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Original
Review of lithium-ion batteries' supply-chain in Europe: Material flow analysis and environmental assessment / Bruno, Martina; Fiore, Silvia. - In: JOURNAL OF ENVIRONMENTAL MANAGEMENT. - ISSN 0301-4797. - ELETTRONICO. - 358:(2024), pp. 1-15. [10.1016/j.jenvman.2024.120758]

Availability:
This version is available at: 11583/2987667 since: 2024-04-09T08:16:03Z

Publisher:
Elsevier

Published
DOI:10.1016/j.jenvman.2024.120758

Journal of Environmental Management 358 (2024) 120758

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Review

Review of lithium-ion batteries' supply-chain in Europe: Material flow analysis and environmental assessment

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ARTICLE INFO

Handling editor: Lixiao Zhang

Keywords:
battery
Critical raw materials
GHG emissions
LCA
Recycling

ABSTRACT

European legislation stated that electric vehicles' sale must increase to 35% of circulating vehicles by 2030, and concern is associated to the batteries' supply chain. This review aims at analysing the impacts (about material flows and CO_2 eq emissions) of Lithium-Ion Batteries' (LIBs) recycling at full-scale in Europe in 2030 on the European LIBs' supply-chain. Literature review provided the recycling technologies' (e.g., pyro- and hydro-metallurgy) efficiencies, and an inventory of existing LIBs' production and recycling plants in Europe. European production plants exhibit production capacity adequate for the expected 2030 needs. The key critical issues associated to recycling regard pre-treatments and the high costs and environmental impacts of metallurgical processes. Then, according to different LIBs' composition and market shares in 2020, and assuming a 10-year battery lifetime, the Material Flow Analysis (MFA) of the metals embodied in End of Life (EoL) LIBs forecasted in Europe in 2030 was modelled, and the related CO_2 eq emissions calculated. In 2030 the European LIBs' recycling structure is expected to receive 664 t of Al, 530 t of Co, 1308 t of Cu, 219 t of Fe, 175 t of Li, 287 t of Mn and 486 t of Ni. Of these, 99% Al, 86% Co, 96% Cu, 88% Mn and 98% Ni will be potentially recovered by pyrometallurgy, and 71% Al, 92% Co, 92% Fe, 96% Li, 88 % Mn and 90% Ni by hydrometallurgy. However, even if the recycling efficiencies of the technologies applied at full-scale are high, the treatment capacity of European recycling plants could supply as recycled metals only 2%-wt of the materials required for European LIBs' production in 2030 (specifically 278 t of Al, 468 t of Co, 531 t of Cu, 114 t of Fe, 95 t of Li, 250 t of Mn and 428 t of Ni). Nevertheless, including recycled metals in the production of new LIBs could cut up 28% of CO_2 eq emissions, compared to the use of virgin raw materials, and support the European batteries' value chain.

1. Introduction

The European Green Deal (European Commission, 2019) committed to make Europe climate neutral by 2050, and 90% reduction in greenhouse gas (GHG) emissions associated to transport, compared to 1990's levels, was set as goal for 2050. European targets for electric vehicles' (EVs) sales set 15% by 2025 and 35% by 2030 as benchmarks (IEA, 2021), while recent legislation updates (European Commission, 2020a) established that EVs' sale must increase to 55% by 2030. The forecasted growth of EVs' market is causing concern about securing the raw materials' supply chain, particularly for Lithium-Ion Batteries (LIBs). Commercial LIBs can be identified according to their cathodes' chemistry as nickel manganese cobalt oxide (NMC), lithium cobalt oxide (LCO), nickel cobalt aluminium oxide (NCA), lithium manganese oxide (LMO) and lithium iron phosphate (LFP). Current bottlenecks of European LIBs' value-chain are mostly related to the supply chain, with China, Africa and Latin America providing 74%-wt of all raw materials, and to cells' production (China supplies 66% of finished LIBs) (European Commission, 2020a). LIBs' composition includes metals having low supply risk in Europe as copper and aluminium (in anode and cathode as current collectors), and manganese and nickel (in NMC cathodes), but whose recycling could contribute to the concept of Sustainable Battery (European Commission, 2020b). LIBs are also made of Critical Raw Materials – CRMs, as cobalt (in cathodes), lithium (in cathodes and electrolyte) and graphite (in anodes), and they imply high supply risks in Europe (European Commission, 2020a). Electric mobility appears to be the main driver for cobalt and lithium global demand (Zhang et al., 2021), and serious shortage is expected for the European and global (Mayyas et al., 2019) battery markets. The raw materials' shortage can be addressed by decreasing CRMs' content in new generation's LIBs (Amici et al., 2022; Bella et al., 2021) and by improving the recycling infrastructure (Bruno and Fiore, 2023) to provide "secondary" raw

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https://doi.org/10.1016/j.jenvman.2024.120758
Received 30 September 2023; Received in revised form 26 February 2024; Accepted 23 March 2024
Available online 8 April 2024

materials as alternatives to "primary" virgin raw materials.

The LIBs' supply chain creates concern also due to geopolitical uncertainties (Habib et al., 2016), and to the high environmental impacts associated to mining activities (Azadi et al., 2020). Material Flow Analysis (MFA) is commonly used to examine supply networks and the recycling potential of critical raw materials, at different geographical and temporal scales (Zhang et al., 2023). From a global perspective, concerns arose because the rate of demand growth is outpacing the material supply capacity and the geographical distributions of mining reservoirs, manufacturing facilities, and market distribution for lithium-ion batteries are misaligned (Miao et al., 2023). In order to overcome this limitation, MFA acknowledged the fundamental role of recycling to supply the expansion of the LIBs market (Lähdesmäki et al., 2023). China and the United States, where LIBs sales are expected to exceed 18 million by 2030, rely strongly on import of raw materials for LIBs manufacturing and would benefit from circular management of resources (Shafique et al., 2022). Given the foreseen shift in market shares of various LIB chemistries toward batteries with lower cobalt content (Tang et al., 2021), the deployment of circular economy strategies is likely to reach the worldwide circularity target between 60% and 85% for cobalt by 2040 (Dunn et al., 2021) and reduce the amount of lithium discarded as waste from 75 to 85% down to 7–47% (Lähdesmäki et al., 2023). Additionally, recycling LIBs can provide significant economic benefits by recovering secondary materials, indeed the copper, aluminium and manganese recovered through LIBs recycling will hold economic values of 7.9, 4.4 and 3.9 billion US dollars (Shafique et al., 2023).

Life cycle assessment (LCA) was extensively applied to LIBs to evaluate the environmental performances of newly developed technologies (Raugei and Winfield, 2019), to compare their lifecycle impacts with other energy storage systems (Terlouw et al., 2019), and to compare different manufacturing (Qiao et al., 2017) or recycling (Golroudbary et al., 2019) processes. The literature data describing the environmental impacts of the different items of LIBs' chain are as follows. Material extraction, transport, and processing have been considered accountable for GHG emissions ranging 96–107 kg CO_2/kWh (Hao et al., 2017), 110 kg CO_2/kWh (Peters et al., 2017), 157 kg CO_2/kWh (Ellingsen et al., 2017), and 150–200 kg CO_2/kWh (Romare and Dahllöf, 2017). While manufacturing is considered the most impacting phase in the whole battery lifecycle (Ahmadi et al., 2017), −67% GHG emissions can be expected by relying on renewable energy (Delgado et al., 2019). The environmental impacts during battery use depend on the type of energy sources used for electricity production (Peters et al., 2017), with lower impacts related to renewable energy sources (Burchart-Korol et al., 2020). LIBs' recycling has lower environmental impacts compared to manufacturing from virgin metals (Bruno and Fiore, 2023; Hao et al., 2017), but higher energy demand and air emissions (Golroudbary et al., 2019). The literature analysis highlighted some knowledge gaps, as the lack of detailed data regarding the recycling processes (Ellingsen et al., 2017), and the fact that most studies focused exclusively on energy consumption and GHG emissions (Ellingsen et al., 2017; Golroudbary et al., 2019; Qiao et al., 2017) and rarely considered other aspects, as raw materials supply (Unterreiner et al., 2016) or metals' criticality (Terlouw et al., 2019).

