JOURNAL OF
BUSINESS CHEMISTRY

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

Research Paper

Luca Stegemann[a*], Moritz Gutsch[a]

Environmental Impacts of Pyro- and Hydrometallurgical Recycling for Lithium-Ion Batteries - A Review

The growing demand for lithium-ion batteries (LIB) leads to an increasing importance of battery recycling. Given the scarcity of resources, effective material recovery is essential for the sustainable production of batteries. Therefore, understanding the environmental impacts of different recycling approaches is crucial. This review is the first to provide a comparative analysis of the environmental impacts across various impact categories for both pyro- and hydrometallurgical recycling processes of nickel-cobalt-manganese (NMC) batteries, including consideration of different data sources and their influence on the results. For global warming potential (GWP), hydrometallurgical recycling achieves an average reduction of - 25.5 kg CO2eq kWh^{-1}, corresponding to a 39% decrease in emissions from battery cell production. In comparison, combined pyro- and hydrometallurgical recycling reduces emissions by 27%. Additionally, the hydrometallurgical method demonstrates greater sustainability in terms of cumulative energy demand (CED), lowering the energy required for battery cell production by nearly 17%. Other environmental categories besides GWP and CED receive significantly less attention in the literature, although the benefits are often more significant. To guide future research, we present three key recommendations for further exploring the environmental impacts of battery recycling.

Keywords: Lithium-ion battery, Battery recycling, Pyrometallurgy, Hydrometallurgy, Environmental impact, Life cycle assessment

Introduction

Driven by growing interest in electric vehicles, portable electronics, and renewable energy storage systems, global annual demand for lithium-ion batteries (LIB) exceeded 1 TWh for the first time in 2023 and is expected to reach around 3 TWh by 2030 (Bürklin et al., 2022; Hettesheimer et al., 2023). At the same time, recycling of used LIBs is increasingly crucial, as improper disposal will cause environmental and safety problems threatening the ecological environment and human health (Islam and Iyer-Raniga, 2022; Zhenghe et al., 2022). Also, recycling helps to preserve critical materials, such as lithium, cobalt, nickel and manganese, which can contribute to a reduction of

[a] Institute of Business Administration at the Department of Chemistry and Pharmacy, University of Münster, Leonardo Campus 1, Münster 48149, Germany.
* Corresponding author (luca.stegemann@uni-muenster.de)

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

the environmental impact associated with battery cell production (Islam and Iyer-Raniga, 2022). On the other hand, recycling of LIBs involves complex processes that require substantial energy and chemical inputs, leading to questions about the overall net environmental benefits of the different recycling methods (Kim et al., 2021; Mohr et al., 2020).

Currently, three main recycling methods are used for LIBs: pyrometallurgical, hydrometallurgical and direct recycling (He et al., 2024). While hydrometallurgy and pyrometallurgy are already employed on industrial scale, direct recycling processes are at lower technological readiness (Davis and Demopoulos, 2023; van Hoof et al., 2023; Xu et al., 2023). In pyrometallurgical recycling, high-temperature processes are used to recover valuable metals from spent LIBs. The process is energy-intensive, can cause harmful emissions, and valuable metals, such as lithium, are not recovered (Gaines, 2014; van Hoof et al., 2023). The hydrometallurgical process involves the use of chemical reagents for the dissolution of valuable metals in aqueous solutions (Wang et al., 2022). This allows manganese and graphite, among other materials, to be recovered (Brückner et al., 2020). Direct recycling separates different battery active substances through physical processes, such as gravity separation and flotation (Jung et al., 2021). The principal distinction between direct recycling and pyro- and hydrometallurgical processes is that direct recycling preserves the crystalline morphology of the cathode (Gaines, 2018).

Apart from economic aspects, which historically have represented a central element in the evaluation of general waste treatment processes, the assessment of environmental impacts has been incorporated into decision-making. Several companies have announced the ramp-up of new recycling capacity, designed to treat different battery chemistries (Bürklin et al., 2022). Here, a sound understanding of the environmental impacts associated with different recycling approaches is key. Previous work has provided some insight into the environmental impacts of different recycling approaches (Lai et al., 2022; Li et al., 2023; Mohr et al., 2020). However, previous work falls short of providing a detailed comparison between different sources of primary data and its impact on the robustness of results. By presenting an up-to-date critical review about environmental impacts of battery recycling, we support industry and policy-makers in shaping their recycling strategies while also providing guidance for future research on battery recycling with low environmental impacts. LIBs with either lithium-iron phosphate (LFP) or lithium nickel cobalt manganese oxide (NMC) account for most of today's battery production volumes (Hettesheimer et al., 2023). Due to a generally higher cycling stability of LFP batteries, it is reasonable to assume that, at first, significant volumes of NMC batteries will have to be treated at end-of-life (EoL). Thus, in the present review we focus on different recycling strategies for NMC batteries.

The structure of the review is as follows: Section two provides technical background on different recycling processes. Due to the aforementioned drawbacks of pyrometallurgical recycling and the lower technological maturity of direct recycling, we focus here on hydrometallurgical and combined pyro- and hydrometallurgical recycling approaches. Next, in section three, we present publications that have conducted life cycle assessments (LCA) for battery recycling. Differences in data sources, system boundaries and technical battery parameters are addressed. Results from different publications are compared in section four, and possible explanations for variability critically analyzed. In addition, we develop an overview of blank spots in literature which could be subject of future research and provide recommendations to improve the consistency of LCAs for battery recycling.

2 Technical background for hydrometallurgy and combined pyro- and hydrometallurgy

Before NMC batteries are treated by pyro- and hydrometallurgical recycling, the battery packs must first be removed from the application, collected and dismantled (Slattery et al., 2021). The dismantling process is usually carried out manually or semi-automatically (Rajaeifar et al., 2021). Subsequently, the non-cellular material is shredded and further treated to recover aluminium, copper and steel (Accardo et al., 2021; van Hoof et al., 2023). In contrast, the battery cells are discharged and submitted to pyro- and hydrometallurgical recycling processes. The system boundary of our study with different treatment routes is shown in Fig. 1.

The hydrometallurgical recycling of LIBs is a widely adopted method in the industry and describes the use of aqueous solutions to recover valuable metals from spent batteries (Wang et al., 2022). This process comprises a number of key stages, including mechanical pre-treatment, leaching, purification and separation (Chen et al., 2019; Liu et al., 2019).

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

The pre-treatment involves crushing and sieving of the batteries to produce the black mass, which contains valuable metals such as lithium, nickel, cobalt and manganese (Lee et al., 2024). Effective pre-treatment not only improves metal recovery rates, it also reduces environmental impact by minimizing the need for aggressive chemicals and energy during the leaching process (Li et al., 2018). Leaching represents a critical stage in the hydrometallurgical recycling process for LIBs, wherein valuable metals are extracted from the pre-treated battery materials through the use of acidic solutions. Widely used leaching agents include sulphuric acid, hydrochloric acid and nitric acid, often supplemented with hydrogen peroxide to increase metal recovery (Zeng et al., 2014). Ongoing research focuses on developing more environmentally friendly leaching agents to further reduce the ecological footprint (Milian et al., 2024). After leaching, the dissolved metals are purified and separated using techniques such as solvent extraction, ion exchange and selective precipitation. These processes ensure the recovery of high-purity lithium, nickel, cobalt and manganese suitable for reuse in new batteries (Chen et al., 2019; Li et al., 2013). Hydrometallurgical recycling of LIBs offers several key benefits. The most significant advantages of this process include low energy consumption, high recovery purity and a high extraction rate (Hua et al., 2020). Conversely, this recycling method faces the challenges of complex processes and long processing times. Furthermore, the consumption of expensive reducing agents and a considerable quantity of acids and alkalis results in the generation of highly saline organic wastewater (Hua et al., 2020; Yang et al., 2022).

