



Review article

## A comprehensive review and classification of unit operations with assessment of outputs quality in lithium-ion battery recycling



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### HIGHLIGHTS

- Overview of capacities and outputs quality in lithium-ion battery recycling.
- Pyrometallurgy-based and hydrometallurgy-based example processes critically described.
- Unit operations classified and ranked according to industrial readiness.
- Hydrometallurgical capacity in EU must be enhanced to recover battery-grade materials.
- Outputs definitions and calculation rules for recycling efficiency need harmonization.

### ARTICLE INFO

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### ABSTRACT

Lithium-ion batteries (LIBs) are the core component of the electrification transition, being used in portable electronics, electric vehicles, and stationary energy storage. The exponential growth of LIB use generates a large flow of spent batteries which must be recycled. This paper provides a comprehensive review of industrial realities of LIB recycling companies in Europe, North America, and Asia. An in-depth description of representative pyrometallurgy-based and hydrometallurgy-based processes is reported, providing classification of unit operations, their industrial readiness, and quality of output materials. The analysis shows that the pyrometallurgical route can treat different LIB chemistries without pre-sorting, but Li is not recovered unless the slag is refined. Hydrometallurgy-based processes are more chemistry-specific and in, although being affected by losses of electrode active materials during the mechanical pre-treatments for black mass separation. Efforts are required to promote in Europe the industrial capacity and readiness of hydrometallurgical processes by facilitating sorting and mechanical separations. There is also the need for harmonization of criteria for outputs definitions and rules for calculating recycling efficiency indicators. This represents an opportunity for modeling to support quantitative techno-economic and environmental assessments of the entire LIB recycling chain.

### 1. Introduction

Nowadays climate change and the abuse of natural resources have become challenging global issues, which cannot be overlooked. Several alternatives have been conceived to move towards a sustainable and climate-neutral society, guided by strategic plans such as the European Green Deal [1] and the Horizon Europe plan [2], which place the European Union (EU) at the forefront of the green transition. One of the main objectives is the decarbonization of the transport sector by the

adoption of electric vehicles (EVs), whose spread is rapidly growing in the last decades. Switching to EV utilization is expected to contribute to a significant reduction in the emission of greenhouse gases (GHGs) from the road transport sector, which accounted for the 11.9% of global emissions in 2020 [3]. The spread of EV adoption is also promoted by strategic legislative initiatives, such as the EU ban to the selling of new internal combustion engine cars by 2035.

There exist different categories of electric vehicles, such as hybrid electric vehicles (HEVs), plug-in hybrid electric vehicles (PHEVs), and battery electric vehicles (BEVs) [4]. HEVs and PHEVs integrate both an

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List of acronyms	
Acronym Description	
BEV	Battery Electric Vehicle
CAM	Cathode active material
CHB	Cyclohexylbenzene
CRM	Critical Raw Material
DEC	Diethyl carbonate
DMC	Dimethyl carbonate
EC	Ethylene carbonate
EMC	Ethyl methyl carbonate
EOL	End-of-life
EV	Electric vehicles
GHG	Greenhouse Gas
HEV	Hybrid Electric Vehicle
LCA	Life cycle assessment
LCO	Lithium cobalt oxide LiCoO <sub>2</sub>
LFP	Lithium iron phosphate LiFePO <sub>4</sub>
LIB	Lithium-ion battery
LMO	Lithium manganese oxide LiMn <sub>2</sub> O <sub>4</sub>
NCA	Lithium nickel cobalt aluminum oxide LiNi <sub>x</sub> Co <sub>y</sub> Al <sub>1-x-y</sub> O <sub>2</sub>
NiMH	Nickel metal hydride
NMC	Lithium nickel manganese cobalt oxide LiNi <sub>x</sub> Mn <sub>y</sub> Co <sub>1-x-y</sub> O <sub>2</sub>
PHEV	Plug-in Hybrid Electric Vehicle
PVDF	Polyvinylidene fluoride
VOC	Volatile organic compound

internal combustion engine and an electric motor of different sizes, where the electric propulsion is limited for HEVs (ca. 2 km electric autonomy given by the energy recovered during braking) and more significant for PHEVs (50–100 km electric autonomy, with the possibility to charge the battery by connecting it to an external electric power source); BEVs are powered solely by a battery pack, allowing up to 500 km of driving range. In any case, the key technology for all these EV categories is the lithium-ion battery (LIB). Thus, along with the growth of EV adoption, there is an equivalent rise in the production of LIBs, which are increasingly used also for stationary energy storage applications. Therefore, considering that the expected lifetime of a LIB system is 10–15 years [5], a large flow of LIBs at end-of-life (EOL) is expected in the future: by 2030, 140 million EVs are predicted to be on the road all over the world, generating cumulatively 11 million tons of spent batteries [6,7]. If such spent batteries cannot be repurposed in second-life energy storage applications, thus delaying their disposal, they effectively become a hazardous waste, which contains flammable organic solvents, polymeric layers, graphite, metallic foils, and oxides of transition metals such as Ni, Co, Mn, and/or Fe in addition to Li ions [8,9]. While such a flow of spent LIBs can represent a serious waste-management challenge, it can be turned into a good recycling opportunity, thus making spent LIBs a valuable source of useful materials [10].

There are multiple driving forces for LIB recycling, including environmental, economic, and strategical matters. In fact, if the disposal of hazardous materials, like flammable solvents of the electrolyte and carcinogenic metals as Ni and Co, is prevented, also materials with a high market price can be recovered, making EOL LIBs particularly attractive from an economic perspective [6,10]. Nevertheless, the core reason pushing LIB recycling, especially in the EU, is linked to the raw materials employed in LIB technology, among which Li, Co, and natural graphite, which are included in the 2020 list of Critical Raw Materials (CRMs) [11]. This means that these materials combine high importance for the EU economy, concerning technological development and elevated supply risk [12–14]. However, recovering such valuable and critical materials entails technological challenges because LIBs are not standardized, neither in cathode chemistries nor in geometrical shapes and sizes, resulting in an extremely complex inventory of materials which are highly mixed together, thus making LIB recycling an open question regarding the ideal process configurations [10,15,16].

Several and quite diverse LIB recycling routes have been proposed and applied, where the common key feature is the recovery of metals contained in the cathode active material (CAM) [17–19]. Pyrometallurgy-based processes are currently the most established battery recycling route, consisting of smelting the whole battery at high temperatures to recover an alloy of valuable metals, like Ni, Co, Cu, which can be further refined in high-purity metal salts through hydrometallurgical steps [20–22]; however, Li ends up in the slag, which has little economic value, so that Li does not re-enter the battery chain. The

hydrometallurgy-based recycling route is a comparatively emerging recycling strategy (although already industrially adopted in some cases) and consists of leaching, crystallizations, selective precipitations, and solvent extractions to recover high-quality single-phase metal salts, especially Co, Ni and Li salts. The co-precipitation recycling route shares similar unit operations, allowing the recovery of CAMs in the form of their precursors directly ready for LIB manufacturing. Finally, the direct recycling route aims at regenerating cathode (and anode) active materials by avoiding their destruction via smelting or leaching, thus representing a so-called closed-loop solution; however, direct recycling is not technologically ready yet at industrial level [7,9,10,16,23–27]. It has to be mentioned that, among these recycling routes, “greener” versions of some operations are recently being studied at research level [28–30].

While the currently established LIB recycling processes are mainly aimed at recovering metals as alloys or individual battery-grade salts, future needs are driving recyclers to move towards closed-loop solutions, so that valuable materials contained in the spent batteries can re-enter the same value chain [31,32]. Moreover, along with the difference among the various recycling routes, each process category varies significantly both in terms of unit operations employed and their sequence in the process to achieve similar goals [15]. As an example, electrolyte removal in the hydrometallurgy-based route can be carried out either before or after the crushing step by using different unit operations, such as thermal drying [33], extraction with solvents [34] or with sub/super-critical CO<sub>2</sub> [35]. Broadly speaking, such different alternatives entail different material recovery efficiencies and quality of recycled products, thus determining the overall sustainability of the recycling process [5,16]. These aspects are becoming increasingly relevant considering that new battery recycling regulations, such as the one proposed by the European Commission [36], need to be supported by detailed technical overviews on recycling capabilities to set meaningful recycling indicators.

This review paper aims at providing a systematic and updated comparison and classification of recycling processes, thus complementing previous surveys about legislative considerations [18,37,38], process chains operations [10,37,39,40], environmental impacts [8,9,41], challenges and opportunities [5,15,16] and future perspectives [42–45]. An extensive survey of existing and established LIB recycling processes and companies in Europe, North America, and Asia is provided, aimed at characterizing, classifying, and comparing the unit operations to identify similarities, critical aspects, and industrial readiness. This wide outlook on the current state-of-the-art allows for a critical analysis of recycling processes in terms of quality of the recycled outputs. These topics are tied to the need of reliable data and information required to develop harmonized rules for evaluating recycling indexes and possible implications at industrial and policy levels. In such regard, the scope of the review paper is limited to recycling processes which are established or, for emerging ones, at an advanced level of development (e.g., pilot scale or demonstration state); a similar approach is

considered for the chemistries of LIBs analyzed, which are limited to the current systems available on the market.