While intensification of electric mobility is crucial to achieve decarbonification target of reducing GHGs emissions by 37.5% set by EU (Tang et al., 2023), recycling has been found to further limit GHGs emissions associated with LIBs (Aichberger and Jungmeier, 2020; Lai et al., 2022). Previous research pointed out the necessity to combine material flow analysis for circularity evaluation with LCA to thoroughly investigate the environmental implications connected circular economy strategies for LIBs management (Picatoste et al., 2022).

To our knowledge, there isn't any previous study performing a systematic analysis of the environmental impacts of the overall LIBs' supply chain at full-scale in a large context as Europe, considering manufacturing (comparing primary and secondary raw materials obtained from recycling), and accounting the impacts of LIBs' production based on secondary raw materials. This study has three main elements of novelty and aims: 1. a systematic and updated analysis of state-of-the-art recycling technologies applied at full-scale, emphasising the technological advancements and limitations; 2. the evaluation of the material flows involved in European LIBs' supply chain, considering the raw materials' demand for the LIBs needed in 2030 according to the legislation targets (35% EVs in the European vehicles' fleet), compared to LIBs' production and recycling capacities at full-scale; 3. the assessment of the environmental impacts comparing the use of primary raw materials and of secondary raw materials deriving from LIBs' recycling. This study will try to answer two key questions: (i) will the European LIBs' supply chain be ready to provide raw materials for the 35% circulating EVs forecasted for 2030? And (ii) what will be the environmental impacts associated to employing the secondary raw materials deriving from LIBs' recycling, compared to primary raw materials, mostly mined and refined outside EU? This study was developed through consequent phases, as follows. Firstly, based on the assessment of up-to-date recycling technologies and on the inventory of LIBs' production and recycling plants existing in Europe in 2020, the requested material flows of valuable metals (aluminium, cobalt, copper, iron, lithium, manganese, and nickel) have been estimated. Secondly, the environmental impacts (global warming potential, acidification potential, eutrophication potential and human toxicity) associated to the mining activities of primary metals required by European production plants have been calculated. Finally, the treatment potential of European recycling plants compared to the raw materials' demand estimated in the previous phases was assessed, and the environmental impact (as global warming potential) associated to the use of secondary metals obtained from recycling and of primary metals have been compared.

2. Methodology

This study was based on a four-phase methodology (Fig. 1). Initially, a literature review was conducted to identify the most applied LIBs recycling technologies at full-scale. Then, an inventory of LIBs recycling plants in Europe was compiled in order to assess the current European recycling capacity. Data from the literature review and from our inventory were combined into a material flow analysis to calculate the amount of metals (Al, Co, Cu, Fe, Li, Mn, Ni) potentially recovered from end-of-life batteries in 2030 and compare it with the material demand set by EVs' fleet in Europe in the same year. Eventually, the environmental assessment of recycling processes applied at full-scale was performed to calculate the GHGs emissions generated by recycling processes and the GHGs emissions avoided by recovery of secondary raw materials from recycling instead of primary raw materials mining.

More in details, the four-phase methodology of this study was as follows.

1. The literature review was conducted on Scopus using the keywords "lithium-ion battery recycling" AND "waste batteries characterisation", "thermal treatment", "pyrometallurgy", "hydrometallurgy", "mechanical treatment" in various combinations. Only research and review articles published in 2011–2023 have been considered. After a pre-screening based on abstract and highlights, the selected references have been inventoried and categorized according to key research topics. The recycling technologies have been considered as sequence of discharge, disassembly, physic-mechanical/thermal pretreatments, concluding with pyrometallurgy and hydrometallurgy as alternatives.
2. The LIBs' production and recycling plants existing in 2020 in the 27 members of the European Union (EU), Switzerland and United Kingdom (UK) were inventoried. When battery production capacity was expressed as energy (kWh/y and GWh/y); a characteristic average energy density of 0.15 MWh/t (Stura and Nicolini, 2006) has

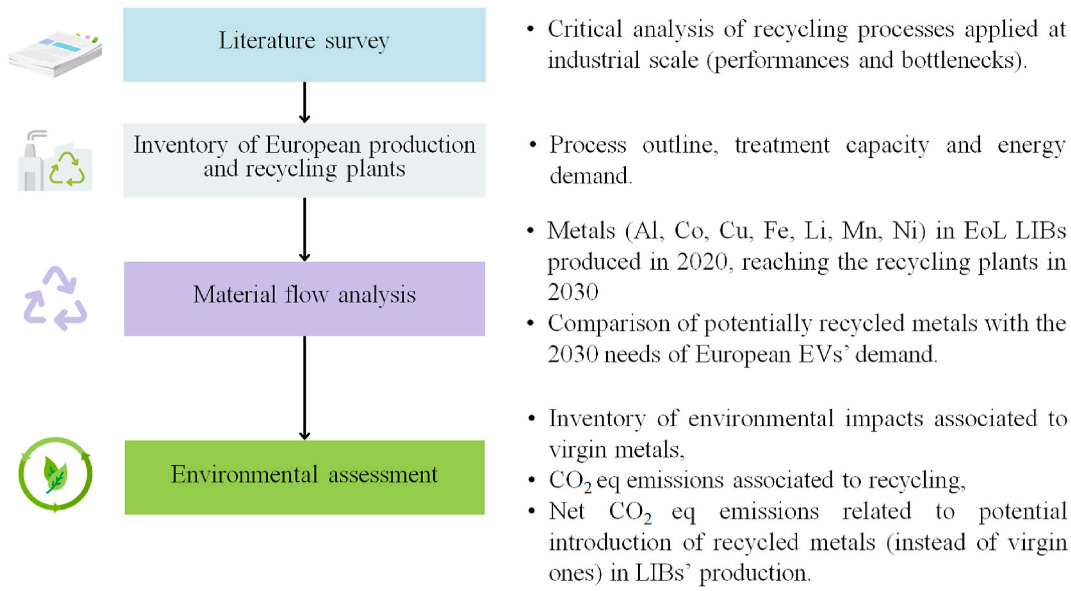

Fig. 1. Outline of the applied methodology (EoL: End of Life, LIBs: Lithium Ion Batteries, EVs: Electric Vehicles).

been accounted to estimate the amount (t/y) of produced and recycled LIBs.

3. The material flow analysis (MFA) of the metals (aluminium, cobalt, copper, iron, lithium, manganese, and nickel) involved by commercial LIBs' was electrodes based on the estimated EVs' demand in Europe according to the targets set by the European legislation (35% of circulating vehicles in 2030) (IEA, 2021), and comparison with the estimated European LIBs' capacity for production and recycling obtained in phase 2. This study involved the above-mentioned metals, as they are recovered at full scale (Mossali et al., 2020). The MFA was based on market shares of different electrodes chemistries forecasted for 2030 and their composition (Gaines et al., 2018; Statista, 2021), and on the average efficiency of recycling processes obtained in phase 1. The metals' recycling potential has been calculated for the recycling plants inventoried in phase 2, based on recovery efficiencies obtained in phase 1 and on market shares in 2020 (Pillot, 2021; Statista, 2021), considering that LIBs entering the recycling facilities were produced 8–10 years earlier (Bruno and Fiore, 2023).
4. The environmental assessment of European LIBs' supply chain was conducted, comparing the use of primary metals and of secondary raw materials deriving from LIBs' recycling. The net GHG emissions (CO_2 eq) associated to recycling were calculated considering the difference between the avoided emissions due to mining activities (calculated for metals' demand related to LIBs' market shares forecasted in 2030), and the emissions associated to recycling (calculated for metals' amounts according to LIBs' market shares in 2020). The specific environmental impacts accounted for mining activities for "primary" virgin metals' extraction and refining (Supplementary Materials, table I) was based on data retrieved from LCA studies (e.g., global warming potential, acidification potential, eutrophication potential and human toxicity). The references in table I have been selected based on accounting all mentioned impact categories for the metals considered in this study. Lithium, nickel and copper mining and processing may involve different routes (Flexer et al., 2018); to apply a conservative approach, this study considered the route associated to the highest environmental impacts. The environmental assessment of recycling reported the GHG emissions -i.e., the global warming potential, the only impact category available in all selected references. GHG emissions due to recycling have been calculated by combining specific coefficients for energy demand and CO_2 eq emissions (retrieved from literature) with the total energy demand of the full-scale recycling plants inventoried in phase 2. Specifically, the overall impact of recycling has been estimated, with a bottom-up approach, as sum of the impacts associated with the routes implemented at full-scale (e.g., mechanical pre-treatments, pyrometallurgy and hydrometallurgy) on LIBs sold in 2020 and recycled in 2030, considering the material flows undergoing each treatment and the associated energy demand. An additional contribution of 1.65 t CO_2/t has been accounted for pyrometallurgy, due to GHG emissions related to the combustion of batteries components (Hu et al., 2021a). The energy demand coefficients were: 4.50 kWh/t for mechanical pre-treatment (Wuschke et al., 2019), 1.08 kWh/t for pyrometallurgy (Hu et al., 2021b), and 1.65 kWh/t for hydrometallurgy (Romare and Dahllöf, 2017).