The pyro- and hydrometallurgical treatment also includes the steps pyrolysis and smelting before hydrometallurgical treatment (Brückner et al., 2020; Rajaeifar et al., 2021). These sub-steps offer the advantage of increased throughput with a reduction in plant size and the removal of organic components. In addition, the technology is highly mature and suitable for the initial recovery of alloys by reduction and smelting (Yao et al., 2018). Subsequently, high-purity individual metals and compounds can be obtained through hydrometallurgical treatment. However, high energy consumption, additional processing of the intermediate products and the need for waste gas treatment are required. Despite these challenges, the combined approach remains a promising method for large-scale battery recycling, offering both efficiency and scalability (Brückner et al., 2020; Windisch-Kern et al., 2022).

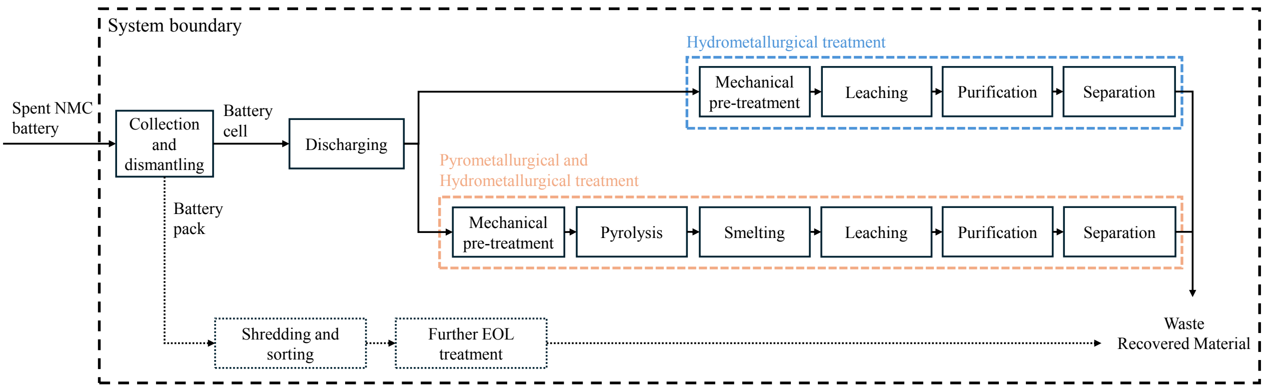

Figure 1: System boundary of this study.

3 Identified literature

The keyword-based search strategy applied in this review was conducted in Scopus and identified a total of 379 potentially relevant publications.[1] After reviewing the abstracts, a total of 64 potentially relevant studies were identified. These 64 studies were analyzed in detail for their relevance to the topic. Possible reasons for exclusion included consideration of a different cell chemistry (n=19), limited detailed LCA (n=13) or different recycling technology (n=8). Finally, 24 studies remained relevant to the environmental assessment of pyro- and hydrometallurgical recycling of NMC batteries. The results of the literature review are presented in Table 1.

[1] Keywords used: (batter* OR lithium-ion) AND (LCA OR Life Cycle Assessment) AND (Recycling OR Recover OR Circular); Field of search: Titel, abstract, keywords; Focus: January 2019 – August 2024

JOURNAL OF BUSINESS CHEMISTRY

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

Table 1: Life cycle assessments of pyro- and hydrometallurgical recycling of lithium-ion based NMC Batteries.

No.	Name, Year	Functional Unit	Background Data, Impact Assessment Method	Energy density	Chemistry	Region
1	Cusenza et al., 2019	11.4 kWh Pack	PEF, Ecoinvent 3	65 Wh/kg	NMC*	Europe
2	Ciez and Whitacre, 2019	1 kg Cell	Ecoinvent, GREET	270 Wh/kg	NMC*	USA
3	Zhu and Chen, 2020	1 kWh Pack	Ecoinvent, GREET	n/a	NMC 622*	China, USA
4	Tao and You, 2020	1 kWh Pack	ReCiPe, Ecoinvent V3.6, GREET	181 Wh/kg	NMC 622*	n/a
5	Mohr et al., 2020	1 kWh Pack	ILCD midpoint, Ecoinvent 3.4	105 Wh/kg	NMC 811*	Europe
6	Sun et al., 2020	1 kWh Pack	CML-IA baseline V3.02, Ecoinvent 3.0, GREET 2018	115 Wh/kg	NMC 622	China
7	Xiong et al., 2020	1 kg Cell	EverBatt	164.37 Wh/kg	NMC 111	China
8	Accardo et al., 2021	1 kWh Pack	CML baseline, Ecoinvent 3.6	213 Wh/kg	NMC 111, NMC 622, NMC 811	China, Europe
9	Rajaeifar et al., 2021	1 kg Cell	Ecoinvent	150 Wh/kg	NMC 111*	China
10	Jiang et al., 2022	1 ton Pack	CML, Ecoinvent V3.6	n/a	NMC 111	United Kingdom
11	Chen et al., 2022	1 kWh Pack	n/a	228 Wh/kg	NMC 811	China
12	Kallitsis et al., 2022	1 kWh Pack	Ecoinvent	105 Wh/kg	NMC 111	China
13	Feng et al., 2022	1 kWh	ReCiPe 2016 Midpoint	120 Wh/kg	NMC*	China
14	Castro et al., 2022	569 g Cell	ReCiPe 2016 v1.1 Midpoint, Ecoinvent v3.6	65,2 Wh/kg	NMC	Europe
15	Quan et al., 2022	1 kWh Pack	CML2001, Ecoinvent, GREET	142.4 Wh/kg	NMC*	China
16	Wang et al., 2022	1 kWh Pack	ReCiPe, GREET	n/a	NMC*	China
17	Blömeke et al., 2022	95 kWh Pack	Ecoinvent 3.8	n/a	NMC 622	Germany
18	Rosenberg et al., 2023	1 kg Pack	CML2001, GaBi Professional Database	142 Wh/kg	NMC 111, NMC 811	Germany
19	Wu et al., 2023	1 kWh	EDIP 2003, CML IA-baseline, ReCiPe 2016, Ecoinvent3, PCF Database	n/a	NMC 811	China
20	Gutsch and Leker, 2023	1 kWh Cell	PEF 3.0, ReCiPe 2016, Ecoinvent 3.8	281 Wh/kg	NMC 811	USA
21	Haupt et al., 2023	1 kg	CML 4.8 2016, Ecoinvent	n/a	NMC 622	n/a
22	Yang et al., 2024	1 kg	PEF 3.0	n/a	NMC*	China
23	Gong et al., 2024	1 kWh	ReCiPe 2016, Ecoinvent 3.8	163 Wh/kg	NMC 811*	China
24	Ali et al., 2024	42.2 kWh Pack	Ecoinvent 3.8	n/a	NMC 622	Europe

*Other cell chemistries are analysed in addition.