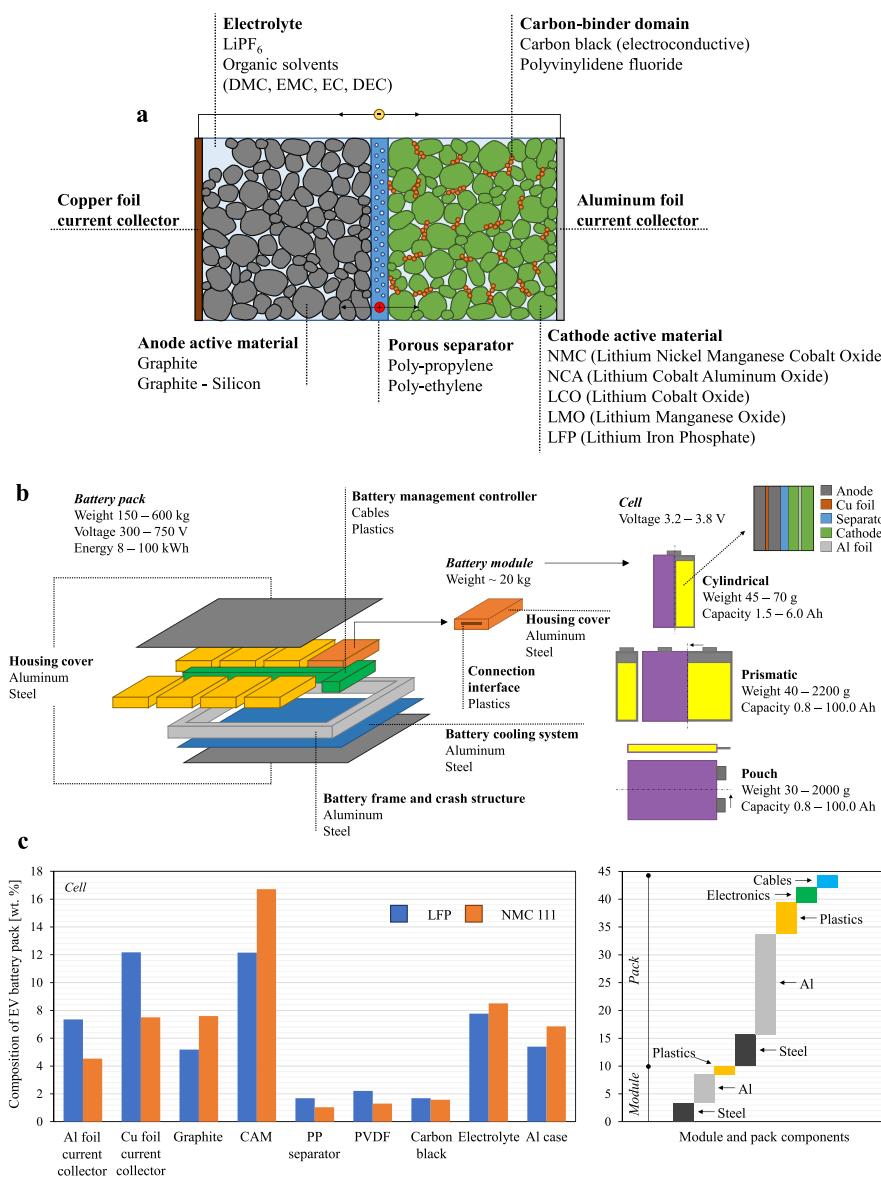
The paper is organized as follows. First, an overview of LIB designs and chemistries is given in Sec. 2 to illustrate the involved materials. Then, LIB recycling routes and processes are discussed in detail in Sec. 3.1, with a further focus on a selected representative pyrometallurgy-based process (Sec. 3.2) and a representative hydrometallurgy-based process (Sec. 3.3). Other industrial processes are described in the supplementary information (SI), whereas a classification and assessment of unit operations is carried out in Sec. 3.4. This is followed by an accurate comparison of recycling processes and recycling companies based on performance and quality of outputs (Sec. 4), ending up in a discussion of implications and possible future scenarios (Sec. 5).

## 2. LIB design and chemistry

A LIB is a complex system, made of different materials organized in a range of functional units, as shown in Fig. 1 [43,46]. The basic unit of a LIB system is the cell, which is schematically depicted in Fig. 1a in its most general design, comprising (from left to right) [47]: a Cu foil current collector, a porous anode made of graphite (and optionally

silicon) particles tightened by a binder [48], a porous separator made of polypropylene or polyethylene [49], a porous cathode made of active material mixed with conductive carbon and a polymeric binder (e.g., polyvinylidene fluoride, PVDF) [50–52], an Al foil current collector. The porous electrodes and the separator are soaked in an electrolyte consisting of LiPF<sub>6</sub> salt (ca. 1 mol L<sup>-1</sup>) dissolved in a mixture of organic solvents (e.g., dimethyl carbonate, DMC, ethyl methyl carbonate, EMC, ethylene carbonate, EC, cyclohexylbenzene, CHB, diethyl carbonate, DEC) [53]. Such a sequence of layers, which has a typical thickness of hundreds of microns or less, is wrapped multiple times within a cell, which is contained in a hard casing made of steel or Al alloy, or in a flexible pouch housing of Al laminated film [54,55].

The material composition of the anode is rather uniform for different LIB typologies, being graphite the most widespread anode active material for Li intercalation [48]. New generations of LIBs may include 5–10 wt% of silicon particles to increase the gravimetric capacity (3578 mAh g<sup>-1</sup> for silicon vs. 372 mAh g<sup>-1</sup> for graphite), although the large volumetric expansion (up to 300%) of silicon during intercalation currently hinders its larger share in the anode composition [54]. In any case, active material particles are bound together by a binder, such as PVDF, enabling a good adhesion with the current collector foil [54].



**Fig. 1.** Global overview of a battery system: a) schematic of the main components of a LIB cell, focusing on chemistries and materials currently available on the market; b) exploded view of a representative battery pack for an EV, showing its organization into modules and cells in three different designs, highlighting the characteristic materials and parameters; (c) on the left, percentage weight distribution of the materials composing a battery cell for two relevant chemistries (LFP in blue, NMC111 in orange), summing up to 55.6% of the total battery weight, where the remainder of the weight is distributed in the module and pack components (which are the same for both cathode chemistries) as reported on the right (elaborated from Refs. [46,54]). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

The cathode composition, instead, is more diverse because, along with an electroconductive additive like carbon black and a binder (PVDF), different chemistries of the cathode active material are used by different manufacturers for various applications [47,54]. Typical cathode active materials are [43,46,54]: lithium cobalt oxide LiCoO<sub>2</sub> (LCO), lithium manganese oxide LiMn<sub>2</sub>O<sub>4</sub> (LMO), lithium nickel manganese cobalt oxide LiNi<sub>x</sub>Mn<sub>y</sub>Co<sub>1-x-y</sub>O<sub>2</sub> (NMC), lithium nickel cobalt aluminum oxide LiNi<sub>x</sub>Co<sub>y</sub>Al<sub>1-x-y</sub>O<sub>2</sub> (NCA) and lithium iron phosphate LiFePO<sub>4</sub> (LFP), whose properties are summarized in Table S1 [42,54,56–58]. LCO is used in LIBs for smartphones, laptops, and digital cameras, although its relatively short life span, low thermal stability, and high cost are significantly slowing down its adoption. LMO batteries are largely used for power tools, medical instruments and, more recently, for powering e-bikes and scooters. The NMC cathodes are especially adopted for EV and come in different stoichiometries, ranging from low Ni excess formulations (such as LiNi<sub>0.33</sub>Co<sub>0.33</sub>Mn<sub>0.33</sub>O<sub>2</sub>, named NMC111 or NMC333, and LiNi<sub>0.5</sub>Co<sub>0.3</sub>Mn<sub>0.2</sub>O<sub>2</sub>, NMC532) to high (LiNi<sub>0.6</sub>Co<sub>0.2</sub>Mn<sub>0.2</sub>O<sub>2</sub>, NMC622) and very high excess (LiNi<sub>0.8</sub>Co<sub>0.1</sub>Mn<sub>0.1</sub>O<sub>2</sub>, NMC811), in order to achieve different properties [50,52]; broadly speaking, increasing the Ni content improves the energy density and the accessible capacity, at the expenses of lower cycle stability and higher production costs due to stricter production conditions (for example, NMC811 synthesis is very sensitive to moisture). NCA cathodes come in different stoichiometries too (e.g., LiNi<sub>0.8</sub>Co<sub>0.15</sub>Al<sub>0.05</sub>O<sub>2</sub> is more widespread compared to LiNi<sub>0.85</sub>Co<sub>0.11</sub>Al<sub>0.04</sub>O<sub>2</sub> or LiNi<sub>0.84</sub>Co<sub>0.12</sub>Al<sub>0.04</sub>O<sub>2</sub>), offering comparatively higher energy density and accessible capacity than NMC, with the drawback of lower thermal stability and larger material and production costs [52,59]. Finally, LFP is attractive for its thermal stability, cycle life, and lower cost due to the abundance of raw materials, but the lower cell potential and gravimetric energy make it more suitable for stationary energy storage applications, although its use in mid-class EVs is rapidly increasing [60].

