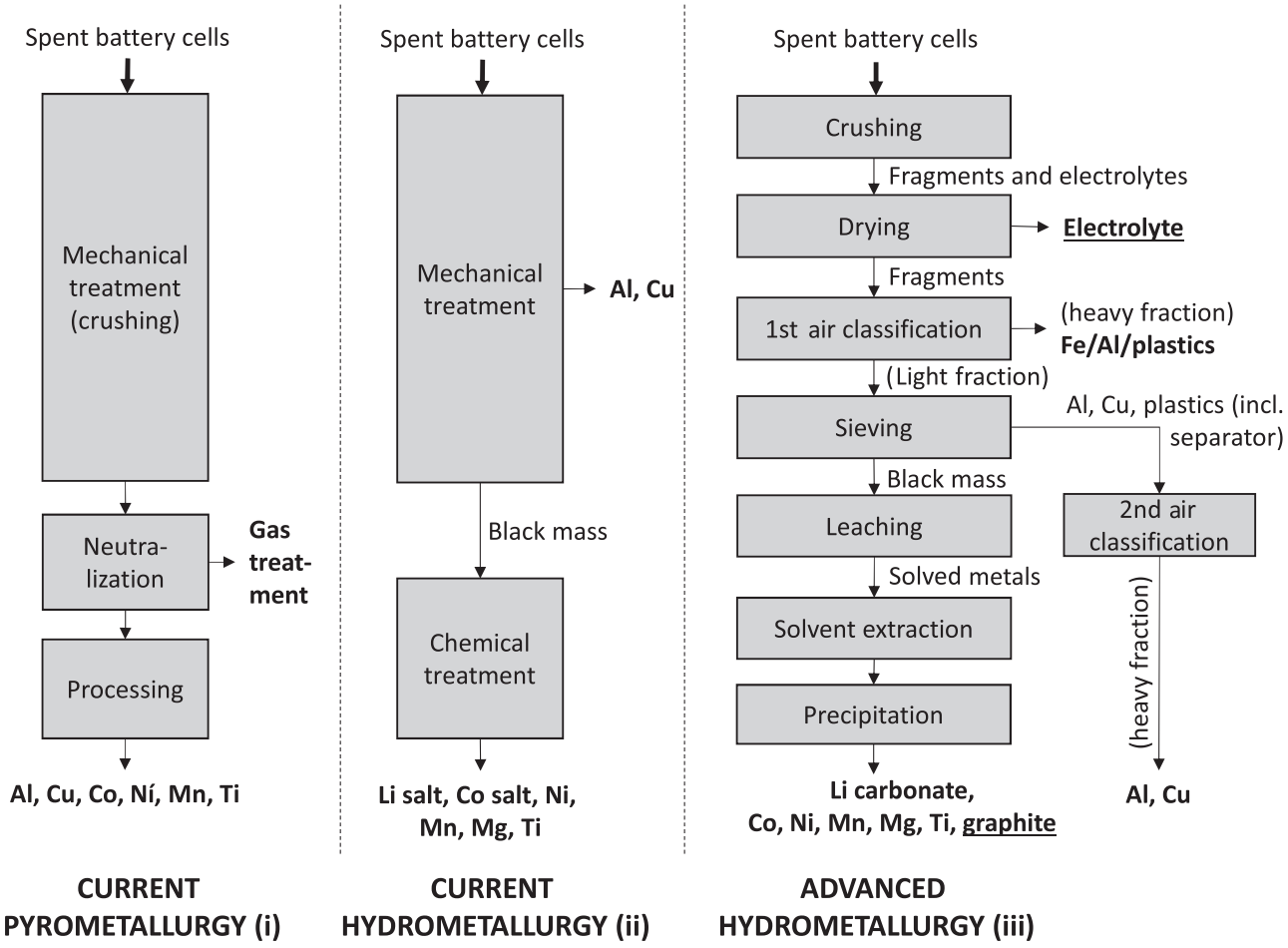

FIGURE 3 Process flows, including all considered product outputs. Inputs, waste and emissions not displayed (Diekmann et al., 2017; Duesenfeld GmbH, 2014; Ekberg & Petranikova, 2015; Fisher et al., 2006)