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

LCA has become a widely used method for evaluating the environmental impact of industrial products and complex systems (Dong et al., 2021; Guinee J. B., 2001). The procedure for a LCA is described in standardized form in ISO 14040/44 (International Organization for Standardization, 2009). Accordingly, an LCA consists of four parts: 1. goal & scope definition, 2. life cycle inventory (LCI), 3. life cycle impact assessment (LCIA) and 4. Interpretation. In the initial phase, the system boundaries and functional unit are defined (Unterreiner et al., 2016). For this study, we defined the grave-to-cradle approach as system boundary to examine the ecological effects of pyro- and hydrometallurgical recycling. Thus, the entire recycling process, including collection, dismantling, discharging, pyro- and hydrometallurgical treatment is considered. In the LCI phase, data are gathered and evaluated to ensure their accuracy, completeness, and consistency for subsequent use in impact assessments (Hauschild et al., 2018). During the LCIA step, inventory data are translated into indicators for environmental impacts categories using an impact assessment methodology (International Organization for Standardization, 2009). In this context, midpoint and endpoint characterization models are basically two different approaches. The midpoint approach treats environmental impacts in a problem-oriented manner and the endpoint approach in a damage-oriented manner (Dong et al., 2021). In this paper, we have focused on studies that use the midpoint approach to better understand the causes of environmental impacts. As shown in Table 2, 17 of the 24 relevant studies used ReCiPe (Huijbregts et al., 2017) or CML (Guinee, 2002) at midpoint level as impact assessment method.

Primary data are included in 11 of the 24 relevant studies (Blömeke et al., 2022; Cusenza et al., 2019; Feng et al., 2022; Haupt et al., 2023; Jiang et al., 2022; Kallitsis et al., 2022; Mohr et al., 2020; Rajaeifar et al., 2021; Sun et al., 2020; Yang et al., 2024; Zhu and Chen, 2020). Among these eleven studies, five LCAs are based exclusively on primary data (Blömeke et al., 2022; Haupt et al., 2023; Jiang et al., 2022; Yang et al., 2024; Zhu and Chen, 2020). Consequently, the remaining 13 studies rely on secondary data (Accardo et al., 2021; Ali et al., 2023; Castro et al., 2022; Chen et al., 2022; Ciez and Whitacre, 2019; Dai et al., 2019b; Gong et al., 2024; Gutsch and Leker, 2024; Quan et al., 2022; Rosenberg et al., 2023; Tao and You, 2020; Wang et al., 2022; Wu et al., 2023; Xiong et al., 2020). These findings are consistent with those of Bauer (Bauer et al., 2022) which further addressed the imbalance between primary and secondary data studies (Aichberger and Jungmeier, 2020; Degen and Schütte, 2022; Ellingsen et al., 2017; Peters et al., 2017). However, the proportion of studies that only use primary data is slightly higher for recycling (21%) than for battery cell production (12%) (Degen and Schütte, 2022). Looking at the links between the studies in Fig. 2, it is noticeable that the studies by Mohr (Mohr et al., 2020) and Dai (Dai et al., 2019b) are central in the literature. Mohr conducted a study based on primary and secondary data, as well as the Ecoinvent database. In contrast, the EverBatt model of Dai relied on secondary data and the Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET) model (Argonne National Laboratory, 2018a, 2018b). The GREET model is from Argonne National Laboratory (ANL), providing researchers with great access to primary data on the production and EoL treatment of LIBs (Benavides et al., 2015; Dunn et al., 2015b; Wang et al., 2023). Approximately 45% of the relevant studies refer directly or indirectly through other studies to data from the EverBatt model of Dai et al. (see Fig. 2).

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

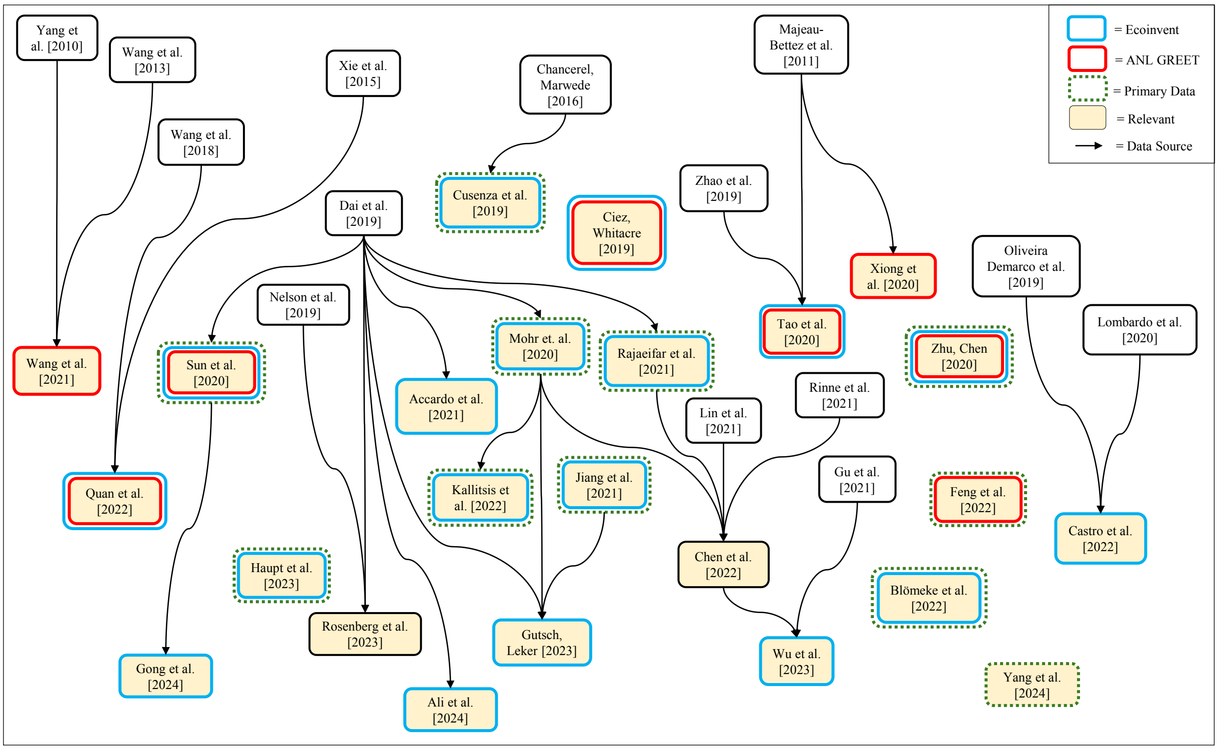

Figure 2: Relationships between LCA studies.

4 Results

To ensure the comparability of the reported results, the functional unit was set to 1 kWh battery pack. When data for the pack level were not available, the analysis was conducted at cell level. Studies where the functional unit differed from 1 kWh of battery capacity were adjusted based on the specific energy density.