GHG emissions associated to recycling have been calculated through

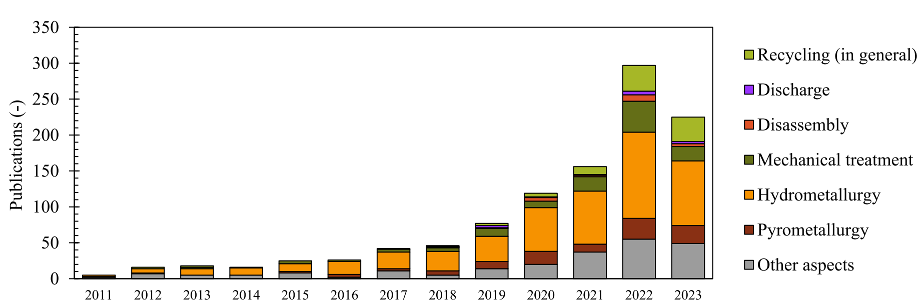

Fig. 2. Categorization of selected references according to year of publication and research topics.

eq. (1) considering the energy production specific impact (I_{energy}) for each country (EEA, 2021) hosting the recycling facilities: 0.207 kg CO_2/kWh for Belgium, 0.111 kg CO_2/kWh for Finland, 0.054 kg CO_2/kWh for France, 0.406 kg CO_2/kWh for Germany, 0.013 kg CO_2/kWh for Sweden and 0.250 kg CO_2/kWh for United Kingdom; lacking more specific data for Switzerland and Norway, the average European value 0.275 kg CO_2/kWh (EEA, 2021) was considered. GHG emissions due to recycling were accounted for as 1.65 t CO_2/t of battery for pyrometallurgy (Hu et al., 2021b), and 1.46 kg CO_2/t of battery for hydrometallurgy (Romare and Dahllöf, 2017). The GHG emissions associated to mechanical pre-treatment have been calculated accounting the GHG emissions associated to the energy mix of the country hosting the plant. As a comparison, the GHGs emissions had been calculated a second time using the emissions factors from Ecoinvent database v. 3.9.1 (Ecoinvent 3.9.1, 2023) applying the LCIA method R eCiPe 2016 v1.03, midpoint (H).

cathodes and decrease for LCO (38%) and LFP (30%). According to the results of phase 1, LIBs' composition exhibited high variability depending on cathode chemistry and manufacturer (Gaines et al., 2018) (Supplementary Materials, table II). The average weight of a LIB pack has been considered 318 kg (Iclodean et al., 2017). The average LIBs' composition for different battery chemistries accounted for in phase 3 was: 2.3–2.5% wt. for plastic compounds (polypropylene, polyethylene and polyethylene terephthalate), 14–22% wt. for electrolyte ($LiPF_6$, ethylene carbonate and dimethyl carbonate), 2.7–3.6% wt. for binder (polyvinylidene fluoride), 29.5–38.9% wt. for anode (copper current collector and graphite) and 39–48% wt. for cathode (aluminium current collector, and Al, Co, Fe, Li, Mn, Ni and P). According to the electrodes chemistries and their market shares, this study considered the following materials demands: 7.94 kg/t of Al, 6.35 kg/t of Co, 15.65 kg/t of Cu, 2.62 kg/t of Fe, 2.08 kg/t of Li, 3.43 kg/t of Mn and 5.81 kg/t of Ni in 2020 and 7.97 kg/t of Al, 4.58 kg/t of Co, 15.69 kg/t of Cu, 1.82 kg/t of Fe, 2.16 kg/t of Li, 3.27 kg/t of Mn and 8.64 kg/t of Ni in 2030.

$$\begin{aligned}\Sigma\ GHGs\ emissions\ [t\ CO2\ eq.] = \{\ mechanical\ pre-treatment\ [t]\cdot 4.50\ [kWh/t]\cdot emissions\ [t\ CO2\ eq.\ /\ kWh]\}\\ +\{\ pyrometallurgy\ [t]\cdot 1.08\ [kWh/t]\cdot emissions\ [t\ CO2\ eq.\ /\ kWh] + pyrometallurgy\cdot 1.65\ [t\ CO2\ eq./t]\}\\ +\{\ hydrometallurgy\ [t]\cdot 1.46\ [t\ CO2\ eq.\ /\ kWh]\ \}\end{aligned} \tag{1}$$

3. Results and discussion

3.1. Overview of literature review

264 references have been selected, out of 1069 found during the literature survey, according to phase 1 of the applied methodology, 94% scientific articles and 6% reviews, 71% published from 2019 to 2023. In overall, the scientific interest towards LIBs' recycling increased exponentially in the last decade. The results of the literature survey were categorized according to their main topic and the trend of different topics over time is displayed in Fig. 2. Besides, according to our survey of existing literature, the temporal evolution of recycling studies dedicated to a specific battery type is displayed in Fig. 3a and the intersection between the most studied battery types and the most popular recycling topics is displayed in Fig. 3b. The research was mostly focused on hydrometallurgy (53% of references), pyrometallurgy (9% references), pre-treatments (e.g., discharge, disassembly, physic-mechanical/thermal processes) (16% of references) and general recycling (8% of references), while material flow analysis of virgin raw materials and of secondary raw materials for LIBs (6%), LCA studies (6%) and economic analysis (1 %) of battery recycling were less considered. Most studies (50%) evaluated the recycling of NMC cathodes (Fig. 3a), and less (24-18%) of LFP and LCO. Studies on pre-treatments (discharge and disassembly) didn't specify cathodes' chemistries, while studies on hydrometallurgical (63%) or pyrometallurgical (15%) recycling involved specific cathodes' types (Fig. 3b). It should be noticed that in the last decade the interest of the scientific community about LIBs' recycling was primarily oriented towards the development and optimization of recycling processes, particularly hydrometallurgy applied to NMC cathodes, while issues as material flow analysis and economic assessment remained rather unexplored.