3 | RESULTS

Production. As a basis for assessing the impacts of recycling, Figure 4 shows the impacts of production for each battery type broken down to cell components. NMC production causes the lowest GWP per 1 kWh capacity (75.50 kg CO_2-Eq), followed by NCA, LFP, and SIB. Here, a high cell energy density plays an important role, since less battery is required for providing a certain capacity. The cell manufacturing energy (electricity and heat) represents the largest part of the production GWPs for each cell chemistry. This is important to note since recycling cannot retrospectively reduce GWP caused by the manufacturing energy demand. Cathode material is responsible for another significant share of the total GWP of NCA and NMC cells, while anode material makes up for the second highest GWP share for SIB production (the anode is made of hard carbon for this battery type, unlike graphite for the LIBs). Regarding the ADP, cathode material (especially when containing cobalt and nickel) and anode current collector (copper) influence the total impacts significantly which is why NCA and NMC production show the highest ADP (0.24 and 0.35 kg Sb-Eq/kWh). LFP and SIB rely on less critical materials, namely iron phosphate and sodium, and the SIB also avoids copper for the anode current collector (Peters et al., 2016).

Recycling. Figure 5 presents the impacts and benefits of the different battery recycling processes for the considered cell chemistries. Impacts are caused majorly by the required process inputs and emissions (e.g., energy and chemicals, off-gases), while the benefits are obtained from the recovered materials, avoiding primary materials. Resting the process impact (positive value in the diagram) from the credit for recovered materials on the negative side gives the environmental benefit for all assessed cell chemistries in both impact categories.

The highest recycling credits are obtained for those cell components that also cause high impacts during primary production. For the considered cells, these materials are copper, nickel, cobalt, and, under GWP aspects, also aluminum. Lithium also plays a minor role. Therefore, the regarded recycling processes offer the highest impact reduction potential for NCA and NMC cells, while especially for LFP cells the possible recycling benefit is significantly lower. The advanced hydrometallurgical process shows the highest benefit in all cases due to the additionally recovered graphite and electrolyte. However, also the process inputs and thus the corresponding impacts increase, especially under ADP aspects.

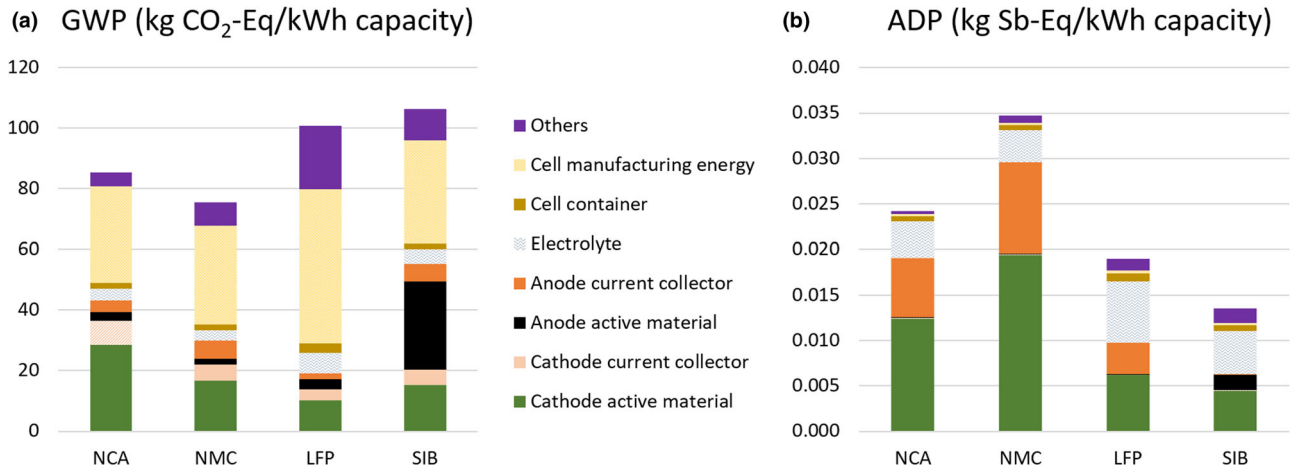

FIGURE 4 Environmental impacts of cell production, broken down to cell components: (a) Global warming potential (GWP) in kg CO_2-equivalents per kWh storage capacity, (b) abiotic resource depletion potential (ADP) in kg Sb equivalents per kWh storage capacity. Underlying data for this figure can be found in Table S2.7 in Supporting Information S2