4.1 Global Warming Potential

Global warming potential (GWP) is the most frequently investigated impact category for LCA in the field of battery cell production and reflects the influence on climate change by kg CO2 to air (Huijbregts et al., 2017; Peters et al., 2017). While 67% of the studies about battery cell production have assessed GWP (Peters et al., 2017), all considered studies on recycling have examined it. The total GWP impact is the sum of the GWP recycling credit and GWP recycling burden (see equation 1).

$$GWP_{total} = \sum GWP_{Credit}\,, GWP_{Burden} \quad (1)$$

It was found that the GWP burden in all studies ranges for the hydrometallurgical treatment between 2.7 and 34.0 kg CO2eq kWh-1. For combined pyro- and hydrometallurgical treatment emissions are between 5.1 and 15.8 kg CO2eq kWh^{-1} (see Fig. 3). The discrepancies in the literature can be primarily attributed to variations in the content of nickel, manganese, and cobalt, as well as different production locations, since greenhouse gas (GHG) emissions resulting from a similar recycling process in China are higher than those observed in most European countries or the US (Xiong et al., 2020). Furthermore, studies based on the same primary data show consistent results (Chen et al., 2022; Gong et al., 2024; Mohr et al., 2020; Sun et al., 2020; Wu et al., 2023). This emphasizes the importance of collecting new primary data to gain new insights. With respect to GWP credit and GWP total impact, the study of Kallitsis (Kallitsis et al., 2022) differs from the remaining studies. This can in part be explained by the functional unit being set at pack level, allowing greater GWP benefits to be achieved through aluminum, copper and steel recovery (Kallitsis et al., 2022). If we compare the results of Mohr and Kallitsis, it is noticeable that the values for burden, credit and total from Mohr are only 53% of Kallitsis results. As the Mohr study was also conducted at the pack

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

level, this alone cannot explain the discrepancies in the results. Another explanation would be that Kallitsis study was conducted in China, where the GWP benefits from the recovered materials are the highest, as production is related to high CO2 emissions (Kallitsis et al., 2022). However, these criteria were also applied in the studies by Sun (Sun et al., 2020) and Accardo (Accardo et al., 2021). One distinguishing characteristic is that the study by Kallitsis collected primary data from the industry in addition to secondary data. This indicates that the recycling process analysed was already further advanced, which also underlines the importance of collecting primary data.

The average burden of the examined studies for recycling of 1 kWh NMC battery with hydrometallurgical treatment is 9.5 kg CO2eq and for combined pyro- and hydrometallurgical treatment 11.9 kg CO2eq. In comparison, the production of 1 kWh NMC 811 battery releases 64.5 kg CO2eq (Gutsch and Leker, 2024). Thus, the CO2 emissions from recycling are about 15-19% of battery cell production. In the studies reviewed, the average total CO2 emissions associated with hydrometallurgical recycling are - 25.5 kg CO2eq kWh^{-1}. In comparison, the average total CO2 emissions associated with combined pyro- and hydrometallurgical recycling are - 17.5 kg CO2eq kWh^{-1}. This indicates that combined pyro- and hydrometallurgical recycling can reduce the GWP of battery cell production by 27%. Hydrometallurgical recycling can result in a reduction of GWP associated with battery cell production by 39%. By comparing the results of the hydrometallurgical approach with those of the combined pyrometallurgical and hydrometallurgical approach, it becomes evident that the studies conducted by Kallitsis (Kallitsis et al., 2022), Chen (Chen et al., 2022), Wu (Wu et al., 2023) and Yang (Yang et al., 2024) are particularly suitable for comparison, since both approaches were investigated under identical conditions. The slight differences in the average values for burden and total can also be observed here. Additionally, the study by Accardo (Accardo et al., 2021) in the context of the combined pyro- and hydrometallurgical approach is unique in reporting a positive value for the total GWP. Therefore, hydrometallurgy is more environmentally friendly in terms of GWP compared to the combination of pyro- and hydrometallurgy (see Fig. 3).

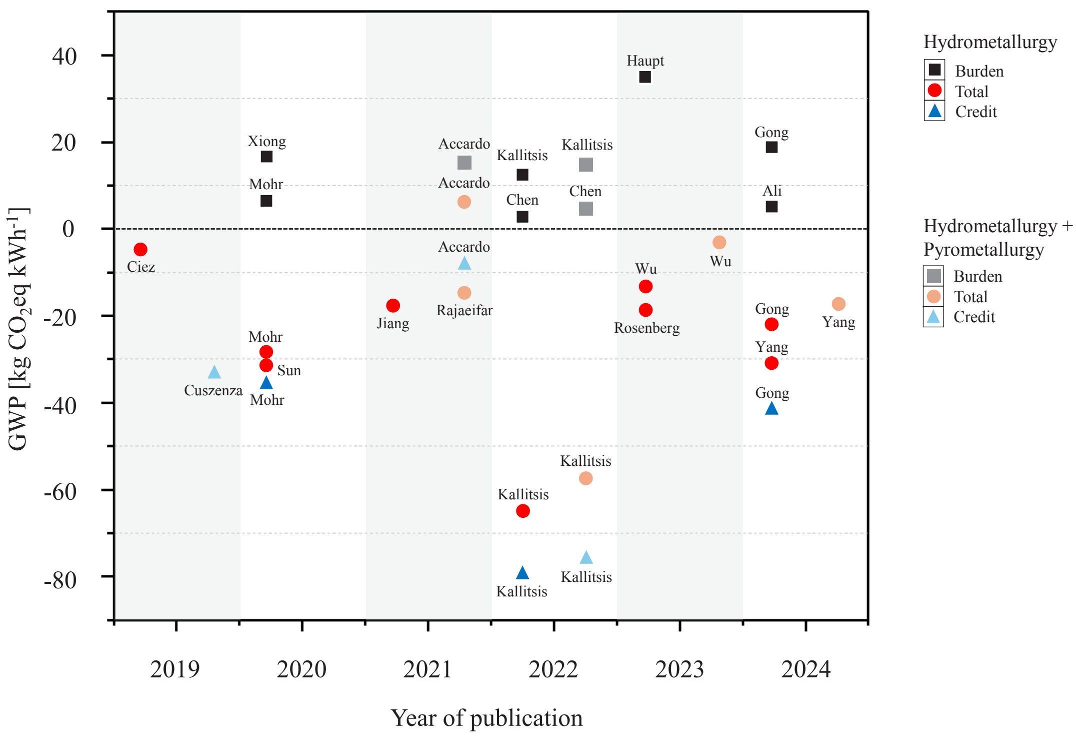

Figure 3: GWP from selected studies in pyro- and hydrometallurgical recycling of 1 kWh NMC battery cell or pack.

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

4.2 Cumulative Energy Demand

According to Peters (Peters et al., 2017), cumulated energy demand (CED) is the second most frequently investigated impact category for LCA in the field of battery cell production and reflects the energy consumption in MJ. While 53% of the studies about battery cell production have evaluated CED (Peters et al., 2017), 54% of the considered studies have analyzed it. The total CED impact is the sum of the CED recycling credit and CED recycling burden (see equation 2).

$$CED_{total} = \sum CED_{Credit}, CED_{Burden} \quad (2)$$

The analysis reveals that the total CED of all relevant studies ranges for the hydrometallurgical treatment between - 453.3 and - 62.9 MJ kWh^{-1} and for the combined pyro- and hydrometallurgical treatment between - 257.6 and 256.8 MJ kWh^{-1} (see Fig. 4). Specifically, the average value for the CED burden of hydrometallurgical recycling is 198.2 MJ kWh^{-1}. In contrast, the combined pyrometallurgical and hydrometallurgical recycling process exhibits a higher average value of the CED burden, with 228.2 MJ kWh^{-1}. A comparative analysis of the average recycling credit values reveals that hydrometallurgical recycling achieves a more significant reduction in energy consumption compared to the combined pyro- and hydrometallurgical recycling process. In particular, the hydrometallurgical process offers an average energy credit of - 492.5 MJ kWh^{-1}, whereas the combined approach yields a comparatively lower credit of - 326.1 MJ kWh^{-1}. This disparity highlights the greater efficiency of the hydrometallurgical method in reducing energy demands and demonstrates that the pyrolysis and smelting sub-steps are characterized by a high energy consumption (Accardo et al., 2021).