3.2. Waste electrodes' features

The market shares of different batteries resulting from phase 1 and applied in phase 3 were derived from literature (Lebedeva et al., 2017): 26% LCO, 23% LFP, 12% LMO, 10% NCA and 29% NMC in 2020, and 16% LCO, 16% LFP, 10% LMO, 10% NCA and 48% NMC in 2030. It may be observed that the market shares of different batteries significantly varied in 2020–2030, implying 66% increase in the demand of NMC

3.3. Performances of lithium-ion batteries' recycling processes applied at full-scale

3.3.1. Discharge

Discharge is a key pre-treatment for EoL LIBs, as residual energy may pose safety issues related to fire and explosion hazard (Wang et al., 2022a). Existing literature on LIBs pre-treatments often neglects details about cell discharging, indeed experimental activities are carried out on cells already discharged (Sunil and Dhawan, 2019) or directly on separated cathodes (Dolotko et al., 2020; Vanderbruggen et al., 2021; Xie et al., 2021). Nonetheless, discharge processes, reducing residual voltage below the threshold of 2 V, ensure safe batteries handling during recycling. Discharge methods can be categorized into: physical discharge, by connection to an ohmic resistance (Ku et al., 2016; Pinegar and Smith, 2019; Widijatmoko et al., 2020a, 2020b), or chemical discharge, by soaking the cells in aqueous solutions, most commonly NaCl with concentration 5% mass for 48 h (Liu et al., 2021; F. Wang et al., 2018; Yu et al., 2018; G. Zhang et al., 2019, 2018b) or 10% mass for 24 h (Fu et al., 2021; Zhang et al., 2013). However, NaCl presents several drawbacks such as galvanic corrosion of the cell casing and potential release of chlorine gas (Kim et al., 2021; Rouhi et al., 2022). Corrosion occurring during discharge with halide salts or $Na_2S_2O_3$, could be avoided or limited by recurring to different salt solutions such as NaOH, K_3PO_4 (Shaw-Stewart et al., 2019) or $MnSO_4$, which prevent organic leakage, however presents only mild discharge rate (Xiao et al., 2020). Corrosion effects are limited with zinc acetate as conductive solution for discharge (Fang et al., 2022). Eventually, metals salts, such as $FeSO_4$ or $ZnSO_4$ are less corrosive and increase the conductivity of the discharging solution, by release of metals ions (Ojanen et al., 2018). $FeSO_4$ in particular has been found to be environmentally friendly solution in terms of contamination of discharging solution and gaseous emissions (Yao et al., 2020). Ultrasounds improved chemical discharge (Torabian et al., 2022).

Physical discharge is based on discharge capacity of iron or graphite powders (Yao et al., 2020), but voltage rebound may happen, hindering safety (Rouhi et al., 2021; Yao et al., 2020). In conclusion, both physical and chemical discharge processes are applied, although an increasing interest for chemical discharge is happening; the existing knowledge gaps are related to solving safety and corrosion issues and investigating discharge behaviour and gas emissions.

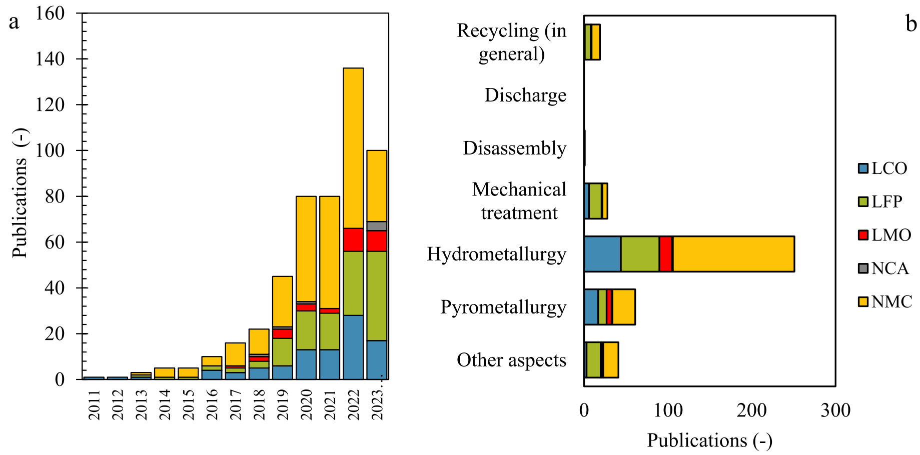

Fig. 3. Categorization of selected references according to (a) cathodes' chemistry and (b) research topics.

3.3.2. *Disassembly*

Disassembly is usually a manual operation entailing high operative costs, ranging from 32 €/kWh to remove a battery from a car to 76 €/kWh for a complete disassembly of modules into cells (Rallo et al., 2020). Compared with automatic shredding, manual disassembly provides higher purity in separated material streams, thus enhancing efficiency and safety of the following recycling steps (Marshall et al., 2020; Werner et al., 2022) and economic benefits (Zhao et al., 2022). Battery disassembly requires removing the plastic casing: automatizing partial disassembly (e.g., casing removal and cells recovery from battery packs) gave positive costs-benefits trade-off (Alfaro-Algaba and Ramirez, 2020); using a hybrid workstation (manually operated) resulted as best option for safety and costs (Tan et al., 2021). New battery designs are necessary to improve agile disassembly, as well as disassembly lines adaptable to different cells' configurations (Stief et al., 2019). Recently, the application of artificial intelligence and sensors to identify battery types and components to improve disassembly and sorting received attention (Lu et al., 2022; Meng et al., 2022). In conclusion, disassembly in full-scale recycling plants is mostly carried out manually due to high variability of cells' design, whereas recent literature explored (semi-) automatic processes. The existing knowledge gaps are related to the need of improving automation to reduce operating costs without affecting efficiency and safety of battery disassembly.

3.3.3. *Thermal and physic-mechanical pre-treatment processes*

Disassembled components undergo thermal and/or physic-mechanical processes depending on consequent recycling process (Kim et al., 2021), with the overall aim of separating active materials from current collectors and/or eliminating binder and electrolyte (Zhang et al., 2020a; Lombardo et al., 2021), and improving metals' leaching in hydrometallurgy (Gu et al., 2022; Ahn et al., 2022; Ma et al., 2022a). Thermal pre-treatments have been applied in a wide range of temperatures, from 200 °C (Li et al., 2022), to 700 °C (Nayaka et al., 2018). At 550–600 °C, thermal pre-treatment has been combined with ball milling (Lombardo et al., 2021), or with ultrasounds (Yan et al., 2022), or froth floatation (Vanderbruggen et al., 2022; Zhan et al., 2021). At higher temperatures (600–1000 °C) carbothermal reduction increased cobalt, manganese, and nickel leaching (Pindar and Dhawan, 2020a).

Physic-mechanical pre-treatments, applied to disassembled batteries to obtain the "black mass", are crushing and ball milling (Peng et al., 2018; Takahashi et al., 2020), followed by separation via sieving, centrifugation, floatation, magnetic, Eddy current or pneumatic classifiers (Zhan and Pan, 2022; Zhang et al., 2018a). Ball milling was combined with magnetic and densimetric separations (Da Costa et al., 2015), and with sieving (Pinegar and Smith, 2019). In overall, physic-mechanical pre-treatments as ball milling (Shen et al., 2019), pneumatic (Bi et al., 2019a; Bi et al., 2020; Zhong et al., 2020), magnetic (Huang et al., 2022a; Hu et al., 2022) and Eddy current separations (Bi et al., 2019b) improved the performances of metals' leaching in hydrometallurgy. Magnetic separation is highly efficient to recover cobalt if preceded by thermal treatments (Li et al., 2022; Peng et al., 2021) or carbothermal reduction (Vishvakarma and Dhawan, 2019).

Concerns about safety of thermal and physic-mechanical treatments have been raised (Lewandowski et al., 2020; Ross et al., 2020), particularly about the release of hazardous volatile components (Diaz et al., 2019; Wuschke et al., 2019; Huang et al., 2022b), requiring adsorption on activated carbon (Stehmann et al., 2017). Energy required for physic-mechanical pre-treatments was 4.50 kWh/t (Wuschke et al., 2019).

Less energy-consuming pre-treatment processes have been developed, as exfoliation of active electrode components from current collectors (Chen et al., 2017a; He et al., 2019, 2020), also based on hydrogen released from aluminium current collector put in contact with water (He et al., 2020). Conversely to recycling treatments, mostly focused on material recovery from cathodes, separation of active materials from current collectors recently drawn attention on both cathodes and anodes (Chen et al., 2017a; He et al., 2021). Effective separation of active materials is essential to ensure high performance standards in subsequent recovery steps; low amounts of manganese (Weng et al., 2013), aluminium and copper (Peng et al., 2020; Zhang et al., 2020b) weren't critical, however lithium impurities (Jo et al., 2018), aluminium in concentrations between 0.02 and 1.48 g/L (Krüger et al., 2014; Zhang et al., 2020c) and copper ranging 4.8 and 91.5 mg/g (Peng et al., 2020) severely hurdled the electrochemical performances of recycled materials. Hydrophobicity of graphite from LIBs' anodes is often exploited by applying separation through froth floatation (Verdugo et al., 2022, 2023).