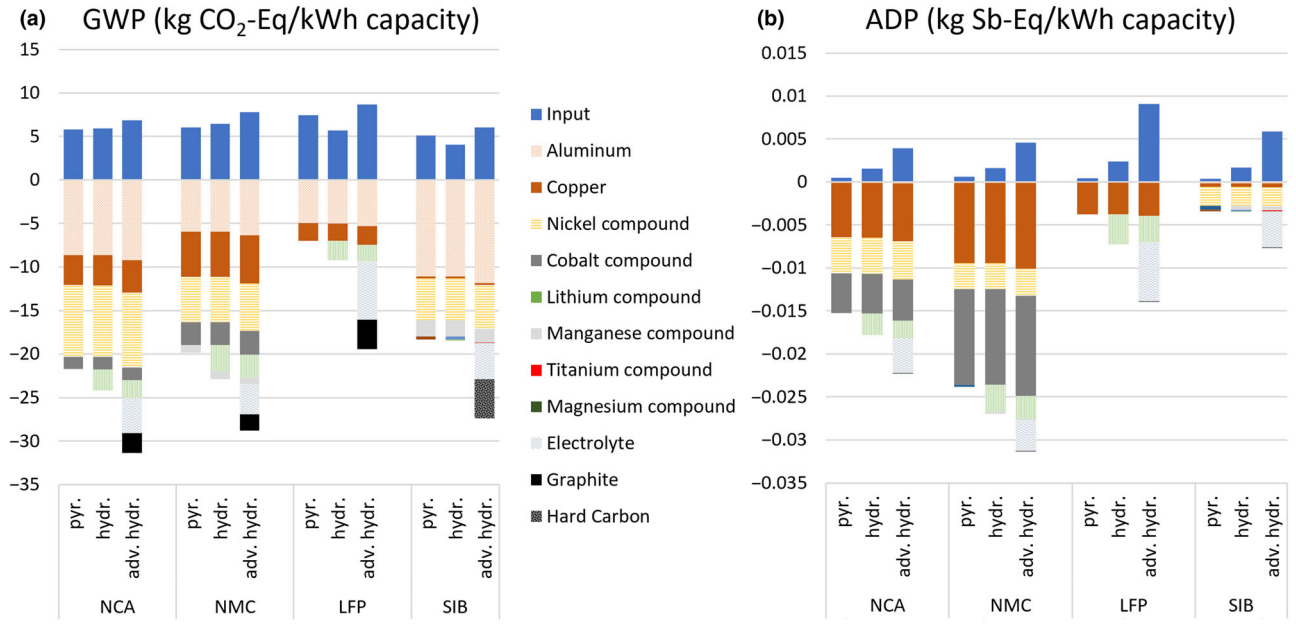

FIGURE 5 Environmental benefits of battery cell recycling, broken down to the contribution of the different fractions recovered by the recycling processes: (a) global warming potential (GWP), (b) abiotic resource depletion potential (ADP). Negative values indicate net benefits (reduction of impacts due to recovered materials), positive values are environmental impacts (due to inputs for the recycling process). Data used for creating this figure can be found in Table S2.8 in Supporting Information S2

Net impact. Figure 6 shows the net impacts obtained for the different recycling processes and cell chemistry, that is, the final impact after subtracting the recycling benefits from the production impacts. For the advanced hydrometallurgical recycling process, the benefit is further broken down into mechanical and hydrometallurgical treatment. Under GWP aspects, the advanced hydrometallurgical recycling shows the best result in all cases, reducing the impacts of the batteries by between 12% and 25% (in comparison to no recycling). For ADP, the same tendency can be observed for the high-energy LIB NCA and NCM, while for the LFP cells and the SIB, the hydrometallurgical treatment does not obtain further benefits, but rather adds burden, as can be observed in Figure 6 (contribution of hydrometallurgical treatment on the positive side, thus increasing impacts instead of reducing them). The comparably high process inputs for this step and the low benefit from the recovered materials make this process unfavorable for recycling these cell chemistries.