There are three different LIB cell designs as shown in Fig. 1b, namely cylindrical, prismatic, and pouch, but their size is not currently standardized. In a cylindrical cell the single repeating unit is wound several times to create a jelly roll, that is placed in a cylindrical steel case. Cylindrical cells, in their different sizes (e.g., 18650, 21700 or the Tesla 4680), are smaller than prismatic and pouch designs, thus entailing lower packing density and gravimetric energy density, while offering good mechanical stability and competitive manufacturing costs [54,61]. In prismatic cells the flat jelly roll is wound in a rectangular area and compressed in a hard casing, thus improving space utilization and permitting flexible design with customizable size, from small format ones for laptops and tablets to larger formats for EV applications [54]. The pouch cell design, whose size and application are similar to the prismatic design, is characterized by a non-rigid case made of heat-sealable laminated multilayer foil, which can swell upon cycling, resulting in better packaging efficiency and weight minimization despite higher manufacturing costs [43,58]. Pouch cells are not provided with safety and protection devices, which need to be arranged externally to the cell.

For electric vehicles and other industrial applications, unit LIB cells are usually connected in series and packaged to create modules which, in turn, make up the battery system (Fig. 1b) [62]. Each module, which is encased in a steel or Al housing, includes a thermal management system [63] and an electronic module management unit, which controls cells temperature, voltage and current [23,62,64]. Modules are connected in series and in parallel to form the battery system. The battery system contains not only the battery pack but also additional peripheries such as the battery management unit, the housing cover, the cooling system, and connection cables [54]. The weight and electric energy of a battery system vary significantly, e.g., ca. 30 kg and 0.6–1.5 kWh [4,54] for HEVs, ca. 170 kg and 9–20 kWh for PHEVs, up to ca. 300 kg and 30–100 kWh for BEVs [54] (see a summary in Table S2 [42]).

The average weight composition of a battery system for EVs is shown in Fig. 1c [46,54]. The left panel reports the material composition of a

single cell, either for LFP (in blue) or NMC111 (in orange) cathode chemistry, while additional data are reported in Fig. S1. The cumulative sum of materials contained within LIB cells is equal to 55.6% at the system level, while the remainder weight is due to the peripheral components (such as, housings, electronics, cables, plastics) that make up the modules and the battery pack, according to the percentage weight distribution reported on the right panel of Fig. 1c. In case of small format batteries used in portable electronics (e.g., for laptops, smartphones, power tools), there are no peripheries since LIB cells are directly used and a steel case is typically adopted, resulting in the weight composition reported in Fig. S2.

### 3. Description of recycling processes

#### 3.1. Introduction to LIB recycling processes

Once framed the importance of LIBs recycling and their morphological differences, this section gives a general description of the main recycling routes, followed by two representative examples in Sec. 3.2 and Sec. 3.3, after which a detailed inventory of each unit operation involved is given in Sec. 3.4.

Collecting and recycling LIBs is technically and economically challenging due to both the complexity of the battery systems, which comprise several components and materials, and the absence of standardization of battery cell geometries and chemistries [7,16,24,25,65]. The main targets that a LIB recycling process should achieve are [16]:

- high-quality products: ideally recyclers should aim at recovering battery CRMs with battery-grade quality, so to achieve a “closed-loop” recycling, and thus avoiding “downcycling”, that is, recovering low-quality materials addressed to construction industry, metallurgy, or pigment production;
- competitive collection and recycling costs: the market price of recycled products should cover the costs of collection, transport, storage, and processing of spent LIBs, as well as a reasonable return on investment for recyclers, so that the price of recycled products is competitive with the cost of raw materials;
- low environmental footprint: LIB recycling is driven also by reducing the negative environmental impacts of landfilling and of mining and refining of virgin raw materials. However, also LIB recycling generates water contaminants and requires energy, although being less intensive than primary production [65]; thus, environmental impact analysis should be carried out to evaluate the sustainability of each LIB recycling process.

Currently there are three main LIB recycling routes to convert spent LIBs into recycled products, which are conventionally classified based on how elements are liberated from electrode active materials, that is, via smelting in pyrometallurgy, via leaching in hydrometallurgy, or without destruction of the crystalline structure of the electrode active material in direct recycling; the co-precipitation route can be classified as an additional recycling route lying between hydrometallurgy and direct recycling because it employs the leaching step while targeting the re-synthesis of cathode active materials. These recycling routes are implemented in industrial recycling processes, which are a chain of subsequent unit operations. Each unit operation can be categorized based on its function in the process, for example: i) pre-treatments such as deactivation and mechanical separations, which are generally operated before the smelting or leaching step, ii) proper pyrometallurgical operations, as the smelting, and iii) hydrometallurgical operations, as the leaching and the following steps required to recover ions from liquid solutions (e.g., precipitations, solvent extractions). A recycling process typically consists of a series of several unit operations of different categories. Thus, in this study the terms pyrometallurgy-based and hydrometallurgy-based, when referred to a recycling process in its entirety, shall not be regarded as a rigid classification applied to all the unit

operations in it, but rather as short cut notations of recycling routes according to the core separation step of metallic materials, namely the smelting for a pyrometallurgy-based process and the leaching for a hydrometallurgy-based process. Fig. 2 provides a general overview of such different recycling routes.

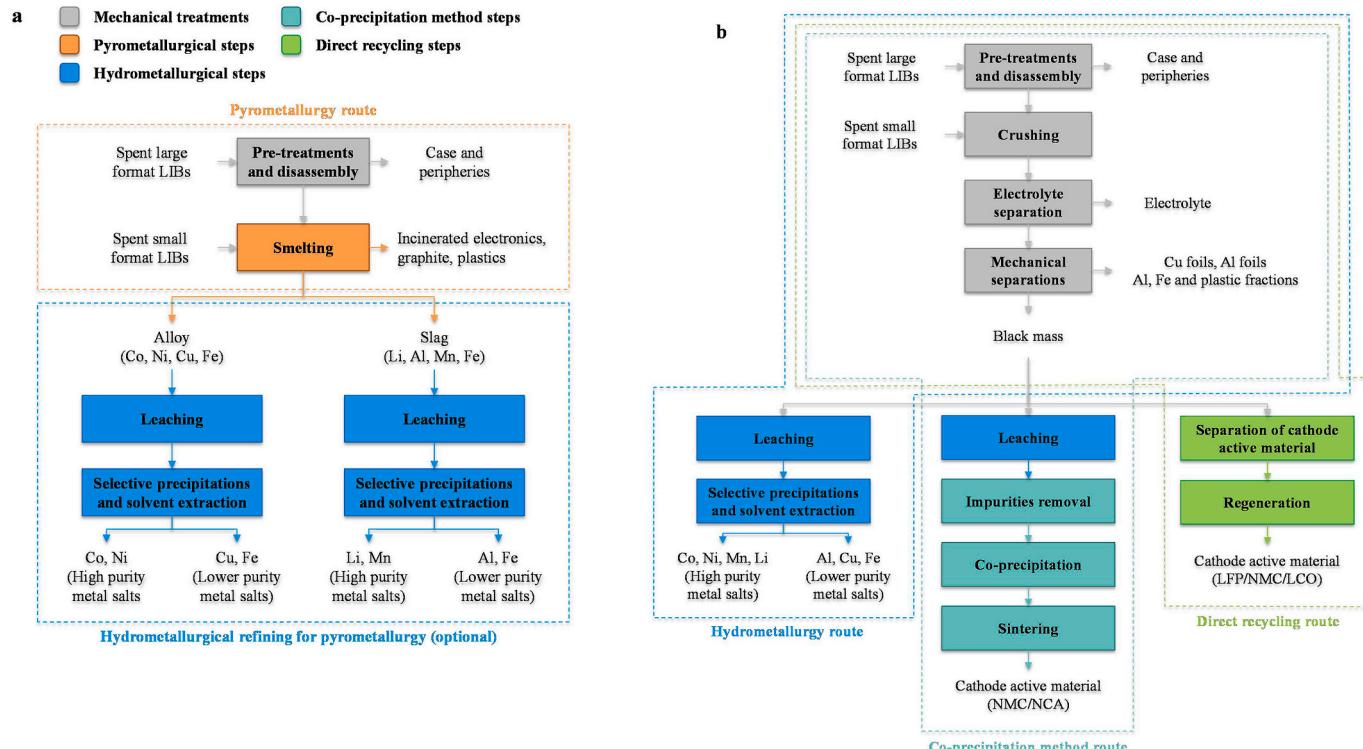
The first pre-treatment, common to all the recycling routes, consists of the disassembly of casing, electronics, and cables from large battery systems. This can be followed by a deactivation step to lower the electrical and flammable risks [24]. This generally includes an over-discharging step, carried out by discharge in a conductive liquid [66–68] (e.g., NaCl brine) or through an electric device with an adjustable load able to recover ca. 20% of the remaining energy to be introduced to the electric grid [54,69,70], followed by electrolyte deactivation, for example by thermal volatilization [71,72], freezing at  $-65^{\circ}\text{C}$  [73] or, as currently explored at laboratory and demonstration levels, electrolyte extraction with super- or sub-critical  $\text{CO}_2$  [74–77]. Notably, electrolyte removal processes can be applied both to the entire battery cells as pre-treatments or to battery fragments after disassembly and crushing [78] (see Fig. 2b); alternatively, over-discharging and electrolyte removal can be skipped in the pyrometallurgical route via directly smelting the battery cells [79] (Fig. 2a). Additional important pre-treatments are represented by mechanical separation steps (Fig. 2b), consisting of crushing, sieving, sorting, and classification [80], which separate active materials contained in the black mass from other outputs as current collector foil scraps, Fe scraps, and plastic fraction [80]. The crushing is executed under inert atmosphere or in a wet mode using an alkaline solution to eliminate the acid gas release and minimize flammable hazard [81].