In conclusion, conventional thermal and physic-mechanical pre-treatments are still commonly applied, even if novel technologies based on exfoliation are gaining attention. The existing knowledge gaps are mostly related to the need of improving efficiency of the separation of active materials from current collectors and to lower energy demand.

3.3.4. *Pyrometallurgy*

Thermal treatments can be applied not only as pre-treatments, but also to recover valuable metals such as cobalt, manganese and nickel (He et al., 2023; He et al., 2021) often without prior pre-treatments (Karabelli et al., 2020). It should be mentioned that pyrometallurgy convert the black mass into metal oxides or alloys (Assefi et al., 2020), and that further hydrometallurgy (see section 3.3.5) is needed to obtain single metals. Smelting takes the black mass above the melting point of metals (1250–1500 °C), allowing to separate the reduced components (Makuza et al., 2021; Windisch-Kern et al., 2021). Recovery yields obtained from smelting are 98% for cobalt and nickel, and 85–92% for manganese (Hu et al., 2021a, 2021b). Thermal treatments at lower temperatures (650–1000 °C) also happen, as pyrolysis (Makuza et al., 2021; Pindar and Dhawan, 2020b), and incineration (Lombardo et al., 2021). Pyrolysis has been found to improve electrode active materials separation efficiency for 83–99% in cathodes, and for 88–97% in anodes (Zhang et al., 2022). High temperature pyrolysis (>950 °C) has been able to recover up to 99% nickel from NMC cathodes (Ebin et al., 2019). Pyrolysis of black mass has different effects depending on cathodes' chemistries: NMC are converted into oxides (Co_3O_4, NiO, Mn_3O_4) and Li_2CO_3, with release of CO, CO_2, and HF from the binder (Lombardo et al., 2021); LCO into metallic cobalt and lithium carbonate (Lie et al., 2020), releasing most lithium (64–97%) and phosphorous (68%) as gases (Windisch-Kern et al., 2021). Carbothermic reduction triggers volatilisation of corrosive gas due to decomposition of polypropylene separator and polyvinylidene fluoride binder (Lombardo et al., 2021). The temperature range 470–599 °C has been associated with highest material loss rates, while release of fluorine decreased with temperature, with no HF released above 470 °C (Yu et al., 2020). Few studies preliminarily investigated lithium recovery through carbothermal reduction (Rostami et al., 2022; Zhang et al., 2022) or sulphation roasting (Biswas et al., 2023) followed by water leaching, and of lithium and phosphorous from gaseous emissions (Holzer et al., 2022). Energy required for pyrometallurgy was 1.08 kWh/t (Hu et al., 2021b).

Recovery rates associated with pyrometallurgical processes resulting from literature (Table 1) are on average 99% for aluminium, 96% for copper, 86 ± 15% for cobalt, 88 ± 4% for manganese and 98 ± 1% for nickel, iron and lithium are not recovered. In conclusion, pyrometallurgy allows recovery with high efficiency mostly NMC cathodes' metals, with few references related to aluminium and copper from current collectors, while lithium and iron recovery are not involved. Production of gaseous corrosive and hazardous fractions during thermal processes should be carefully managed.

Table 1
Recovery efficiencies of pyrometallurgy processes applied to Lithium-Ion Batteries.

Co	Mn	Ni	Al	Cu	Reference
97.90%	91.50%	97.70%	–	–	Hu et al. (2021a)
98.20%	85.30%	98.40%	–	–	Hu et al. (2021b)
68.00%	–	–	–	–	Pindar and Dhawan (2020b)
–	–	–	99.34%	96.25%	Zhong et al. (2019)
81.30%	–	–	–	–	Ruismäki et al. (2020)
–	–	99.00%	–	–	Ebin et al. (2019)
95.6%					Li et al. (2022)
86.35% ± 15%	88.40% ± 4%	98.37% ± 1%	99.34%	96.25%	average values[a]

[a] Standard deviation was detailed when at least 3 values were available.

3.3.5. Hydrometallurgy

Hydrometallurgy consists in leaching valuable metals from cathodic active materials in acidic environment (Larouche et al., 2020), with various available routes (Ma et al., 2020). Leaching usually happens after mechanical (Diaz et al., 2020; Takahashi et al., 2020) or thermal (Chen et al., 2020; Fu et al., 2020; Vieceli et al., 2021; Zhang et al., 2020b) pre-treatments, using acidic solvents at temperatures between 40 °C (Chen et al., 2017a; He et al., 2017; Jian et al., 2020) and 100 °C (Chen et al., 2018, 2020; Musariri et al., 2019; Wu et al., 2020), and also at room temperature (20–25 °C) (Takahashi et al., 2020). Most common leaching agent is sulfuric acid (Chan et al., 2020; Chen et al., 2018; Diaz et al., 2020; Dutta et al., 2018; He et al., 2017; Sattar et al., 2019; Urbańska, 2020; Vieceli et al., 2021; Zhao et al., 2020), followed by hydrochloric acid (Guo et al., 2016; Jian et al., 2020; Xu et al., 2020), phosphoric acid (Chen et al., 2020; Meng et al., 2017; Zhuang et al., 2019), and organic acids, mostly citric (Chabhadiya et al., 2021; Patil et al., 2020; Wu et al., 2020; Xing et al., 2021), alginic (Cai et al., 2022), benzenesulfonic (Fu et al., 2019), formic (Chen et al., 2021; Gao et al., 2017), gluconic (Fan et al., 2020) and L-tartaric (Ma et al., 2022) acids. Leaching agent's concentrations are 1–2 M for organic and inorganic acids except for sulfuric acid, occasionally exceeding 5M (Fan et al., 2021; Zhao et al., 2020). Organic acids required higher process temperatures and lower solid/liquid ratio compared to inorganic acids; contact time is similar (60–120 min) for all acids, while citric and ascorbic acids required longer times (Nayaka et al., 2018). Leaching is favoured when metals are oxidised, thereby reducing agents are often added to increase hydrometallurgy efficiency (Nicol, 2020). The most used is hydrogen peroxide, in concentration equal to 1.5–2% vol. (Chabhadiya et al., 2021; Musariri et al., 2019) and 4–5% vol (Chen et al., 2017b; Sattar et al., 2019; Shin et al., 2019). Other common reducing agents are glucose (Chen et al., 2018; Meng et al., 2017), starch (Jian et al., 2020), orange peel powder (Wu et al., 2020), ethanol (Zhao et al., 2020), citric acid (Zhuang et al., 2019) and glutaric acid (Jian et al., 2020). Full details about the experimental conditions (leaching agent, concentration, solid-to-liquid ratio, contact time, reducing agent) adopted in the selected references are in Supplementary Materials, table III.

Post leaching, many routes have been explored to recover metals from the leachate: solvent extraction (Nadimi and Karazmoudeh, 2021; Shen et al., 2019b), also in multi-step processes (Nguyen and Lee, 2021; Yang et al., 2020), or followed by crystallization (Sattar et al., 2019; Djoudi et al., 2021); deep eutectic solvents (DES) (Tran et al., 2019; Wang et al., 2019; Schiavi et al., 2021; Zante et al., 2020), supercritical extraction in water (Lie et al., 2020) or carbon dioxide (Rothermel et al., 2016); ion-exchange resins (Virolainen et al., 2021) and sequential application of chelating and cation-exchange resins (Chiu and Chen, 2017); ionic liquids (Zante et al., 2020; Schaeffer et al., 2020); precipitation (Zhang et al., 2020b; Chu et al., 2020; Yang et al., 2020; Peng et al., 2019b), and co-precipitation (Zhao et al., 2020; Swain, 2018; Guo et al., 2017; Beak et al., 2021). Recovery of Li and Co proved to be effective in recovering materials with purity grades appropriate to ensure electrochemical performances comparable to commercially available electrodes (Jo et al., 2018). Impurities (0.05–0.006 mol/L) of iron and copper improved cobalt leaching efficiencies (Peng et al., 2019a). The energy required for hydrometallurgy was accounted 1.65 kWh/t (Romare and Dahllöf, 2017).