Under GWP aspects, recycling generally shows a lower relative reduction potential than for ADP. This outcome is partially influenced by the high share that cell manufacturing contributes to the total GWP, which cannot be reduced by recycling. NCA and NMC cells show the lowest production impacts and thus also lowest net GWP. This is due to their higher energy densities compared to LFP and SIB; less cell mass needs to be produced

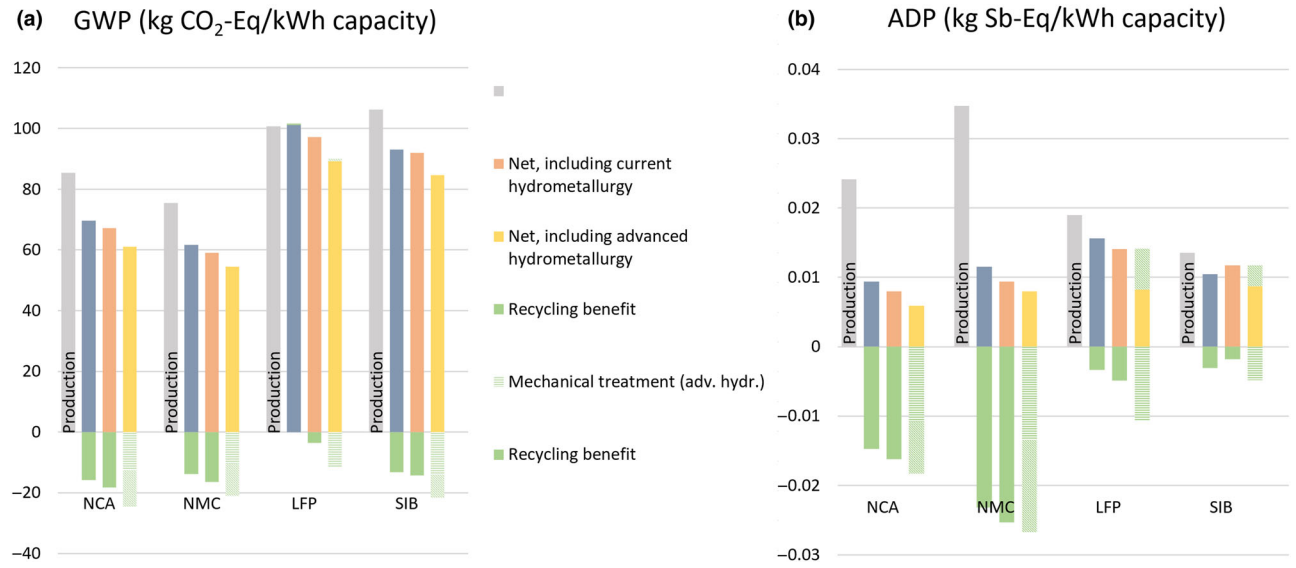

FIGURE 6 Production and "net" environmental impacts after subtracting recycling benefits obtained by the different recycling processes: (a) global warming potential (GWP), (b) abiotic resource depletion potential (ADP). Recycling benefits are negative (green bars), indicating reduction of impacts. For advanced hydrometallurgy, benefits are divided into mechanical and hydrometallurgical treatment (hatched areas). Underlying data for this figure can be found in Tables S2.9 and S2.10 in Supporting Information S2

for providing the same capacity and thus, less production energy is required. Pyrometallurgical recycling shows lower benefits and thus higher net impacts when compared to the current hydrometallurgical process, mainly due to the higher energy consumption of the high temperature processing and the loss of lithium in the slag. Regarding the LFP cell, pyrometallurgical treatment even adds GWP, requiring significant process inputs for recovering a comparable small share of the contained materials. The additional recovery of electrolyte and graphite contribute to the lower net impacts of the advanced hydrometallurgical process, despite the highest process inputs. Depending on the recycling process, relative impact reduction potentials for NCA and NMC cells range between 18% and 29% leading to net impacts between 69.58 and 61.00 kg CO_2-Eq (NCA), respectively, 61.69 and 54.45 kg CO_2-Eq (NMC).