Furthermore, it can be determined that 83% of the studies demonstrate a benefit in total CED, establishing a general consensus within the research community. Therefore, the average total CED for hydrometallurgical recycling is calculated to be - 189.5 MJ kWh^{-1}. Hydrometallurgical recycling can reduce the CED by almost 17%, given that the production of 1 kWh NMC battery requires 1,126 MJ of energy (Dai et al., 2019a). However, the study by Accardo differs from this trend with a positive total CED, due to high energy requirements of the pyrometallurgical steps (Accardo et al., 2021). Nevertheless, Rajaeifar conclude that the combined pyro- and hydrometallurgical recycling process yields a net benefit (Rajaeifar et al., 2021). This can be attributed to the closed-loop approach, which is considered the best-case scenario, as it allocates optimistically high energy and environmental credits to the system. Additionally, since 2022, fewer studies have analyzed the values for recycling CED total and recycling CED credit (see Fig. 4). To gain a comprehensive understanding of the impact of recycling, it is essential to address these aspects in future research.

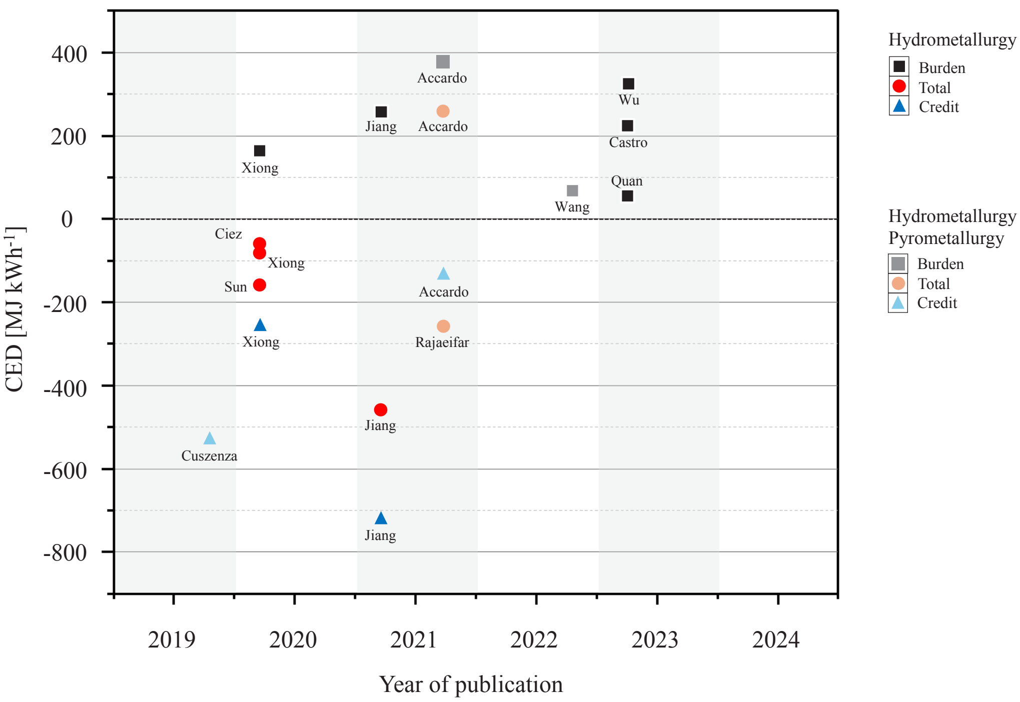

Figure 4: CED of selected studies in pyro- and hydrometallurgical recycling of 1 kWh NMC battery cell or pack.

JOURNAL OF
BUSINESS CHEMISTRY

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

5 Further Impact categories

The objective of a LCA is to quantify the overall environmental impacts of a system (International Organization for Standardization, 2009). Accordingly, in the LCIA phase exists a wide set of characterisation models and impact categories that can be used to evaluate the environmental impact (Finkbeiner et al., 2006; International Organization for Standardization, 2009). In the analyzed studies, the used impact categories consist of the frequently applied impact assessment methods ReCiPe (Guinee, 2002; Huijbregts et al., 2017), CML (Guinee, 2002) and the impact category CED. The matrix presented in Fig. 5 summarizes the considered impact categories in the relevant studies. However, looking at the impact categories, it appears that many studies consider only GWP and very few other categories in addition, while still referring to the concept of LCA. In particular, only the studies by Cusenza (Cusenza et al., 2019), Kallitsis (Kallitsis et al., 2022), Castro (Castro et al., 2022) and Yang (Yang et al., 2024) analyse a wide spectrum of impact categories. The study by Gutsch and Leker (Gutsch and Leker, 2024) employs a combined approach to assess a number of impact categories simultaneously. Furthermore, it is evident that since 2022, none of the analyzed studies have examined the impact category CED, including its burden, credit and total recycling values (see Fig. 5). This absence highlights a gap in the current research, as CED represents a crucial indicator for comprehending the total energy implications of recycling processes. This includes the energy conserved through material recovery and the energy consumed in the recycling operations themselves.

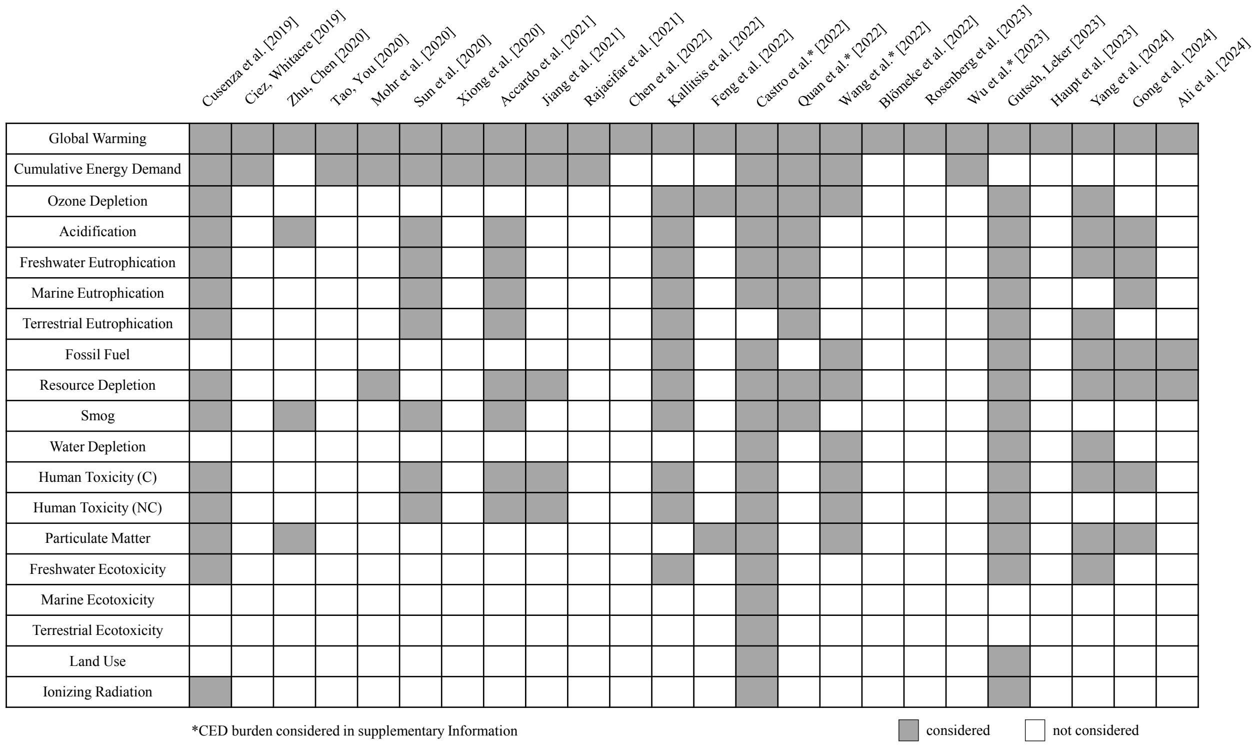

Figure 5: Overview of the impact categories considered within relevant studies.