In conclusion, compared to pyrometallurgy (Table 1), hydrometallurgy average efficiencies calculated from literature (Table 2) are extremely high (71 ± 34% for Al, 91.7 ± 12% for Co, 91.5 ± 13% for Fe, 96.2 ± 5% for Li,88.4 ± 19% for Mn and 90.1 ± 15% for Ni), and more metals are involved (i.e., iron, lithium). However, critical issues are significant energy required, numerous post-leaching phases, and application of toxic solvents. Recent literature is shifting towards more environmentally friendly leaching agents and downstream processes, adopting lower temperatures and milder (possibly organic) acids in safe conditions.

3.3.6. Challenges and future opportunities

There are still many challenges associated with LIBs recycling. First of all, the complexity of accurately sorting different battery chemistries during collection limits the technological feasibility of materials recovery. Moreover, safety concerns arise from the initial process of discharge, due to the potential release of harmful gasses and potential hazards posed by voltage rebound, which may limit safety during the subsequent disassembly step. During disassembly, the presence of hazardous chemicals in batteries' electrolytes and binders, as well as the ignition risk caused by residual voltage, pose a risk to workers' safety. Following dismantling and separation of the batteries' casings, the electrodes are subject to pre-treatments aimed at concentrating the black mass from other less valuable components. Pre-treatments consume a lot of energy demand and have still limited efficiency. Metallurgical recycling of LIBs is usually performed by

Table 2
Recovery efficiencies of hydrometallurgy processes applied to Lithium-Ion Batteries.

Leaching agent	Co	Mn	Ni	Al	Li	Fe	Reference
Hydrochloric acid					99.4		Guo et al. (2016)
					97		Yang et al. (2020)
	97.25				97.72		Xing et al. (2021)
					95	95	Liu et al. (2022)
					98		Ilyas et al. (2022)
Phosphoric acid	99						Chen et al. (2017b)
	98				100		Meng et al. (2017)
					94.29	97.67	Yang et al. (2020)
	91.63	92.35	83.1		100		Zhuang et al. (2019)
					93.51	97.96	Wang et al. (2022b)
					97.72	98.24	Li et al. (2022)
	98.9	97.3	99.1		99.6		Zhou et al. (2023)
Sulfuric acid	70				99.1		Jha et al. (2013)
	66.2	50.2	96.3		93.4		Meshram et al. (2015)
	74	65	68				Tanong et al. (2016)
					96.85		Huang et al. (2022b)
	99.7	99.7	99.7		99.7		He et al. (2017)
	96				86		Pagnanelli et al. (2016)
	96	94	68				Tanong et al. (2017)
	97				99		Dutta et al. (2018)
	79.4	91.9	66.2			68.5	Sobianowska-Turek (2018)
	98				96		Chen et al. (2018)
	93.8				95.7		Peng et al. (2018)
	94.63				98.62		Jiang et al. (2018)
	93.2	90.3	91.5		80.2		Cheng et al. (2019)
	68	34.8			92		Sattar et al. (2019)
	99						Zhao et al. (2020)
	99.29	99.91	98.62		99.78		Wang et al. (2020)
	98.91				92.67		Ghassa et al. (2020)
	100	100	100		100		Chan et al. (2021)
	92.84	90.18	93.11				Wang et al. (2018)
	23.2	18.3	25.6		100		Fan et al. (2021)
	70	70	70		70		Vieceli et al. (2021)
	99				99		Aboulaich et al. (2022)
	90				90		Partinen et al. (2022)
		98.13			99.62		Chang et al. (2022)
	99				99		Kong et al. (2022)
	88.6				99.9		Jiang et al. (2022)
			99.9				Permatasari et al. (2022)
	97.24	96.88	99.46		99.79		Li et al. (2022)
	95	92	95		98		Jian et al. (2020)
	98.7	99.5			99.9		Natarajan et al. (2018)
	91.7	97.3	99.1		99.6		Zhou et al. (2023)
Acetic acid	94.61	97.97	96.39	94.7	98.56		Wang et al. (2022b)
	90				99		Prasetyo et al. (2022)
Alginic acid	97.58				98.59		Cai et al. (2022)
Ascorbic acid	94.8				98.5		Li et al. (2012)
	90						Nayaka et al. (2018)
	99.56	99.87	99.6		99.69		Chen et al. (2018)
	96.53				99.58		Fu et al. (2019)
Benzenesulfonic acid	95	94	97		99		Fu et al. (2019)
Citric acid	96				99		Gao et al. (2019)
					92		Golmohammadzadeh et al. (2017)
	95				99		Chen and Zhou (2014)
	95				95		Musariri et al. (2019)
				47.24	94.83		Kumar et al. (2020)
	90	98	98		94		Pindar and Dhawan (2020b)
	90	89	94		91		Meng et al. (2017)
	96		96		100		Esmaeili et al. (2020)
	96.1		97.2		94.1		Nakajima et al. (2022)
		94			94		Wang et al. (2022b)
	95.6	94.9	90.7		98.3		Choi et al. (2022)
	85				85		Patil et al. (2020)
DL-malic acid	94.3	96.4	95.1		98.9		Sun et al. (2018)
	90				90		de Oliveira Demarco et al. (2019)
	97.6	97.3	97.8		98		Ning et al. (2020)
	98.86				98.13		Zhou et al. (2021)
	90.58	98.66	90.14		98.53		Wang et al. (2022b)
	97.1	97.6	96.2		98.1		Cheng et al. (2022)
	95	95	95		95		Sidiq et al. (2022)
Formic acid					99.93		Gao et al. (2017)
Gluconic acid	95	95	95		95		Ersha Fan et al., 2020
L-tartaric acid	98.5	98.5	98.5				Ma et al. (2022)
Maleic acid	98	98	98		95		Fan et al. (2020)

(continued on next page)

Table 2 (*continued*)

Leaching agent	Co	Mn	Ni	Al	Li	Fe	Reference
Oxalic acid	97				98		Zeng et al. (2015)
					98.9		Rouquette et al. (2023)
Trifluoroacetic acid	91.8	89.8	93		99.7		Zhang et al. (2015)
Average	91.7 ± 12%	88.4 ± 19%	90.1 ± 15%	71.0 ± 34%	96.2 ± 5%	91.5 ± 13%	

pyrometallurgical processes, which aim primarily to recover Co, Ni and Mn as metals alloys and requires careful management of corrosive gases and hazardous fractions released during thermal processes. Alongside pyrometallurgy, novel recycling plants operate hydrometallurgical recycling in order to expand material recovery also to lithium, which is lost as slag during pyrometallurgy. However, the use of hazardous chemicals and the numerous post-leaching phases, raise environmental issues and render waste management challenging.

Safety and environmental sustainability could be improved along the whole LIBs recycling process and research should focus on achieving this target. For instance, the use of non-corrosive discharge solutions could limit the release of gaseous emissions and by avoiding galvanic corrosion they could increase the amount of metals recovered from batteries' casings, however the research on the topic is still limited. Automation of disassembly could reduce operative costs while minimizing workers' exposure to hazards, however improvements are still required to achieve efficiency comparable to manual disassembly. Pre-treatments are often overlooked by research, despite their importance in determining efficiency of following metallurgical processes. Eventually, future research should improve environmental sustainability of hydrometallurgical processes by limiting operative temperature, using milder organic leaching agents and simplifying materials recovery after leaching.