The cells' net ADP is highly influenced by their recyclability, and the results strongly depend on the specific cell chemistries. NCA and NMC cells benefit greatly from recycling, resulting in a significantly lower net impact. Overall, LFP and SIB score worse, despite lower initial ADP impacts from their production phase. In advanced hydrometallurgy, especially the recovery of electrolyte adds to a further reduced net impact. It becomes clear that hydrometallurgy (processing of the black mass) for LFP and SIB in the advanced process adds environmental burden. This underlines the company's previous statement that the hydrometallurgy represented in the dataset is not adapted to LFP or SIB type cells. The displayed net impact of LFP and SIB therefore only considers the benefits of mechanical treatment—and is significantly lower than with current hydrometallurgical recycling. Net impacts might still be reduced if adding an adapted, cell-specific hydrometallurgy with, for example, lower input of chemicals. However, possible benefits of additionally recovered cell components must be balanced with environmental impacts of the additional process efforts. Depending on the recycling method, the ADP of NCA cells is reduced by 61–76% to a net impact between 0.0059 and 0.0094 kg Sb-Eq (NMC: 67–77% reduction potential, 0.0080–0.0115 kg Sb-Eq net impact). In advanced hydrometallurgical recycling, LFP and SIB cells reach reduction potentials of 57% and 35%, respectively, reducing their ADP to 0.0083 (0.0088) kg Sb-Eq/kWh.

4 | UNCERTAINTY AND SENSITIVITY

When interpreting the results presented above, the associated uncertainties need to be considered. These are significant and attributable to the numerous assumptions and simplifications in the process and assessment model and the often insufficient information about inputs and emissions. Also, battery chemistries are developing, and low cobalt NMC cathodes (NMC_{622} and NMC_{811} are already on the rise, expected to substitute NMC_{111} in near future (Shobana, 2019). Energy densities are expected to increase further while the energy and GHG intensity of the cell manufacturing process is decreasing (Emilsson & Dahllöf, 2019). While the available data is insufficient as to determine meaningful value ranges for an extensive uncertainty analysis, the influence of these factors on the recycling benefits are evaluated in the following via a sensitivity analysis.