After the pre-treatment phases, the core of recycling routes starts. Pyrometallurgy (Fig. 2a) is based on smelting the whole battery in a furnace at extremely high temperature (ca.  $1500^{\circ}\text{C}$ ) to recover an alloy of valuable metals (Ni, Co, Cu), which can be further refined via

hydrometallurgical steps to obtain high-purity metal salts [54,82,83]. The electrolyte is evaporated in the low-temperature zone of the furnace and addressed to energy recovery while plastics and graphite are burnt in the higher temperature zone. Li, Al, Mn, and part of Fe are contained in the slag fraction which is typically addressed to low-quality markets as construction material additive, although in principle it might be refined via hydrometallurgical operations [22].

The core of hydrometallurgy is leaching (Fig. 2b) in strong inorganic acids such as  $\text{H}_2\text{SO}_4$ , with  $\text{H}_2\text{O}_2$  as an additional reducing agent [84,85]. Other more recent and environmentally friendly options, namely bio-hydrometallurgy, exploit the microbiological metal dissolution [86] and/or weak organic acids (e.g., citric, malic) [87] to recover precious metals, but their application to spent LIB recycling (mostly portable LIBs) is still far to be industrially ready [28,29]. The leaching aims at dissolving CAMs into solution, to recover them as single-phase metal salts through crystallization, selective precipitation, solvent extraction, and electrochemical methods (Fig. 2b, dashed blue contour) [88,89]. The process aims at recovering battery-grade salts, so that they can be potentially reused in the battery production chain contributing to a closed loop of CRMs [90]. The hydrometallurgical route, differently from the pyrometallurgical one, necessarily requires pre-treatments such as dismantling, over-discharging, and electrolyte separation followed by complex mechanical separations (in grey in Fig. 2b), which allow recovering several scraps of Al, Fe, and Cu, and, in particular, to extract the black mass [91].

The black mass is the starting point of direct recycling route (dashed green contour in Fig. 2b). This consists of separating and then regenerating the cathode (and anode) active materials without destructing them into elements through leaching or high-temperature treatments [7]. The regeneration aims at restoring the Li inventory, lost during battery life, in the CAM by keeping the particle morphology and crystalline structure, so that such a recycled product can be directly reused



**Fig. 2.** Simplified block flow diagrams of general LIB recycling processes based on: (a) pyrometallurgical route, with optional hydrometallurgical refining of the alloy and/or the slag, (b) hydrometallurgical/co-precipitation/direct recycling routes, preceded by a mechanical separation section. The colourmap on the top left indicates the category of each step of the process, that is, mechanical treatments (light grey), pyrometallurgy (orange), hydrometallurgy (blue), co-precipitation method (light blue/green) or direct recycling (green). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

for manufacturing new LIBs [92,93]. While still seeking established methods to directly regenerate CAMs [30], recent studies have been performed on spent LFP [93–95] proceeding via chemical [96] or electrochemical [97] lithiation. Finally, in the co-precipitation route (Fig. 2b, light blue/green) the black mass is leached, impurities are removed, and cathode material precursors are co-precipitated to sinter the CAM. Thus, the crystalline cathode structure is broken as in the hydrometallurgical route, but the same CAM of the starting spent LIB is re-synthesized from the recovered metals [98,99].

As a general comparison between the different recycling routes, pyrometallurgy allows versatility in the process feed with batteries of different geometries and chemistries, accepting even other types of batteries like Ni-metal hydride (NiMH) or primary Li [19]. In addition, it is the most technologically developed one and ready at industrial scale, making it capable to process large flows of spent LIBs [9]. The major drawbacks of the pyrometallurgical route lie in the typical low purity of recycled products and the downcycling of CRMs like Li; then, the high energy consumption entailed in the smelter could result in a net increase in GHG emissions for all the LIB chemistries through their entire life cycle [65,100]. On the other hand, the hydrometallurgical route enables for high purity and selectivity of recovered metals, with low energy consumption, and low toxic gas emissions. Ciez et al. [65] estimate that a net reduction of GHG emission is expected from the hydrometallurgical recycling of NMC and NCA-types LIBs, despite wastewater pollution due to hazardous chemicals and strong acids cannot be ignored. However, the high complexity of the chain of mechanical separations prior to the proper hydrometallurgical steps and the specificity to limited LIB chemistries slow down the application of this recycling route at the industrial scale, resulting in plants of smaller capacity compared to pyrometallurgy-based processes [100,101]. Focusing specifically on the environmental impacts, recent life cycle assessment (LCA) studies have evidenced that, considering the whole life cycle of a battery pack (i.e., from production to operation and then recycling), the pyrometallurgical-based recycling contributes less than 11% to all of the assessed impact categories, except for the hydrometallurgical refining of the produced slag which highly affects water contamination,

contributing up to 60% of the life cycle impact [102,103]. Nevertheless, the recovered valuable materials are still relevant in terms of environmental credits to many categories of the LCA [104,105], while their economic value is not always balanced with the scarcity of resources that will be faced in the next future (e.g., for Li) [106,107], given also the dramatic increase of raw materials prices in the early 2022 [108]. Therefore, although LIB recycling generally entails lower environmental impacts compared to the mining of virgin materials, economic aspects are equally important. All recycling routes can be profitable at high volume of EOL LIBs considering current raw material prices and battery composition [109], even if the reverse supply chain must still be optimized [110]. In general, both pyrometallurgy-based and hydrometallurgy-based routes get revenues from recovering Co, Ni, and Cu [109], while only the second route may also consider selling Li given its recent high price [108]. A comparison among pyrometallurgy-based, hydrometallurgy-based and direct recycling routes revealed a cost saving relative to the use of virgin materials of 6, 13 and 27% respectively for NMC111, increasing to the range 38–43% when treating LCO only [111]. Considering the technological readiness of pyrometallurgy-based and hydrometallurgy-based processes and their adoption in existing industrial LIB recycling realities, the pros and cons of these two processes are schematically summarized in Table 1.

The direct recycling route has good potential in reducing GHG emissions, especially for high-value chemistries as NMC and NCA [65, 113,114]. However, technological difficulties, such as the need for an ideal separation of cathode from anode materials as well as the “one-chemistry” specificity of the processes, hinder the scale-up at industrial scale. Moreover, this may result in the production of obsolete CAMs which may not have a market considering the average life span of, say, an EV LIB (8–15 years) [115]. Thus, the co-precipitation method seems to offer a good trade-off between the most established hydrometallurgical steps and the ideal principles of direct recycling. Similarly to the hydrometallurgical route, the co-precipitation method requires preliminary mechanical separations (with corresponding loss in active materials) and may be susceptible to impurities in the leached solution. Nevertheless, this recycling route can potentially handle a wider mix of

**Table 1**

Summary of the key advantages and disadvantages of pyrometallurgical and hydrometallurgical routes for LIB recycling. Notably, pyrometallurgy and hydrometallurgy here refer to processes whose core unit operations are the smelting and the leaching, respectively.

Recycling route	Advantages	Disadvantages
Pyrometallurgy [20,21]	<ul style="list-style-type: none"> <li>• Versatile: applicable to several battery chemistries and geometries</li> <li>• High recycling capacity</li> <li>• No sorting or other pre-treatments</li> <li>• High recovery of valuable metals (Co, Ni, Cu)</li> <li>• Established method and industrial know-how</li> </ul>	<ul style="list-style-type: none"> <li>• Graphite, plastics, and electrolyte are necessarily burnt</li> <li>• Downcycling of Li and Al affecting the supply chain costs of the raw material</li> <li>• Expensive off-gas treatments to avoid toxic air emissions</li> <li>• High energy consumption</li> <li>• Low purity of products or further hydrometallurgical refinement</li> </ul>
Hydrometallurgy [84, 112]	<ul style="list-style-type: none"> <li>• High recovery rates, which can potentially include the entire electrolyte</li> <li>• High purity of products</li> <li>• Low energy consumption</li> <li>• Low GHG emissions</li> </ul>	<ul style="list-style-type: none"> <li>• High GHG emissions</li> <li>• High sorting efficiencies and mechanical pre-treatments required</li> <li>• Crushing of batteries involves safety problems</li> <li>• Efficient electrolyte separation methods have to be industrially demonstrated</li> <li>• Lower recycling capacity</li> <li>• Contaminated wastewater</li> <li>• High operating costs (e.g., solvents)</li> <li>• Process complexity</li> <li>• High costs for the treatment of the final effluent (neutralization)</li> </ul>

**Table 2**

Overview of LIB recycling companies located in Europe, North America, and Asia. For each company, the table reports the declared plant capacity as the flow of spent batteries, along with the battery chemistry treated in the facilities. The category of prevalent unit operations used in the industrial reality, namely mechanical (M), pyrometallurgical (P), hydrometallurgical (H), co-precipitation (CP), direct recycling (DR) or a combination thereof, is reported in the penultimate column, where round brackets indicate a future addition to the facility.