3.4. Analysis of European lithium-ion batteries' supply chain

3.4.1. Inventory of plants producing and recycling lithium-ion batteries

From selected literature (Lebedeva et al., 2017; Transport and Environment, 2019; Danino-Perraud, 2020; Larouche et al., 2020; Mossali et al., 2020; Romare and Dahllöf, 2017; Winslow et al., 2018), a list of European LIBs' production and recycling full-scale plants (existing and forecasted in 2016–2025) was compiled. 14 production plants (Supplementary Materials, table IV), initially corresponding to 2.5 GWh in 2016 (foreseen to reach 378 GWh also considering plants forecasted by 2025) were inventoried. In overall, European production plants display production capacity (2,525,893 t/y) adequate for the expected 2030 needs (2,257,800 t/y). LIBs' production capacity evolved in recent years; while in 2016 it was entirely concentrated in United Kingdom (2030) will see Germany (52.5%), Poland (20%), Sweden (12.7%) and Norway (8.4 %) as key players (Supplementary Materials, fig. I), accounting in overall for 2,433,333 t/y out of the 2,525,893 t/y of total European production capacity.

Considering LIBs' recycling capacity (Supplementary Materials, table V), 14 recycling facilities were identified, accounting in total for 55,810 t/y (Bruno and Fiore, 2023). In 2016, four countries were responsible for 93% of total European recycling capacity: Belgium (13%), France (37%), Germany (30%) and Norway (13%). Three key technologies have been identified: mechanical treatment, pyrometallurgy and hydrometallurgy. These can be combined, also with pre-treatments, as happens in plants managed by Umicore in Belgium and Accurec in Germany. Pyrometallurgy, eventually preceded by pre-treatment, is applied in 4 plants out of 14 (SNAM and Eramet in France, Akkuser in Finland, and Duesendeld in Germany). Hydrometallurgy, preceded by pre-treatment, is applied in 4 plants out of 14 (Redux and Lithorec in Germany, Batrec Industrie in Switzerland, AEA Technologies in the United Kingdom). Only two plants (Pilagest in Spain, and uRecycle in Sweden) apply a purely mechanical treatment. In conclusion, pyrometallurgy provides 35 % of European recycling capacity while hydrometallurgy 28%, their combination 21%, and mechanical treatment the remaining 16%.

3.4.2. Comparison of European lithium-ion batteries' production and recycling capacity with electric vehicles' demand

The demand of metals involved by commercial LIBs' cathodes based on the estimated EVs' demand - 35% of EVs in European vehicles' fleet by 2030 - in Europe was calculated according to average composition and market shares (Section 3.2). Being nickel the main component of NMC cathodes (5.81 kg of nickel/t of battery in 2020) (Gaines et al., 2018) and considering the forecasted market shares for 2030, its demand is expected to grow up to 8.64 kg/t of battery. LCO and LFP requests expected to decrease by 2030, and related amounts of cobalt and iron are expected to drop for cobalt (from 6.35 in 2020 to 4.58 kg/t of battery in 2030) and iron (from 2.62 in 2020 to 1.82 kg/t of battery in 2030). Changes in market share and composition from 2020 to 2030 are key issues, implying that LIBs produced in 2020 and reaching the recycling facilities in 2030 will be able to provide metals for a higher number of batteries. According to the results of this material flow analysis, the amount of different metals reaching the recycling infrastructure in 2030 and treated by pyrometallurgical and hydrometallurgical plants, is displayed in Fig. 4a; whereas, the amount of recovered metals, which could re-enter the LIBs supply chain in Europe as secondary raw materials, is displayed in Fig. 4b.

Among LIBs' components, lithium, cobalt, nickel, and manganese present the highest supply risk in Europe (European Commission, 2020b). While refining and manufacturing mainly happen in China, their mining activities are geographically diversified, mostly occurring outside Europe (Sun et al., 2019). Lithium is mainly extracted in Australia, Chile, and Argentina (106,200 t/y in total) (U.S.G.S., 2023), while in Europe it happens in Portugal (800 t/y) (Oliveira et al., 2015). Finland provides cobalt to Europe (2100 t/y) (European Commission, 2018), and the world leading producer is the Democratic Republic of Congo (130,000 t/y), followed by Indonesia (10,000 t/y) and Russia (8900 t/y)(U.S.G.S., 2023).Manganese is supplied by South Africa, Gabon, Australia and Ghana (16,400 t/y in total) (U.S.G.S., 2023), and by Ukraine, Hungary and Romania in Europe (700,200 t/y in total) (European Commission, 2018). Indonesia, Russia and the Philippines supply nickel (2,159,000 t/y in total) (U.S.G.S., 2023),and the United Kingdom, France, Italy, Finland, Poland and Greece (228,340 t/y in total) (European Commission, 2018); 50% of globally extracted aluminium (40,000 t/y out of 69,000 t/y globally) is mined in China, and 3% in Europe, in Norway and Iceland (2.150 t/y) (U.S.G.S., 2023). Global reservoirs of iron are located in Australia, Brazil, China and India (196,000 t/y), while in Europe mining of iron ore is concentrated in Turkey and Ukraine (57,000 t/y) (U.S.G.S., 2023). Copper is mostly mined in Russia, China, Congo, Peru and Chile (12,500 t/y in total), and less than 2% in Europe, in Poland (390 t/y) (U.S.G.S., 2023). In conclusion, metals' demand matching European EVs' fleet target for 2030 (IEA, 2021) is for lithium 2.16 kg/t of battery (4885 t/y in total), for cobalt 4.58 kg/t (10,338 t/y in total), for nickel 8.64 kg/t (19509 t/y in total), for manganese 3.27 kg/t (7393 t/y in total) and for iron 1.82 kg/t (17,986 t/y in total).

The plants producing the highest amounts of recycled metals (e.g., Eramet in France, Redux in Germany, and Umicore in Belgium) make the host countries key players of the game. In 2030, the European recycling capacity will allow to recover each year 278 t of aluminium (out of 17,986 t necessary to supply European EVs' market in 2030 (IEA, 2021)), 468 ± 69 t of cobalt (out of 10,338 t necessary), 531 t of copper

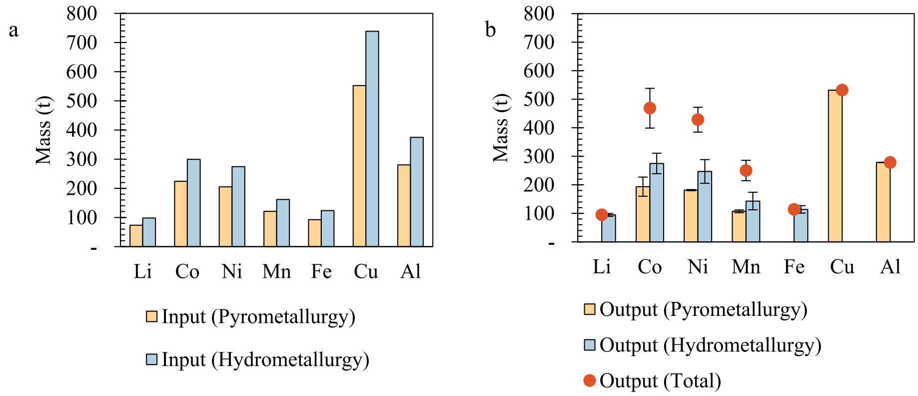

Fig. 4. Material flow analysis of European lithium-ion batteries' recycling infrastructure in 2030: (a) input and (b) output streams (red: total; yellow: pyrometallurgy; blue: hydrometallurgy). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

(out of 17,986 t necessary), 114 ± 13 t of iron (out of 7393 t necessary), 95 ± 5 t of lithium (out of 4885 t necessary), 250 ± 35 t of manganese (out of 7393 t necessary) and 428 ± 43 t of nickel (out of 19,509 t necessary).In total, the recycled metals provided by European recycling infrastructure will supply about 2165 t out of the 99,658 t of metals required by EVs' demand forecasted for 2030, corresponding to 2% of the demand.