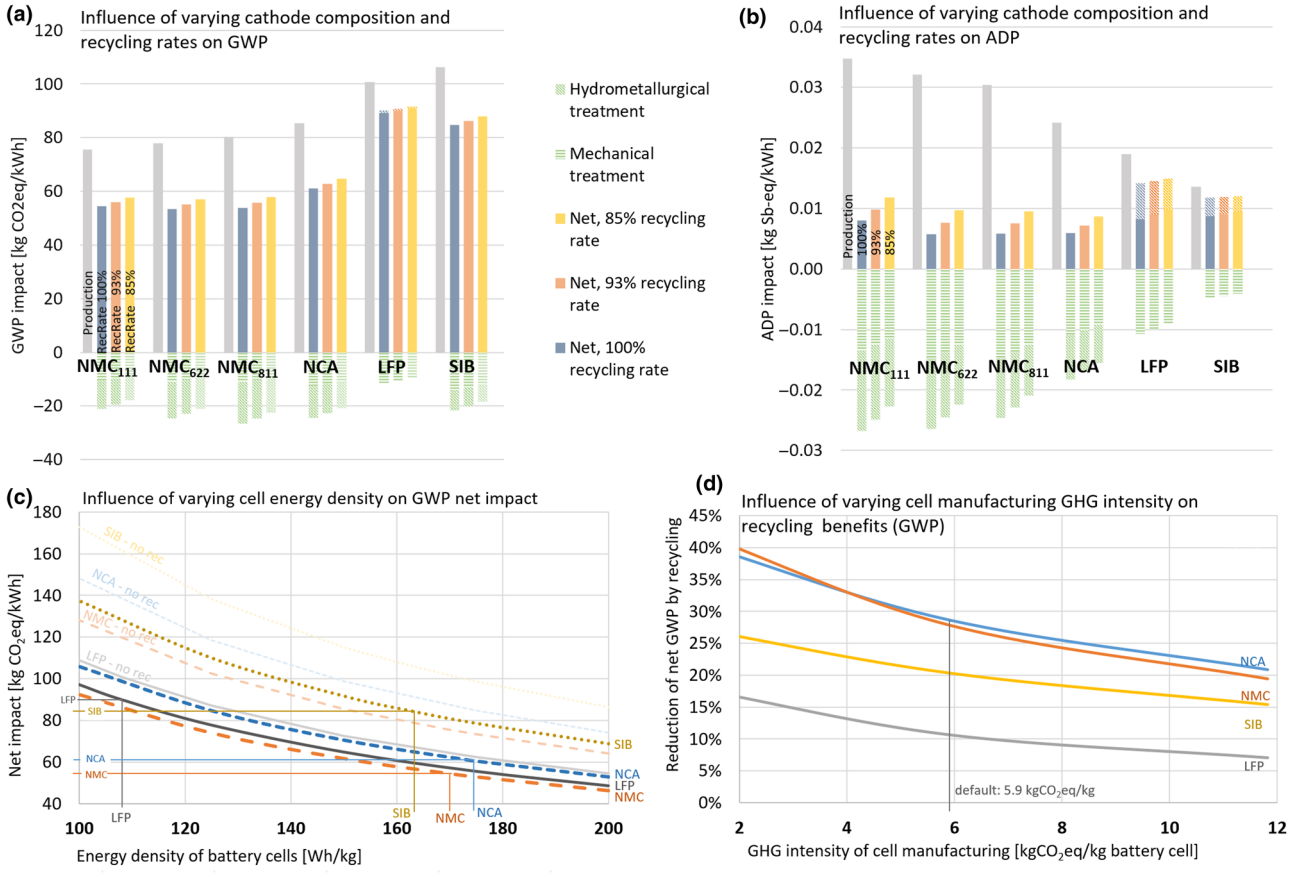

FIGURE 7 Sensitivity analysis. (a) Influence of varying cathode compositions and recycling efficiencies on GWP. (b) Influence of varying cathode compositions and recycling efficiencies on ADP. (c) Influence of varying cell energy density on net GWP impact, with default energy densities indicated by fine solid lines, net impacts of cell production including recycling by the dark colored lines (dotted yellow for SIB, solid grey for LFP, dashed blue for NCA, and long dashed orange for NMC), and corresponding gross impact (without recycling benefit in light color). (d) Influence of varying cell manufacturing greenhouse gas intensity on the GWP benefit achievable by battery recycling (lower right). The underlying data is provided in Tables S2.13–S2.16 in Supporting Information S2

4.1 | Recovery rate and recycled material quality

Although a closed-loop recycling is assumed in the assessment, recycled materials do not always fulfil the quality requirements for battery production (Peters, Baumann, & Weil, 2018). Evaluating this aspect comprehensively would require expanding system boundaries and allowing the recovered materials to replace primary ones in other sectors (like, e.g., construction or packaging), while maintaining a minimum share of primary material in the battery production inputs. However, assuming that secondary material is assimilated by the market (not discarded) and recycled material does not displace recycling elsewhere, such an expanded scope would obtain practically the same results, being the same amount of primary material displaced simply elsewhere. On the other hand, recycling efficiency is highly relevant, and varying it can also be regarded as a proxy for the variation of recycled product quality, assuming that the recovered products only correspond to primary material quality up to a certain percentage. Figure 7 (top) provides the net benefits obtained for the advanced hydrometallurgical treatment (iii) with varying recycling efficiencies; 100% (the base case (Duesenfeld GmbH, personal communication, July 2014)), 93% (as assumed by Fisher et al.), and a comparatively low 85%. For dropping recovery efficiencies, the mechanical recycling of LFP and SIB becomes more favorable, these cells being (due to the lower production impacts) less sensitive on high recovery rates. As a consequence, high recovery efficiency is especially important for cell chemistries that contain higher amounts of scarce metals like copper, nickel, and cobalt and thus cause higher production impacts.

4.2 | Low cobalt cell chemistries and electrolyte recovery

The results for the advanced hydrometallurgical recycling (iii) of different NMC cathode materials (NMC_{111} with stoichiometric relation 1:1:1of nickel, manganese, and cobalt, NMC_{622} with 6:2:2, and NMC_{811} with low cobalt content and stoichiometric relation 8:1:1). Interestingly, in terms of GWP, there is no significant difference between the NMC chemistries, leading increased nickel contents to higher production impacts, which are then offset by the recycling benefits (higher amounts of recovered nickel). ADP shows a contrary trend, lower impacts for high-nickel cathodes, but

correspondingly lower recycling benefits. In sum, the net impacts do not change significantly with varying NMC composition, assuming identical energy density (the influence of increasing energy density is treated in the following section).