Company	Location	Capacity (year) [t y <sup>-1</sup> ]	Battery chemistry treated	Industrial reality	Source
<b>Europe</b>					
Umicore	Belgium	7000 (2020)	Li-ion	P H	[18,126]
Accurec Recycling	Germany	4000–6000 (2020)	Li-ion	M P H	[122]
Glencore (Xstrata)	Norway	7000 (2020)	Li-ion, NiMH	P	[18,121]
Duesenfeld	Germany	2000–3000 (2020)	Li-ion	M (H)	[127]
Akkuser Oy	Finland	1000 (2020) 3000 (2020)	Li-ion Others	M	[128,129]
Recupyl Valibat	France	110 (2020)	Li-ion	M H	[130]
SNAM	France	300 (2020)	Li-ion, NiMH, NiCd	P	[122,131]
Erasteel Recycling (Valdi)	France	20,000 (2020)	Mainly others, accepts Li-ion	P	[132]
Batrex Industries	Switzerland	200 (2020)	Li-ion	M	[18,23]
Euro Dieuze Industrie	France	200 (2020)	Li-ion	M	[122,133]
Redux	Germany Austria	10,000 (2020)	Li-ion	M	[134]
Neometals	Germany	10,000 (2022)	Li-ion	M H	[135]
Northvolt AB	Norway	8000 (2022) 125,000 (2030)	Li-ion	M H	[136]
Fortum Oyj	Sweden	Unknown (2023) up to 125,000 (2030)	Li-ion	M H	[137]
Stena	Sweden	3000 (2022) 10,000 (2023)	Li-ion	M H	[138]
<b>North America</b>					
Retriev (Toxco)	USA	4000 (2020)	Li-ion	M H	[24,121,139]
	Canada	4500 (2020)	Li-ion	M H	
Glencore (Xstrata)	Canada	7000 (2020)	Li-ion, NiMH	P	[18,121]
INMETCO	USA	6000 (2020)	NiMH, accepts Li-ion	P	[18,140]
Ascend Elements (Battery Resourcers)	USA	1500 (2020)	Li-ion	M CP	[141]
OnTo	USA	-	Li-ion	DR	[18,75]
Li-Cycle	Canada	5000 (2020)	Li-ion	M	[142]
	USA	5000 (2020)	Li-ion	M	
	USA	10,000 (2020)	Li-ion	M	
	USA	60,000 (2023)	Li-ion	CP (H)	
American Manganese Inc	Canada	150 (2020)	Li-ion	CP (M)	[143,144]
	Italy	40,000 <sup>a</sup> (2024)	Cathode scraps	CP (M)	
Lithion Recycling	Canada	200 (2020)	Li-ion	M H	[121,145]
	Canada	2000 (2023)	Li ion	M H	
<b>Asia</b>					
Sony Sumitomo	Japan	150 (2020)	Li-ion	P H	[18,82]
Hunan BRUNP	China	30,000 (2020)	Li-ion, NiMH, others	M CP	[122]
Shenzhen Green Eco Manufacturer Hi Tech (GEM)	China	20,000–30,000 (2020)	Li-ion, NiMH	M H	[122]
SungEel Hi Tech	South Korea	8000 (2020)	Li-ion	M H	[122,146]
Huayao Cobalt	China	65,000 (2020)	Li-ion	M H	[122,147,148]
Highpower International	China	10,000 (2020)	Li-ion - NiMH	M P H	[122]
Guanghua Sci-Tech	China	10,000 (2020)	Li-ion	M H	[122]
Tele Battery Recycling	China	3000 (2020)	Li-ions, others	M H	[122]
Kobar	South Korea	800 (2020) 1200 (2020)	Li-ion NiCd, NiMH	M P	[122]
Dowa Eco-System Co Ltd.	Japan	1000 (2020)	Li-ion, others	P H	[122]
Nippon Recycle Centre Corp.	Japan	5000 (2020)	Li-ion, NiCd, NiMH, alkaline	P	[122]
JX Nippon Mining and Metals Co. Ltd.	Japan	-	Li-ion, others	M H	[122]

<sup>a</sup> American Manganese Inc recycling plant will recycle ca. 7700 t y<sup>-1</sup> of cathode production scraps from ItalVolt gigafactory, corresponding to 40,000 t y<sup>-1</sup> of spent LIBs.

spent LIBs compared to direct recycling by adjusting the composition of the dissolved metal salts to modify the chemistry of the re-sintered CAM, partially solving the obsolescence problems [98,116,117].

Worldwide these different recycling routes are adopted (and sometimes integrated each other) by different companies, which process LIBs as their main feed (e.g., Umicore, Retriev Technologies, Recupyl Valibat, Akkuser, Sumitomo-Sony, Accurec) or as a secondary input, resulting in different recycling efficiencies, recovered materials purity, and declared plant capacities [18,38,40,73,118–123]. Table 2 summarizes the main LIB recycling companies, grouped according to their geographical location and reporting their prevalent categories of unit operations adopted in their industrial reality, according to the declared information from scientific literature and company websites or reports. Considering the large number of emerging technologies reported in the literature [115], only those at pilot plant stages, under permitting procedure, under commercialization, or that have produced several patents are listed in Table 2 to provide a snapshot of the current technologies ready

at industrial level. Notably, some hydrometallurgical companies are organized in dislocated hubs, which perform LIB collection and mechanical separation of the black mass, which is then treated in a single hydrometallurgical facility, thus minimizing transportation cost and flammable risks [124,125]. Other companies stop the recycling process at the black mass, which is then addressed to other recycling companies. A discussion of Table 2 is also reported in Sec. 4 in terms of claimed process capacities and quality of products.

Finally, to explain in detail LIB recycling routes, the main steps and the comparison of unit operations at different levels of development, two representative industrial realities are selected: one is the established Umicore process, as an example of a pyrometallurgical route followed by the hydrometallurgical refining of the alloy [126]; whereas the second reality is an emerging process which combines the mechanical section of Duesenfeld and the hydrometallurgical/co-precipitation method section of Ascend Elements [141]. The reasons behind such choices are mainly based on availability of data and technological readiness. In particular,

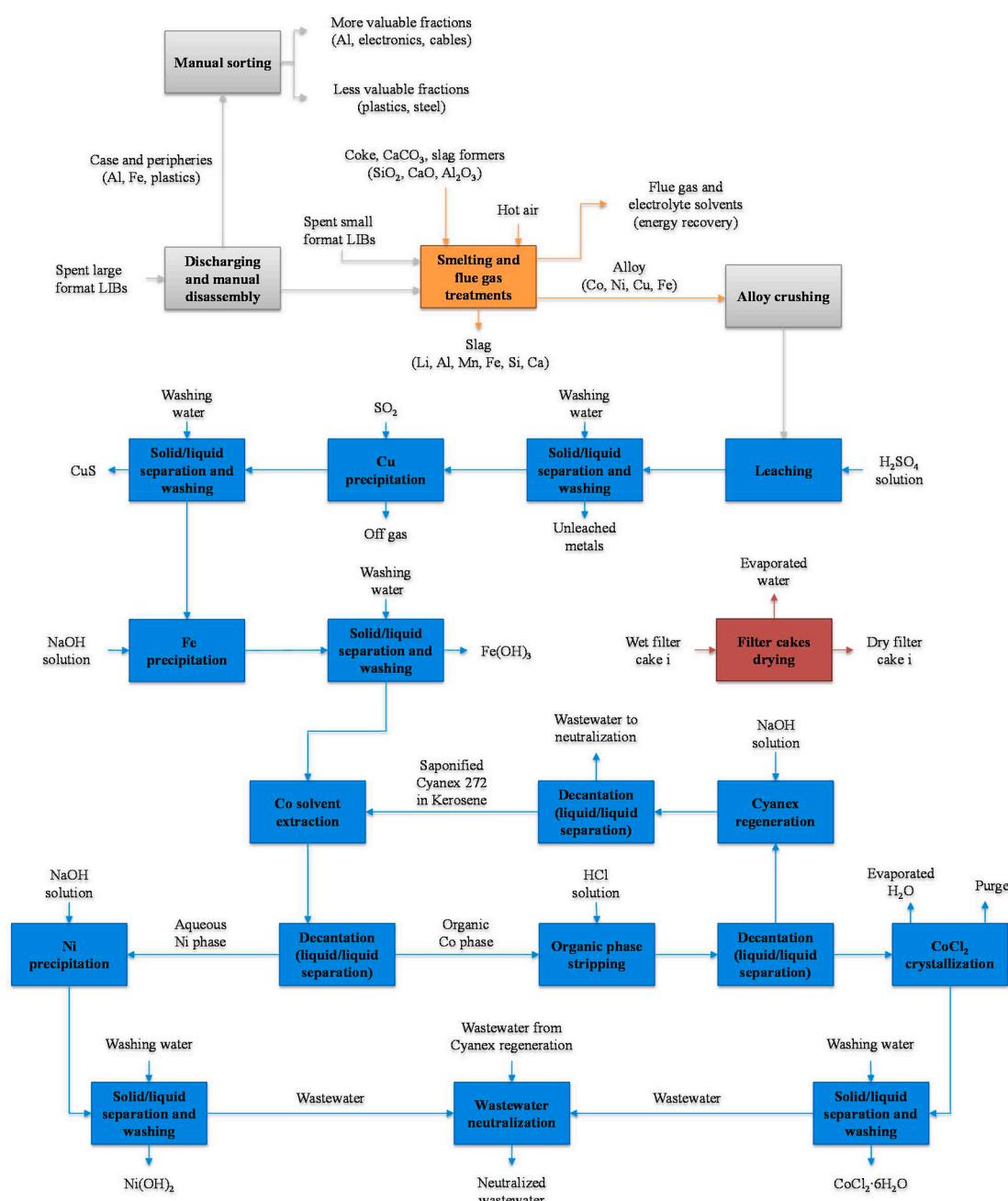
the second process is explained in detail in the LithoRec project [141] and stands out as one of the best available technologies in terms of material recovery. Only the mechanical separations envisaged in the LithoRec project are currently developed at industrial scale by Duesenfeld, while the hydrometallurgical/co-precipitation steps are not operative yet; nevertheless, the latter are similar to the hydrometallurgical/co-precipitation section carried out by Ascend Elements, which is thus considered here.