European policies encourage the electrification of transportation networks to reduce GHGs emissions from transportation, while also requiring material circularity in the battery sector. However, our study indicates that the present European recycling infrastructure is still insufficient to satisfy the EU's collection and recovery targets. Similarly, the mandatory minimum levels of recycled content, which are set at 16% for cobalt, 6% for lithium and 6% for nickel could not possibly be met considering secondary raw materials provided exclusively from European recycling facilities. Increasing European recycling capacity will be essential in meeting regulatory requirements whilst securing the supply chain of critical raw materials mined outside of Europe and frequently connected with geopolitical tensions.

3.4.3. Environmental assessment

The environmental assessment was based on the comparison of environmental impacts of LIBs' production in Europe in 2030 according to two scenarios: 1. Production based entirely on primary metals; 2. Production based on secondary metals provided by European recycling infrastructure (section 3.4.2), topped up with primary metals. Even if the share of recycled metals supplied by European plants is 2% (corresponding to 2165 t of metals out of 99,658 t of metals required by European EVs fleet in 2030) of the necessary amount, investigating the environmental outcomes of such a contribution is interesting.

The calculation of environmental impacts (GWP: global warming potential, AP: acidification potential, EP: eutrophication potential, HTP: human toxicity potential) associated to mining activities providing the metals required by LIBs' manufacturing was based on specific impact coefficients retrieved from literature (Supplementary Materials, table I). Among several impact categories, GWP, AP, EP and HTP were selected due data availability for every metal considered in this study. To have a reference for comparison with GWP associated with European LIBs' supply chain, the GWP (along with AP, EP and HTP) ascribable to primary metals employed in global LIBs' production have been calculated (Supplementary Materials, fig. II). Total GWP associated to global LIBs' production in 2030 is equal to 491.07 t of CO_2 (36% due to aluminium, 20% to copper, 19% to nickel, 15% to cobalt and 7% to lithium), corresponding to 4,910,000 km by car (fed by gasoline) (European Environmental Agency, 2023). China, the leading LIBs' producer, is unsurprisingly responsible for the highest (44%, 16.5 kt of CO_2) share. Copper mining is the main contributor to the impact categories of eutrophication and human toxicity potential, whereas Nickel and Aluminium mining are mostly accountable for impacts related to global warming potential and acidification potential.

The amount of GWP generated by the recycling of 1 t of LIBs, with state-of-the-art full-scale recycling processes according to the results of our inventory, and the potential saving of emission from mining activities, based on the amount of secondary raw materials recovered from the recycling of 1 t of LIBs, is displayed in Fig. 5. Specifically considering the European supply chain, GWP due to mining of primary metals required by LIBs' production in 2030, based on specific impact coefficients retrieved from literature (Supplementary Materials, table I) and on results of phases 1–3 of the applied methodology, was calculated. GWP of recycling (1053.10 kg CO_2 eq/t) was decreased by avoiding CO_2 eq emissions (315.76 kg CO_2 eq/t) related to the mining of primary metals corresponding to the share of recycled metals calculated in section 3.4.2. GHG emissions balance showed that introducing recycled metals in European LIBs' supply chain could avoid 28% of GHGs

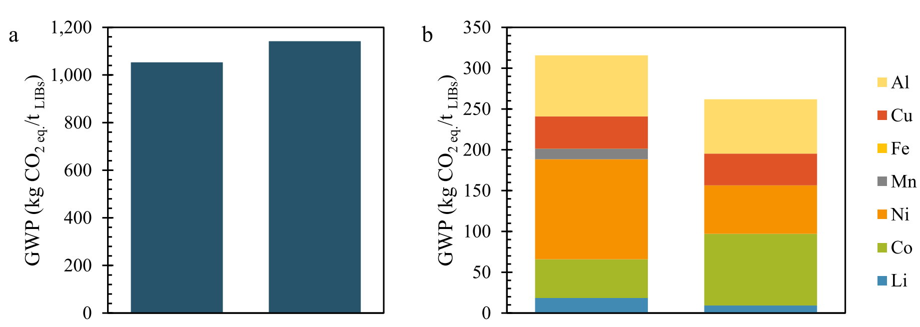

Fig. 5. GHG emissions associated to (a) lithium-ion batteries' recycling in Europe in 2030 and (b) avoided by employing the recycled metals (instead of primary metals) in the production of new batteries.

emissions. The trade-off was achieved by plants applying hydrometallurgy (1.46 kg CO_2/t of battery) (Romare and Dahllöf, 2017), as pyrometallurgy entails significant CO_2 eq emissions (1.65 t CO_2/t of battery) (Hu et al., 2021b) due to graphite incineration, and hydrometallurgy allows to recover lithium. Emissions associated with metals' mining and potentially saved by secondary materials exiting from recycling plants in 2030 have been calculated (data referred to 1 ton of recycled LIB): 3.34 17 kg CO_2 eq. for Al, 4.04 ± 0.8 kg CO_2 eq. for Co, 6.37 17 kg CO_2 eq. for Cu, 0.95 ± 0.16 kg CO_2 eq. for Fe, 1.17 ± 0.06 kg CO_2 eq. for Li, 2.86 ± 0.4 kg CO_2 eq. for Mn and 7.62 ± 0.5 kg CO_2 eq. for Ni. The highest saving is associated with Nickel, whose recycling avoids significant GWP impact and presents a high recovery rate.

In conclusion, the environmental benefits of electric vehicles, when compared to traditional internal combustion engines, are frequently overshadowed by the environmental impacts associated with mineral processing to extract the critical raw materials for LIBs manufacture. Recovery of secondary raw materials from LIBs recycling will pose an additional contribution to GHGs emissions throughout the whole life cycle. Nonetheless, recycling processes avoid mining of primary raw materials, which in the specific context of Europe, will reduce supply risks for raw materials required by future LIBs manufacturing.

4. Conclusions

The growing interest for LIBs recycling is driven by the necessity of securing the supply chain, affected by extensive employment of critical raw materials. LIBs recycling at full-scale needs to be carefully optimized across all phases. Further research related to pre-treatments should explore ionic solutions able to avoid galvanic corrosion, and the optimization of trade-off between higher precision of manual dismantling and lower costs of automatic disassembly systems. Metallurgical recovery of valuable metals was extensively investigated, obtaining high recovery efficiencies via pyrometallurgy (99% for Al, 86 ± 15% for Co, 96% for Cu, 88 ± 4% for Mn and 98 ± 1% for Ni), as well as hydrometallurgy (71 ± 34% for Al, 91.7 ± 12% for Co, 91.5 ± 13% for Fe, 96.2 ± 5% for Li, 88.4 ± 19% for Mn and 90.1 ± 15% for Ni), which allows to recover a wider range of metals, including Li and Fe. Future research should minimize their environmental impacts and economic costs, assuring the high technical performances.

Despite hydrometallurgy attracts most attention of the scientific community, pyrometallurgy still represents the dominant technology in current industrial plants. Considering the 14 recycling facilities inventoried in Europe by this study (55,810 t/y total recycling capacity), 35%-wt of EoL LIBs is destined to pyrometallurgy and 21%-wt to combined pyrometallurgy and hydrometallurgy. In 2030, European LIBs' production capacity will be adequate for forecasted needs. However, EU recycling plants will provide only 2%-wt of needed metals (278 t of Al, 468 ± 69 t of Co, 531 t of Cu, 114 ± 13 t of Fe, 95 ± 5 t of Li, 250 ± 35 t of Mn and 428 ± 43 t of Ni). This will avoid mining primary metals and considering the GHGs emissions associated with recycling (1053.10 kg CO_2 eq/t of battery), the net GHG emissions will be cut by 28%–737.34 kg CO_2 eq/t battery. In conclusion, further research should aim at decreasing the environmental impacts of full-scale recycling processes through revamping existing facilities based on processes associated to lower energy demand and GHG emissions. However, while the scientific community should put effort in overcoming the mentioned bottlenecks, the reinforcement and development of European recycling infrastructure is highly needed to secure the LIBs' supply chain.

CRediT authorship contribution statement

Martina Bruno: Conceptualization, Data curation, Investigation, Visualization, Writing – original draft. **Silvia Fiore:** Conceptualization, Methodology, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

The authors declare no conflict of interest. The research was funded by internal resources.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2024.120758.

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