The Bauer commentary (Bauer et al., 2022) highlights that the EoL management of batteries is potentially harmful to the natural environment and human health. Moreover, the findings of the study by Kallitsis indicate that the recycling credits can be even higher in impact categories other than GWP and CED (Kallitsis et al., 2022). This is in line with the results reported by Accardo, who identified the greatest benefits from recycling in the impact categories of human toxicity and acidification (Accardo et al., 2021). Therefore, LCA about battery EoL treatment should examine impact

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

categories like Human Toxicity Potential, Resource Depletion, Acidification Potential and Terrestrial Eutrophication Potential in addition to GWP and CED to address aspects of human health and natural environment. However, the analysis of the relevant studies shows that on average only 40% of these four categories are analyzed. Consequently, future research should prioritise a more comprehensive investigation.

6 Conclusions and outlook

This work provides an overview about the current state of research on the environmental impacts of pyro- and hydrometallurgical recycling of NMC batteries. It was found that most LCAs were conducted in China, followed by Europe and the United States. As background data, GREET (Argonne National Laboratory, 2018b, 2020; Dunn et al., 2014; Dunn et al., 2015a) and Ecoinvent (Wernet et al., 2016) are used by a large number of studies. We improved the comparability of relevant studies by analyzing the system boundaries and adjusting the functional units to a standard of 1 kWh battery pack. Our analysis results show that pyro- and hydrometallurgical recycling of NMC batteries is described in the literature with predominantly positive environmental effects. Combined pyro- and hydrometallurgical recycling can reduce the GWP of battery cell production by 27%. Conversely, hydrometallurgical recycling can result in a reduction of GWP by 39%. Furthermore, it can be summarised that the pyrolysis and smelting sub-steps are distinguished by a high level of energy consumption. Conversely, hydrometallurgical recycling can reduce the CED of battery cell production by almost 17%. The findings suggest that studies using secondary data generally show a high degree of consistency with those using primary data. In the literature, environmental categories beyond GWP and CED generally receive less attention, even though their benefits are often more significant.

When evaluating this study, it is important to acknowledge that it encompasses battery cells with varying compositions of nickel, manganese, and cobalt. Moreover, LIBs were analyzed at both the pack and cell levels, which can lead to variations in the results. Further research could investigate the EoL environmental impacts of other cell chemistries, such as LFP or Lithium nickel cobalt aluminium oxide (NCA), in terms of data sources and different environmental impact categories. Based on our findings, we offer three recommendations for conducting future LCAs:

- *Recommendation 1:* In addition to the total impact, it is advisable to include the credit and burden in absolute terms. This allows a more differentiated analysis and comparison of the individual recycling processes and ensures that both the benefits and the drawbacks of each process are clearly identified.
- *Recommendation 2:* To facilitate direct comparisons between studies, it is recommended to standardize the functional unit to 1 kWh at pack level. Additionally, given that the majority of current studies rely on secondary data, it is recommended that more primary data be collected to ensure the gathering of new insights.
- *Recommendation 3:* Besides GWP and CED, other impact categories are not regularly considered. To gain a comprehensive understanding of the impact on both humans and the environment, it is important to consider additional impact categories such as Human Toxicity Potential, Acidification Potential, Terrestrial Eutrophication Potential and Resource Depletion.

Funding sources

This research was partially funded and supported by the German Federal Ministry of Education and Research within the project ProRec [03XP0537A] and the Ministry of Culture and Science of the State of North Rhine-Westphalia within the project BIOSTORE [PB22-014].

CRediT authorship contribution statement

Luca Stegemann: Conceptualization, Methodology, Investigation, Data curation, Formal analysis, Writing – original draft, Writing – review & editing.
Moritz Gutsch: Writing – review & editing, Supervision, Validation, Project administration.

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

References

Accardo, A., Dotelli, G., Musa, M.L., Spessa, E., 2021. Life Cycle Assessment of an NMC Battery for Application to Electric Light-Duty Commercial Vehicles and Comparison with a Sodium-Nickel-Chloride Battery. Applied Sciences 11, 1160.

Aichberger, C., Jungmeier, G., 2020. Environmental Life Cycle Impacts of Automotive Batteries Based on a Literature Review. Energies 13, 6345.

Ali, A.-R., Bartie, N., Husmann, J., Cerdas, F., Schröder, D., Herrmann, C., 2023. Simulation-Based Life Cycle Assessment of Secondary Materials from Recycling of Lithium-Ion Batteries. Elsevier BV.

Argonne National Laboratory, 2018a. Cobalt life cycle analysis update for the GREET Model.

Argonne National Laboratory, 2018b. Update of Bill-of-materials and Cathode Materials Production for Lithium-ion Batteries in the GREET Model.

Argonne National Laboratory, 2020. Update of bill-of-materials and cathode chemistry addition for lithium-ion batteries in GREET 2020.

Bauer, C., Burkhardt, S., Dasgupta, N.P., Ellingsen, L.A.-W., Gaines, L.L., Hao, H., Hischier, R., Hu, L., Huang, Y., Janek, J., Liang, C., Li, H., Li, J., Li, Y., Lu, Y.-C., Luo, W., Nazar, L.F., Olivetti, E.A., Peters, J.F., Rupp, J.L.M., Weil, M., Whitacre, J.F., Xu, S., 2022. Charging sustainable batteries. Nat Sustain 5, 176–178.

Benavides, P.T., Dai, Q., Sullivan, J.L., Kelly, J.C., Dunn, J.B., 2015. Material and Energy Flows Associated with Select Metals in GREET 2. Molybdenum, Platinum, Zinc, Nickel, Silicon. Argonne National Lab. (ANL), Argonne, IL (United States) ANL/ESD-15/11. https://www.osti.gov/biblio/1224976.

Blömeke, S., Scheller, C., Cerdas, F., Thies, C., Hachenberger, R., Gonter, M., Herrmann, C., Spengler, T.S., 2022. Material and energy flow analysis for environmental and economic impact assessment of industrial recycling routes for lithium-ion traction batteries. Journal of Cleaner Production 377, 134344.

Brückner, L., Frank, J., Elwert, T., 2020. Industrial Recycling of Lithium-Ion Batteries—A Critical Review of Metallurgical Process Routes. Metals 10, 1107.

Bürklin, B., Latz, T., Schenk, L., Degen, F., Diehl, M., Krätzig, O., Paulsen, T., Wessel, S., Kampker, A., Lackner, N., Neef, C., Wicke, T., 2022. Umfeldbericht zum europäischen Innovationssystem Batterie 2022. Fraunhofer ISI.

Castro, F., Mehner, E., Cutaia, L., Vaccari, M., 2022. Life cycle assessment of an innovative lithium-ion battery recycling route: A feasibility study. Journal of Cleaner Production 368, 133130.

Chen, M., Ma, X., Chen, B., Arsenault, R., Karlson, P., Simon, N., Wang, Y., 2019. Recycling End-of-Life Electric Vehicle Lithium-Ion Batteries. Joule 3, 2622–2646.