### 3.2. Representative established pyrometallurgy-based process: Umicore

Umicore is a multinational material processing group dealing with energy materials, performance materials, catalysts, and recycling [18, 23, 24, 40, 80, 82, 83, 120–122, 149–151]. Umicore developed a recycling

process called Valéas in 2000–2003, specific for rechargeable NiMH and Li-ion batteries. After testing the process in a pilot plant in Höfers, Sweden, in 2011 the 7000 t y<sup>-1</sup> plant in Hoboken, Belgium, was established [121]. The Umicore process is a standard example of pyrometallurgical route in battery recycling, with a subsequent hydrometallurgical section added to refine Ni and Co [149]. The feed can be composed of NiMH, Li-ion, and Li polymer batteries. EV LIBs are disassembled manually in the facility at Hanau, Germany, and the resulting battery cells and modules are shipped to the recycling facility in Hoboken, Belgium, where the pyrometallurgical process is carried out. The obtained alloy is sent to Olen, Belgium, to recover high-quality metals via hydrometallurgical refinement. The overall flowchart of the Umicore process is depicted in Fig. 3.

The core of the Umicore process is the smelting (orange in Fig. 3).



**Fig. 3.** Simplified flowchart of Umicore process [18, 23, 24], including pyrometallurgical smelting (orange), mechanical treatments (grey) and hydrometallurgical refining of the alloy (blue); the heat treatments, as the drying of filtered materials, are reported in red and apply to all the humid output streams. For the main streams the most relevant components are specified. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Disassembled battery cells are mixed with limestone ( $\text{CaCO}_3$ ), sand, coke, and reducing agents (i.e., metallic Al and Zn). The feed must contain 30–50 wt% of spent batteries to produce an alloy with a sufficient content of Co and Ni to have economic revenues. The feed is introduced into a vertical shaft furnace with a top submerged lance [152]. The material enters the so-called “preheating zone” where the temperature of the mixture does not exceed 300 °C. A slow increase in temperature allows the electrolyte to be slowly evaporated, and thus reduces the risk of explosions to a minimum [40,54]. In the second zone, the “plastics pyrolysis zone”, the temperature increases up to 700 °C, whereby the plastic components and binders of the cells melt, decompose, and partly evaporate in an overall exothermic process which heats the hot gases rising from the bottom of the furnace. The hot gases rise into the “preheating zone”, cool down and are fed together with the electrolyte vapors to the flue gas treatment section. In the third zone, “the smelting and reduction zone”, a regulated flow of pre-heated air (500 °C), optionally enriched in oxygen, is injected at the bottom of the shaft furnace. In this zone, where the temperature is between 1200 and 1450 °C, the remaining battery waste is transformed into two fractions: a slag fraction, consisting of Li, Al, Si, Mn, Ca, and residual Fe in oxidized state, along with an alloy, which mainly contains Cu, Co, Ni, and Fe in reduced state [18,24].

Different smelter feeds depend on the battery casing: since steel does not act as a reducing agent during the smelting while Al does, the latter decreases the amount of carbon used for the reduction of cathode metals. The temperature inside the smelter is determined by the highly exothermic combustion reaction of carbon and endothermic thermal decomposition of  $\text{CaCO}_3$ . The temperature of the flue gas leaving the furnace is increased to 1150 °C by a plasma torch before entering the post-combustion chamber [149]. Toxic halogens, produced by the decomposition of electrolyte salt  $\text{LiPF}_6$  and the binder PVDF [153], and volatile organic compounds (VOCs) [154] are captured by injection of Ca, Na, or  $\text{ZnO}$  in the post-combustion chamber. The hot gases are then cooled to 300 °C by a quench operation with water vapor and then conventional filtration is carried out. The Ultra High Temperature technology of the gas cleaning system based on the plasma torch allows to fully decompose the organic compounds and avoids the production of harmful dioxins or VOCs [155]. The toxic halogen compounds are captured in the flue dust which is filtered out and sent to disposal. For 1 t of spent batteries, 5000 MJ of heat is required for the smelter and the gas clean-up [82]. The following is an example of the smelter feed described in Umicore patent US 7169206B2 [149]: the furnace is fed with 1200 kg of LIBs with stainless steel casing, 200 kg of slag (40 wt%  $\text{CaO}$ , 34 wt%  $\text{SiO}_2$ , 11 wt%  $\text{Al}_2\text{O}_3$ , 15 wt% others), 110 kg of  $\text{SiO}_2$ , 100 kg of  $\text{CaCO}_3$ , 400 kg of coke and a flow of  $120 \text{ m}^3 \text{ h}^{-1}$  of pre-heated air at 500 °C. The quantity of  $\text{SiO}_2$ ,  $\text{CaCO}_3$  and slag is increased in case of LIBs with Al casing. The smelting process, which has a mean residence time of 455 min, results in the following metal recovery rates defined as the mass ratio between the metal recovered in the alloy and the total metal in the feed: 92.8% for Cu, 99.0% for Ni, 64.5% for Fe, and 94.0% for Co. The other fractions of these metals, in particular of Fe, together with other slag formers and other metals like Li, Al, and Mn, end up in the slag phase in the form of metal oxides. The slag from LIBs could undergo hydrometallurgical Li recovery [156,157], but currently this option is not economically attractive, so the slag is sold as an additive for construction material.

For the hydrometallurgical refinement (in blue in Fig. 3), the alloy is crushed and leached with  $\text{H}_2\text{SO}_4$  solution, thus forming metal sulfate salts dissolved in aqueous solution [80]. Next, Cu is removed from the solution by precipitation of  $\text{CuS}$  and  $\text{Cu}_2\text{S}$  using  $\text{SO}_2$  at elevated temperatures [54], then Fe is precipitated as hydroxide using  $\text{NaOH}$  solution. The resulting solution, which contains Co and Ni ions, is pumped into a mixer-settler, where an organic extractant such as Cyanex 272 (organophosphinic acid) is contacted with the aqueous solution to separate the Co, which goes to the organic phase, from the Ni, which stays in the aqueous phase [150,151]. Generally, the extraction with Cyanex 272 is

carried out at room temperature with a volumetric ratio between organic solvent and aqueous solution in the range 1–5 and pH around 5, achieving extraction efficiencies of Co and Ni above 80 wt% and under 10 wt%, respectively [151,158,159]. Cyanex 272 is typically diluted in kerosene, and partially saponified with a  $\text{NaOH}$  solution to enhance its reactivity towards Co extraction. The selectivity series of Cyanex 272 for metal ions is the following:  $\text{Fe}^{3+} > \text{Zn}^{2+} > \text{Cu}^{2+} > \text{Mn}^{2+} > \text{Co}^{2+} > \text{Mg}^{2+} > \text{Ca}^{2+} > \text{Ni}^{2+} > \text{Li}^+$  [160]. Thus, Fe, Cu, and Mn impurities, which are not removed from leachate, end up in the organic Co-rich phase, whereas the aqueous solution, containing mostly Ni sulfate, includes the remaining Co sulfate [54,161,162]. A solution of  $\text{NaOH}$  is added to the Ni-rich phase to precipitate  $\text{Ni(OH)}_2$  at pH ca. 9, leading also to the precipitation of  $\text{Co(OH)}_2$  as an impurity. The Co-rich organic phase, instead, is stripped with a HCl solution to remove Co from the organic extractant as an aqueous  $\text{CoCl}_2$  solution. Such a solution is sent to a crystallization section to obtain solid  $\text{CoCl}_2$ , setting the operative condition to avoid the crystallization of  $\text{NiCl}_2$  and  $\text{CuCl}_2$ , which are more soluble than  $\text{CoCl}_2$  [18,39,163]. The organic extractant is regenerated with  $\text{NaOH}$  solution, thus creating a closed loop for the extraction unit operation.