4.3 | GHG intensity of battery manufacturing

Since the impacts of manufacturing make up the share of battery production impacts that cannot be compensated by recycling, it is worth looking into the impact of improved production process on the benefits of recycling in terms of GHG emissions. Figure 7 (bottom right) shows the relative benefit obtained from advanced hydrometallurgical recycling for varying GHG intensity of cell manufacturing. The GHG benefit achievable from recycling clearly increases when the contribution of the manufacturing process to the total production impacts drops. Still, even then more than half of the GHG emissions from cell manufacturing cannot be offset by recycling, marking a certain limit in terms of GHG benefits of recycling. For SIB and LFP, the maximum benefit is significantly lower, also because the recycling process is not optimized for their specific cell composition.

4.4 | Battery energy density

The use phase of the batteries is explicitly excluded from the assessment. However, the energy density is a performance parameter that affects the results strongly, being the recycling impacts and benefits calculated on a per-kWh basis. Figure 7 (bottom left) displays the influence of varying cell energy density on the net GWP impact obtained for the different battery cells including recycling (dark colored lines) and the corresponding gross impacts without recycling (light colored lines). The relevance of energy density for the net impact is clearly observable. If all cells had the same energy density, LFP would be situated in between NMC and NCA cells, despite the comparable low benefit obtained from recycling, while SIB would score significantly worse.

5 | DISCUSSION

The results of the present cell-chemistry specific assessment indicate highest recycling benefits and lowest net impacts for NCM-type LIB under GWP aspects, while for ADP, NCA cells obtain the best results. Benefits for LFB and SIB cells are comparably low in both considered impact categories, and hydrometallurgical treatment of the black mass without a specifically tailored process potentially even increases environmental burdens. This is in line with the findings by Ciez and Whitacre (2019), who assess the GWP of NCA, NMC, and LFP pouch cells on a mass basis. In their study, NMC cells show the highest GWP reduction, while in the present work NCA cells perform slightly better. This can be attributed to varying assumptions regarding the cell compositions and different underlying inventory databases, but also different energy densities. In both studies, LFP cells show the least benefit (respectively the highest added burden) in both of the two assessed recycling processes. However, in their work pyrometallurgical recycling adds burden for all three cell chemistries, while in the present study it performs unfavorable only for LFP cells.

When comparing the results with those by Buchert, Jenseit, Merz, and Schüler (2011) and Buchert and Sutter (2015a, 2015b), who provide results for NMC and LFP type cells on a mass basis, it is noteworthy that they obtain a higher GWP reduction for the recycling of LFP than for NMC cells. A closer look, however, reveals that the difference results from the high benefit from the battery pack disassembling step (recovered steel and aluminum from the housing, which contributes a higher relative share for low energy density batteries) which is excluded in the present study focusing on single cells.

Dunn, Gaines, Kelly, James, and Gallagher (2015) also provide results for different recycling routes of different cathode materials, and indicate a relative GWP reduction potential of recycling. Their considered intermediate process (comparable to a mechanical–hydrometallurgical route) shows 20% GWP reduction for NMC and 18% for LFP recycling. The current hydrometallurgical process in the present study process offers roughly 22% GWP reduction potential for NMCs and 3.5% for LFPs. However, these results can hardly be compared since Dunn et al. (2015) only consider cathode material manufacturing and not cell production. Cell manufacturing energy, which makes up large parts of the cell production impact, is not taken into account. Moreover, the modeling of the production phase also plays a significant role for the outcome since the authors refer to the solid-state preparation technique of the cathode material.