Chen, Q., Lai, X., Gu, H., Tang, X., Gao, F., Han, X., Zheng, Y., 2022. Investigating carbon footprint and carbon reduction potential using a cradle-to-cradle LCA approach on lithium-ion batteries for electric vehicles in China. Journal of Cleaner Production 369, 133342.

Ciez, R.E., Whitacre, J.F., 2019. Examining different recycling processes for lithium-ion batteries. Nat Sustain 2, 148–156.

Cusenza, M.A., Bobba, S., Ardente, F., Cellura, M., Di Persio, F., 2019. Energy and environmental assessment of a traction lithium-ion battery pack for plug-in hybrid electric vehicles. Journal of Cleaner Production 215, 634–649.

Dai, Q., Kelly, J.C., Gaines, L., Wang, M., 2019a. Life Cycle Analysis of Lithium-Ion Batteries for Automotive Applications. Batteries 5, 48.

Dai, Q., Spangenberger, J., Ahmed, S., Gaines, L., Kelly, J.C., Wang, M., 2019b. EverBatt: A Closed-loop Battery Recycling Cost and Environmental Impacts Model. Office of Scientific and Technical Information (OSTI).

Davis, K., Demopoulos, G.P., 2023. Hydrometallurgical recycling technologies for NMC Li-ion battery cathodes: current industrial practice and new R&D trends. RSC Sustain. 1, 1932–1951.

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

Degen, F., Schütte, M., 2022. Life cycle assessment of the energy consumption and GHG emissions of state-of-the-art automotive battery cell production. Journal of Cleaner Production 330, 129798.

Dong, Y., Hossain, M.U., Li, H., Liu, P., 2021. Developing Conversion Factors of LCIA Methods for Comparison of LCA Results in the Construction Sector. Sustainability 13, 9016.

Dunn, J.B., Gaines, L., Barnes, M., Sullivan, J.L., Wang, M., 2014. Material and Energy Flows in the Materials Production, Assembly, and End-of-Life Stages of the Automotive Lithium-Ion Battery Life Cycle. Argonne National Lab. (ANL), Argonne, IL (United States) ANL/ESD/12-3 Rev. https://www.osti.gov/biblio/1177517.

Dunn, J.B., James, C., Gaines, L., Gallagher, K., Dai, Q., Kelly, J.C., 2015a. Material and Energy Flows in the Production of Cathode and Anode Materials for Lithium Ion Batteries. Argonne National Lab. (ANL), Argonne, IL (United States) ANL/ESD-14/10 Rev. https://www.osti.gov/biblio/1224963.

Dunn, J.B., James, C., Gaines, L., Gallagher, K., Dai, Q., Kelly, J.C., 2015b. Material and Energy Flows in the Production of Cathode and Anode Materials for Lithium Ion Batteries. Argonne National Lab. (ANL), Argonne, IL (United States) ANL/ESD-14/10 Rev. https://www.osti.gov/biblio/1224963.

Ellingsen, L.A.-W., Hung, C.R., Strømman, A.H., 2017. Identifying key assumptions and differences in life cycle assessment studies of lithium-ion traction batteries with focus on greenhouse gas emissions. Transportation Research Part D: Transport and Environment 55, 82–90.

Feng, T., Guo, W., Li, Q., Meng, Z., Liang, W., 2022. Life cycle assessment of lithium nickel cobalt manganese oxide batteries and lithium iron phosphate batteries for electric vehicles in China. Journal of Energy Storage 52, 104767.

Finkbeiner, M., Inaba, A., Tan, R., Christiansen, K., Klüppel, H.-J., 2006. The New International Standards for Life Cycle Assessment: ISO 14040 and ISO 14044. Int J Life Cycle Assess 11, 80–85.

Gaines, L., 2014. The future of automotive lithium-ion battery recycling: Charting a sustainable course. Sustainable Materials and Technologies 1-2, 2–7.

Gaines, L., 2018. Lithium-ion battery recycling processes: Research towards a sustainable course. Sustainable Materials and Technologies 17, e00068.

Gong, W.T., Daigo, I., Dunuwila, P., Sun, X., 2024. The change of material flows and environmental impacts along with future upgrading scenarios of NCM battery in China. Journal of Cleaner Production 463, 142758.

Guinee, J.B., 2002. Handbook on life cycle assessment. Operational guide to the ISO standards. Kluwer, Dordrecht.
Guinee J. B., 2001. Life Cycle Assessment: An operational guide to the ISO standards.

Gutsch, M., Leker, J., 2024. Costs, carbon footprint, and environmental impacts of lithium-ion batteries – From cathode active material synthesis to cell manufacturing and recycling. Applied Energy 353, 122132.

Haupt, J., Kononova, N., Cerdas, F., Zellmer, S., Herrmann, C., 2023. Challenges of prospective life cycle assessment of emerging recycling processes: case study of battery materials recovery. Procedia CIRP 116, 23–28.

Hauschild, M.Z., Rosenbaum, R.K., Olsen, S.I. (Eds.), 2018. Life cycle assessment. Theory and practice. Springer International Publishing; Imprint; Springer, Cham.

He, B., Zheng, H., Tang, K., Xi, P., Li, M., Wei, L., Guan, Q., 2024. A Comprehensive Review of Lithium-Ion Battery (LiB) Recycling Technologies and Industrial Market Trend Insights. Recycling 9, 9.

Hettesheimer, T., Neef, C., Rosellón Inclán, I., Link, S., Schmaltz, T., Schuckert, F., Stephan, A., Stephan, M., Thielmann, A., Weymann, L., Wicke, T., 2023. Lithium-Ion Battery Roadmap - Industrialization Perspectives toward 2030. Fraunhofer ISI.

Hua, Y., Zhou, S., Huang, Y., Liu, X., Ling, H., Zhou, X., Zhang, C., Yang, S., 2020. Sustainable value chain of retired lithium-ion batteries for electric vehicles. Journal of Power Sources 478, 228753.

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

Huijbregts, M.A.J., Steinmann, Z.J.N., Elshout, P.M.F., Stam, G., Verones, F., Vieira, M., Zijp, M., Hollander, A., van Zelm, R., 2017. ReCiPe2016: a harmonised life cycle impact assessment method at midpoint and endpoint level. Int J Life Cycle Assess 22, 138–147.

International Organization for Standardization, 2009. ISO 14040: 2006 Environmental Management–Life Cycle Assessment–Principles and Framework.

Islam, M.T., Iyer-Raniga, U., 2022. Lithium-Ion Battery Recycling in the Circular Economy: A Review. Recycling 7, 33.

Jiang, S., Hua, H., Zhang, L., Liu, X., Wu, H., Yuan, Z., 2022. Environmental impacts of hydrometallurgical recycling and reusing for manufacturing of lithium-ion traction batteries in China. The Science of the total environment 811, 152224.

Jung, J.C.-Y., Sui, P.-C., Zhang, J., 2021. A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments. Journal of Energy Storage 35, 102217.

Kallitsis, E., Korre, A., Kelsall, G.H., 2022. Life cycle assessment of recycling options for automotive Li-ion battery packs. Journal of Cleaner Production 371, 133636.

Kim, S., Bang, J., Yoo, J., Shin, Y., Bae, J., Jeong, J., Kim, K., Dong, P., Kwon, K., 2021. A comprehensive review on the pretreatment process in lithium-ion battery recycling. Journal of Cleaner Production 294, 126329.

Lai, X., Chen, Q., Tang, X., Zhou, Y., Gao, F., Guo, Y., Bhagat, R., Zheng, Y., 2022. Critical review of life cycle assessment of lithium-ion batteries for electric vehicles: A lifespan perspective. eTransportation 12, 100169.