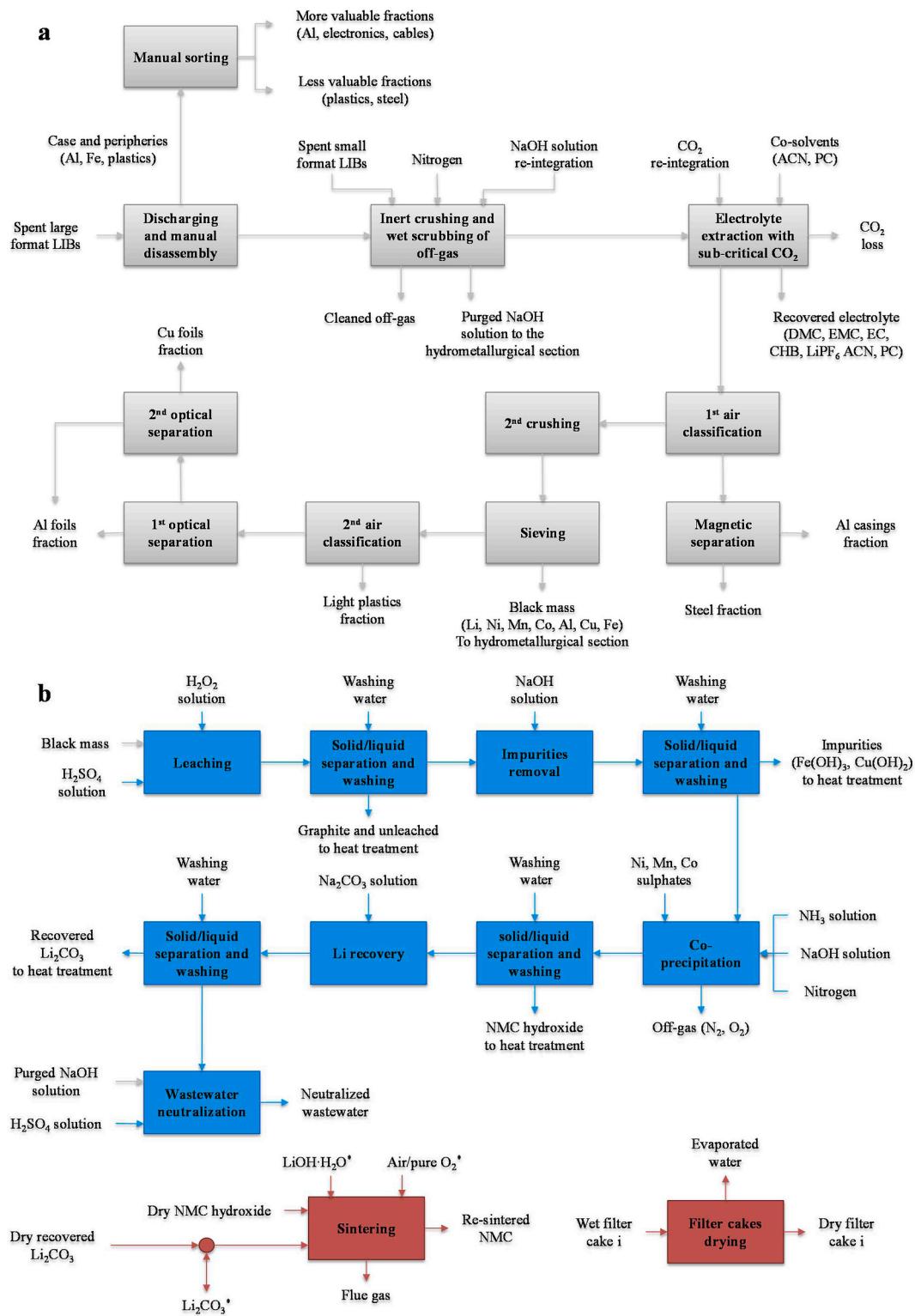
In summary, the Umicore pyrometallurgical route is based on the smelting, which produces an alloy then refined via hydrometallurgical steps to obtain Co, Ni, Cu and Fe salts. The key features of the process are the high material recovery rates of such metals, while Li, Al and Mn end up in the slag. Plastics, graphite, and electrolyte are burnt for energy recovery and the resulting gases require a dedicated cleaning system [17,19,79].

### 3.3. Representative emerging hydrometallurgy-based process: LithoRec

The LithoRec process was conceived in two projects funded by the German Federal Ministry of the Environment, Nature Conservation Building, and Nuclear Safety and VDI/VDE Innovation + Technik GmbH in 2009 and 2012, coordinated by the Technische Universität Braunschweig and the Westfälische Wilhelms-Universität Münster [18, 24,35,54,91,122,164–170]. The projects aimed at developing a new recycling process by combining electrical, mechanical, thermal, and hydrometallurgical treatments, specifically designed for NMC-based LIBs from EVs. To prove the industrial feasibility of the process, a temporary demonstration plant was built in Braunschweig and, during four months of operation, it recycled 1.4 t of battery systems reaching an overall material recycling efficiency of 75–80% [54], which could be potentially increased if also graphite and electrolyte salts were recycled as shown in the project at lab scale [141]. The mechanical treatments of the LithoRec process are implemented by Duesenfeld company in Wendeburg, Germany, which treats about  $2000 \text{ t y}^{-1}$  of LIBs. The resulting black mass is sent to another company for the recovery of Co, Ni, and Mn as single-phase hydrated sulfates in addition to  $\text{Li}_2\text{CO}_3$  and graphite. Instead, the LithoRec project [141] suggests a hydrometallurgical route based on the co-precipitation method to recover a ternary Ni–Co–Mn hydroxide and  $\text{Li}_2\text{CO}_3$  to be used as precursors for the re-sintering of NMC powders: such an original approach is described here and represented in Fig. 4.

The first step is discharging the battery system to 0 V, which is carried out in ca. 90 min and allows for the recovery of approximately 20% of the battery electric energy, followed by a 24 h short-circuit time to minimize the relaxation voltage of the battery [171,172]. The battery packs are manually disassembled to recover system peripheries such as housing, cables, and the battery management unit; automation of this process does not seem practicable due to the different geometries of battery packs [6].

The battery modules are then crushed under nitrogen atmosphere [150]. The crushing is performed with a shredder consisting of a single rotary shear with forced feeding, equipped with a discharge screen at a mesh size of 20 mm, which limits the upper size of fragments [54,173]. The connection from the crusher to the following equipment is



**Fig. 4.** Simplified flowchart of LithoRec process [18,24,35,54,91,122,164–170], including mechanical treatments (grey), hydrometallurgical steps (blue) and heat treatments (red). (a) First section of the process, comprising all the mechanical treatments. (b) Second part of the process, related to the hydrometallurgical treatments of the black mass. The asterisks denote streams which may be optionally required in case of adjustments in NMC stoichiometry. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

performed via a gastight screw conveyor, which is closed before starting the operation. The exhaust gas from the crusher is processed in a sequence of condenser, carbon filters, and wet scrubber with a recirculating alkaline solution, to recover respectively evaporated electrolyte solvents, the remaining VOCs, and the toxic HF originated by binder and

Li salt decomposition [174].

Next, the remaining electrolyte is removed to minimize the flammable hazard and improve the following separation processes thanks to a lower adhesion among the fragments. Several electrolyte removal strategies were investigated in the LithoRec projects, such as solid-liquid

extraction via DMC, extraction with sub-critical CO<sub>2</sub>, and thermal drying [54]. The first approach is not practicable because several extraction stages with DMC and water are required to reduce LiF impurities below the threshold required for the hydrometallurgical section [34,175]. An alternative solid-liquid extraction method, as reported in Fig. 4a, uses sub-critical CO<sub>2</sub> to recover both the organic solvents and the conductive Li salt. As investigated by Grützke et al. [164] at laboratory scale, the electrolyte components are extracted with an efficiency of 89% through a flow of CO<sub>2</sub> at 60 bar and 25 °C with additional co-solvents (a mixture of acetonitrile and propylene carbonate to enhance the extraction yield of EC and LiPF<sub>6</sub>) for 30 min, followed by 20 min without additional co-solvents to maximize the extraction of non-polar linear carbonates (EMC and DMC). After lamination, the solvents condense and are separated from CO<sub>2</sub> that leaves the shredded fragments completely dry, with additional benefits of avoiding fire hazard associated to any traces of Li metal, which is converted to Li<sub>2</sub>CO<sub>3</sub>. The applicability of super-critical CO<sub>2</sub> at industrial level has still to be demonstrated [35, 176–178], although OnTo company has patented such operation and is going to test it in a pilot plant [75–77]. The third option for electrolyte removal, which is currently adopted in the Duesenfeld plant at industrial scale [54], consists of a thermal drying at temperatures between 80 and 140 °C at 100–300 mbar [179]. The organic solvents of the electrolyte are vaporized and then condensed, or alternatively they are sent to a post-combustion process to obtain thermal energy. In this case, the conducting salt LiPF<sub>6</sub> is decomposed, producing toxic HF gas [8].