In any case, it has to be pointed out that the general lack of detailed inventory data for the recycling processes in combination with a cell chemistry specific assessment makes a comparison with previous works difficult. This highlights the relevance of a transparent and parametrizable inventory for the different recycling processes as provided in the present work. Furthermore, the specific system boundaries defined for the present assessment have to be taken into account. We explicitly excluded the use phase of the batteries in order to sharpen the contrast between production and recycling phase and thus the potential reduction of production impacts achievable by the different recycling processes. This provides an intuitive picture of the potentials and limitations of the assessed recycling process for the different battery types, but is no complete LCA and can therefore not provide any information or recommendation regarding the environmental impact of any of the assessed battery types. For this purpose, a full life cycle perspective would be required, considering also charge–discharge efficiency and lifetime, which can vary substantially between different batteries. This, in turn, would require defining representative application cases, since lifetime and efficiency

depend on load profiles and cycle depths required by the specific application (Baumann, Peters, Weil, & Grunwald, 2017). Being such an analysis out of the scope of the present work, the focus on production versus recycling impacts is considered the most intuitive one, allowing the inventory data provided in Supporting Information an easy adaptation of the recycling models for future, more application-oriented assessments.

6 | CONCLUSION

This work provides a cell-specific assessment of LIB and post-LIB cell recycling by means of different recycling processes based partly on original primary data. Two main findings can be pointed out in this regard.

First, recycling can lead to a significant reduction of the potential environmental impacts associated with the production of LIB cells and is therefore crucial for a future LIB industry. However, the environmental benefit depends strongly on the processed cell chemistry, but also on the considered impact category. Particularly when regarding the resource depletion potential of NMC and NCA cells, closed-loop recycling can reduce their impact to an extent that even leads to a lower net impact than that of LFP or sodum-ion batteries (SIB), which show a more favorable performance in their production phase. However, the benefits highly depend on the cell composition and layout, but also their energy densities (as the assessment is done on a capacity basis, i.e., per kWh). The results are thus only valid for specific battery cells as modeled in this work. For other cell layouts and energy densities, results may vary, as pointed out in the sensitivity analysis. The extent of the recycling benefit also depends on which impact category is considered. Resource depletion benefits (ADP) are much higher than those for greenhouse gas emissions (GWP), since recycling can only reduce impacts from mining, not from cell manufacturing or active material. The latter are also major contributors to the GWP of LIB cells, and the potential for reducing GWP of LIB by recycling is therefore limited. However, recycling benefits for GWP are likely to increase in future with a growing share of green electricity used in the manufacturing process or, generally, less energy intensive LIB cell production (Emilsson & Dahllöf, 2019; Peters et al., 2017). Other impact categories are not discussed further for the sake of comprehensiveness but might be equally relevant and can be reduced substantially by the assessed recycling processes. The corresponding results are provided in Supporting Information (Table S2.8 in Supporting Information S2 and the corresponding figures).

Second, due to substantial differences between the compositions of battery cells, a cell chemistry specific approach for recycling is necessary. Recycling processes need to be tailored and adapted to different cell chemistries in order to obtain the best possible output quality and highest environmental benefit. Otherwise, recycling efforts might outweigh the benefits obtained from the recovered materials to a large extent, or even cause additional impacts, as pointed out for hydrometallurgical treatment of LFP and SIB cells. For these cells, maximum recycling depth does not automatically mean maximum environmental benefit. This is especially relevant in future, with the development of new battery technologies going toward minimizing the share of expensive metals like cobalt or toward using less scarce and environmentally critical materials such as, for example, phosphorus or sodium. It is thus of great importance to enable a reliable identification of cell chemistries, for example, by labeling the cells, allowing recyclers to tailor the recycling processes cell chemistry specifically for minimum input of chemicals and process energy. Direct recycling that recovers cathode material maintaining its crystal structure is often mentioned as an option for low-cost cells such as LFP, offering significantly higher potential benefits (Dunn, Gaines, Sullivan, & Wang, 2012). However, this method is not commercial yet and there are still quality concerns regarding the recovered cathode material (Peters et al., 2018).

Also, the limitations of the present study must be taken into account. The results are bound to uncertainties due to often weak or insufficiently modeled background data. While the advanced hydrometallurgical process is based on primary data, the other two processes are adapted from the literature, containing several ambiguities and modeling inconsistencies. Disclosing more detailed process flows and cell-chemistry specific input of chemicals and energy for the different recycling processes could thus be targeted by future works, helping to improve data quality and thus the robustness of the results.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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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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