Lee, J., Park, K.W., Sohn, I., Lee, S., 2024. Pyrometallurgical recycling of end-of-life lithium-ion batteries. Int J Miner Metall Mater 31, 1554–1571.

Li, J., Li, L., Yang, R., Jiao, J., 2023. Assessment of the lifecycle carbon emission and energy consumption of lithium-ion power batteries recycling: A systematic review and meta-analysis. Journal of Energy Storage 65, 107306.

Li, L., Dunn, J.B., Zhang, X.X., Gaines, L., Chen, R.J., Wu, F., Amine, K., 2013. Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment. Journal of Power Sources 233, 180–189.

Li, L., Zhang, X., Li, M., Chen, R., Wu, F., Amine, K., Lu, J., 2018. The Recycling of Spent Lithium-Ion Batteries: a Review of Current Processes and Technologies. Electrochem. Energ. Rev. 1, 461–482.

Liu, C., Lin, J., Cao, H., Zhang, Y., Sun, Z., 2019. Recycling of spent lithium-ion batteries in view of lithium recovery: A critical review. Journal of Cleaner Production 228, 801–813.

Milian, Y.E., Jamett, N., Cruz, C., Herrera-León, S., Chacana-Olivares, J., 2024. A comprehensive review of emerging technologies for recycling spent lithium-ion batteries. The Science of the total environment 910, 168543.

Mohr, M., Peters, J.F., Baumann, M., Weil, M., 2020. Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes. J of Industrial Ecology 24, 1310–1322.

Peters, J.F., Baumann, M., Zimmermann, B., Braun, J., Weil, M., 2017. The environmental impact of Li-Ion batteries and the role of key parameters – A review. Renewable and Sustainable Energy Reviews 67, 491–506.

Quan, J., Zhao, S., Song, D., Wang, T., He, W., Li, G., 2022. Comparative life cycle assessment of LFP and NCM batteries including the secondary use and different recycling technologies. The Science of the total environment 819, 153105.

Rajaeifar, M.A., Raugei, M., Steubing, B., Hartwell, A., Anderson, P.A., Heidrich, O., 2021. Life cycle assessment of lithium-ion battery recycling using pyrometallurgical technologies. Journal of Industrial Ecology 25, 1560–1571.

Rosenberg, S., Glöser-Chahoud, S., Huster, S., Schultmann, F., 2023. A dynamic network design model with capacity expansions for EoL traction battery recycling - A case study of an OEM in Germany. Waste management (New York, N.Y.) 160, 12–22.

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

Slattery, M., Dunn, J., Kendall, A., 2021. Transportation of electric vehicle lithium-ion batteries at end-of-life: A literature review. Resources, Conservation and Recycling 174, 105755.

Sun, X., Luo, X., Zhang, Z., Meng, F., Yang, J., 2020. Life cycle assessment of lithium nickel cobalt manganese oxide (NCM) batteries for electric passenger vehicles. Journal of Cleaner Production 273, 123006.

Tao, Y., You, F., 2020. Comparative Life Cycle Assessment of three Recycling Approaches for Electric Vehicle Lithium-ion Battery after Cascaded Use.

Unterreiner, L., Jülch, V., Reith, S., 2016. Recycling of Battery Technologies – Ecological Impact Analysis Using Life Cycle Assessment (LCA). Energy Procedia 99, 229–234.

van Hoof, G., Robertz, B., Verrecht, B., 2023. Towards Sustainable Battery Recycling: A Carbon Footprint Comparison between Pyrometallurgical and Hydrometallurgical Battery Recycling Flowsheets. Metals 13, 1915.

Wang, M., Elgowainy, A., Lee, U., Baek, K., Balchandani, S., Benavides, P., Burnham, A., Cai, H., Chen, P., Gan, Y., Gracida-Alvarez, U., Hawkins, T., Huang, T.-Y., Iyer, R., Kar, S., Kelly, J., Kim, T., Kolodziej, C., Lee, K., Liu, X., Lu, Z., Masum, F., Morales, M., Ng, C., Ou, L., Poddar, T., Reddi, K., Shukla, S., Singh, U., Sun, L., Sun, P., Vyawahare, P., Zhang, J., 2023. Summary of Expansions and Updates in R&D GREET® 2023. Argonne National Laboratory (ANL), Argonne, IL (United States) ANL/ESIA-23/10. https://www.osti.gov/biblio/2278803.

Wang, M., Liu, K., Dutta, S., Alessi, D.S., Rinklebe, J., Ok, Y.S., Tsang, D.C., 2022. Recycling of lithium iron phosphate batteries: Status, technologies, challenges, and prospects. Renewable and Sustainable Energy Reviews 163, 112515.

Wernet, G., Bauer, C., Steubing, B., Reinhard, J., Moreno-Ruiz, E., Weidema, B., 2016. The ecoinvent database version 3 (part I): overview and methodology. Int J Life Cycle Assess 21, 1218–1230.

Windisch-Kern, S., Gerold, E., Nigl, T., Jandric, A., Altendorfer, M., Rutrecht, B., Scherhaufer, S., Raupenstrauch, H., Pomberger, R., Antrekowitsch, H., Part, F., 2022. Recycling chains for lithium-ion batteries: A critical examination of current challenges, opportunities and process dependencies. Waste management (New York, N.Y.) 138, 125–139.

Wu, W., Cong, N., Zhang, X., Yue, Q., Zhang, M., 2023. Life cycle assessment and carbon reduction potential prediction of electric vehicles batteries. The Science of the total environment 903, 166620.

Xiong, S., Ji, J., Ma, X., 2020. Environmental and economic evaluation of remanufacturing lithium-ion batteries from electric vehicles. Waste management (New York, N.Y.) 102, 579–586.

Xu, P., Tan, D.H.S., Jiao, B., Gao, H., Yu, X., Chen, Z., 2023. A Materials Perspective on Direct Recycling of Lithium-Ion Batteries: Principles, Challenges and Opportunities. Adv Funct Materials 33.

Yang, C., Zhang, J., Liang, G., Jin, H., Chen, Y., Wang, C., 2022. An advanced strategy of "metallurgy before sorting" for recycling spent entire ternary lithium-ion batteries. Journal of Cleaner Production 361, 132268.

Yang, H., Hu, X., Zhang, G., Dou, B., Cui, G., Yang, Q., Yan, X., 2024. Life cycle assessment of secondary use and physical recycling of lithium-ion batteries retired from electric vehicles in China. Waste management (New York, N.Y.) 178, 168–175.

Yao, Y., Zhu, M., Zhao, Z., Tong, B., Fan, Y., Hua, Z., 2018. Hydrometallurgical Processes for Recycling Spent Lithium-Ion Batteries: A Critical Review. ACS Sustainable Chem. Eng. 6, 13611–13627.

Zeng, X., Li, J., Singh, N., 2014. Recycling of Spent Lithium-Ion Battery: A Critical Review. Critical Reviews in Environmental Science and Technology 44, 1129–1165.

Vol.22, Iss.1, February 2025
URN: urn:nbn:de:hbz:6-43998534359
DOI: 10.17879/43998523309

Zhenghe, X., Zhenda, L., Shubin, W., Zhouguang, L., 2022. Review on hydrometallurgical recovery of valuable metals from spent lithium-ion batteries.

Zhu, L., Chen, M., 2020. Research on Spent LiFePO4 Electric Vehicle Battery Disposal and Its Life Cycle Inventory Collection in China. International Journal of Environmental Research and Public Health 17, 8828.