After electrolyte removal, the battery fragments undergo a series of mechanical separations as a combination of air-sifting, crushing, and sieving processes to separate several output streams [54,91]. Dried battery fragments are fed to the first air zig-zag-sifter, which separates the light fractions from the heavy fractions. This air classification is carried out with a mass load of 109 g kg<sup>-1</sup> air and an air velocity of 3.34 m s<sup>-1</sup> [9]. The heavy fraction consists of steel (13.8 wt%), Al housing (47.7 wt%), Al modules (26.6 wt%), plastics (4.0 wt%), Cu (5.0 wt%), and inclusions (2.9 wt%), whereas the light fraction is composed by Al and Cu foils, cathodic and anodic active materials, and plastic separators. The heavy fraction is fed to the following magnetic separation to recover steel and Al case fractions. Instead, the light fraction undergoes homogenization of the fragments via a cutting mill with a discharge screen of 10 mm (2<sup>nd</sup> crushing), followed by a vibration sieve at a mesh size of 500 µm to separate coating active materials of the electrodes [180]. Such a final powder resulting from the sieving process is known as black mass and contains electrode coatings (97 wt%) with impurities of Cu (1.9 wt%), Al (0.8 wt%), and steel (0.3 wt%).

The recovery efficiency of the black mass in the mechanical sections ranges between 75 and 90 wt%. The loss of black mass in other streams is due to the organic binder, which is not removed during previous treatments and is therefore responsible for the adhesion of coating materials to the oversized current collector foils [24,181]. In the current process, the second crushing preceding the sieving aims indeed at decreasing the adhesion of active materials from current collector foils, but its efficiency is not ideal, resulting to be the bottleneck of the recovery of CAMs in the mechanical section. Alternative methods have been investigated to remove the binder and enhance the black mass yield; for example, Hanisch et al. [24,182] suggest an incineration step at 500 °C to decompose the PVDF binder (which produces HF, thus requiring gas cleaning), then the battery fragments are fed to an impact separator (which is a modified jet sieve) where an air jet pushes the fragments to the wall of the chamber, thus separating the black mass particles with a recovery efficiency of 99 wt%, with only 0.1 wt% of current collector impurities. Nevertheless, this method has not been implemented yet at industrial scale.

While the black mass fraction is sent to the hydrometallurgical section, described later in this section, the fraction remaining on the sieve undergoes a second air classification via zig-zag sifting [183]. The second zig-zag sifter operates with a mass load of 25 g kg<sup>-1</sup> air and an air velocity of 1.1 m s<sup>-1</sup> [54,91], separating the plastic separators light

fraction from the heavier fraction made of current collector foils [73]. Al and Cu are recovered by using an optical separation thanks to a CCD (Charge-Coupled-Device) camera. The Cu fraction needs two runs of optical separation to obtain high-purity Cu scraps with only 0.3 wt% Al [184].

The black mass containing the cathode active materials, graphite, and traces of current collector foils is fed to the hydrometallurgical section [54,168–170] (Fig. 4b). The process is specifically designed for NMC chemistries but could accept also other LIB chemistries as secondary input. However, LFP content should be lowered as much as possible because, at alkaline pH typical of the following co-precipitation step, almost all the phosphorous amount inside the LFP is turned into phosphate ions, leading to the unwanted precipitation of highly insoluble Cu, Co, Ni, and Mn phosphates. This phenomenon represents a double drawback since it causes Co and/or Ni losses as well as contamination of the NMC precursor that might affect its crystalline structure [44,185].

The black mass is first leached in a stirred tank at 70–80 °C for 2–3 h using H<sub>2</sub>SO<sub>4</sub> 4 mol L<sup>-1</sup> and 30–50 wt% H<sub>2</sub>O<sub>2</sub> as an additional reducing agent, then insoluble graphite is removed via filtration [168]. The leachate contains Co, Ni, Mn, Li Al, Cu, and Fe ions, thus an impurity removal step is employed by adding NaOH solution: at pH ca. 6.5, Fe, Al, and Cu ions precipitate as hydroxides, whereas Co, Ni, Mn, and Li ions remain in solution [186,187]. After filtering out the precipitated salts, a stoichiometry adjustment can be performed by adding MnSO<sub>4</sub>, NiSO<sub>4</sub>, and CoSO<sub>4</sub> to the solution to obtain the right molar ratio between Ni, Mn, and Co, according to the selected NMC formulation desired as recycling product. Then, a transition metal hydroxide precursor (Ni<sub>x</sub>Mn<sub>y</sub>Co<sub>1-x-y</sub>(OH)<sub>2</sub>) is co-precipitated by increasing the pH to ca. 11 by using NaOH in presence of NH<sub>4</sub>OH as a chelating agent, which avoids the precipitation of metals as single-phase hydroxides. The operation is realized in a stirred tank under N<sub>2</sub> atmosphere to avoid the oxidation of Mn<sup>2+</sup> to Mn<sup>4+</sup>. The co-precipitation step, which is the core of the homonymous recycling route, is realized in the same operative conditions and with the same reagents as it occurs in the manufacturing of CAM from virgin raw materials [15,116,188].

After filtering out the NMC hydroxide precursor, the Li-rich solution is mixed with Na<sub>2</sub>CO<sub>3</sub> solution at 40 °C to precipitate Li<sub>2</sub>CO<sub>3</sub>. The NMC hydroxide precursor and the Li<sub>2</sub>CO<sub>3</sub> are dried (see the red box at the bottom right of Fig. 4b) and mixed with additional virgin Li<sub>2</sub>CO<sub>3</sub> to reach the right stoichiometry, that is, a molar ratio of Li to metal precursor in the range 1.0–1.1. The blended powders are ball milled for 48 h and compressed into pellets [168]. Finally, the pellets are sintered at 900 °C for 12–15 h to produce the selected NMC product. The operative conditions in the sintering step, involving the choice of Li source (Li<sub>2</sub>CO<sub>3</sub> or LiOH), the oxidizing atmosphere (air or pure oxygen), and a single or double step lithiation, strongly depend on the desired NMC formulation (e.g., NMC111 or NMC811).

In summary, the LithoRec hydrometallurgical/co-precipitation route consists of a series of mechanical separation steps, which enable the recovery of current collector scraps, light plastic fractions and even the electrolyte, despite being the bottleneck for the recovery efficiency of black mass. The following hydrometallurgical steps of the black mass, which separate graphite from the leachate metal salt solution, are centered on the co-precipitation of Ni<sub>x</sub>Mn<sub>y</sub>Co<sub>1-x-y</sub>(OH)<sub>2</sub> followed by the precipitation of a Li salt precursor. These two products are mixed to sinter the NMC cathode material.

### 3.4. Survey of unit operations in LIB recycling

After analyzing two representative processes, a comprehensive survey of all the unit operations employed in the studied recycling companies, including those reported in the SI, is developed in this section. Table 3 provides an inventory of such unit operations, both included in established industrial realities or still under development, classified according to their use in a specific section of the process (deactivation

**Table 3**

Inventory of typical unit operations used in LIB recycling processes and their possible and most common combinations. The industrial readiness is defined in three levels as follows: Laboratory (only at research level), Prototype (prototyped in established process), Industrial (in established and competitive realities).

Section	Unit Operation	Industrial Readiness	References
Deactivation	Discharge in conductive brine	Industrial	[66–68,189,190]
	Discharge with electronic devices	Industrial	[54,69,70]
	Electrolyte freezing with liquid N <sub>2</sub>	Prototype	[73,191,192]
	Thermal electrolyte evaporation	Industrial	[33,71,72,179]
	Incineration of battery fragments to remove binder	Prototype	[122,183]
	Electrolyte extraction with sub/super critical CO <sub>2</sub>	Prototype	[35,74–76,164,176–179]
Mechanical treatments	Inert crushing	Industrial	[18,81,130,193,194]
	Wet crushing in alkaline brine	Industrial	[133,135,139,142,195–197]
	Magnetic separation	Industrial	[8,81,198]
	Eddy-current separation	Prototype	[78,199,200]
	Air classification (zig-zag sifter)	Industrial	[54,73,201]
	Sieving	Industrial	[78,180,202]
	Detachment of active materials from metal foils through binder dissolution	Prototype	[203,204]
	Second crushing	Industrial	[141]
	Dense media separation	Prototype	[18,205–207]
	Optical separation of current collectors	Industrial	[141]
Pyrometallurgy	Froth floatation to separate active materials	Prototype	[143,208–210]
	Smelting in electric arc furnace of whole battery	Industrial	[211]
	Smelting in electric arc furnace of black mass	Industrial	[81,183]
	Smelting in shaft furnace	Industrial	[149]
	Smelting in rotary hearth furnace	Industrial	[20,140]
Hydrometallurgy	Off-gas treatments	Industrial	[17–19,79,82,154]
	Leaching of the black mass with H <sub>2</sub> SO <sub>4</sub> with optional reducing agent	Industrial	[143,145,168,212]
	Al and Fe impurities removal through pH adjustment	Industrial	[186,187]
	Cu impurities removal through precipitation or solvent extraction	Industrial	[142,145,207,213,214]
	Li <sub>2</sub> CO <sub>3</sub> precipitation with Na <sub>2</sub> CO <sub>3</sub> or CO <sub>2</sub>	Prototype	[99,130,215]
	Co and Ni separation with solvent extraction	Industrial	[175,216]
	Ni, Mn and Co ternary hydroxide or carbonate co-precipitation	Prototype	[15,98,116,188]
Direct recycling	Electrochemical production of Co, Cu and MnO	Prototype	[195,217–219]
	Solid-state regeneration	Laboratory	[26,220,221]
	Hydrothermal regeneration	Laboratory	[113,222–224]
	Precursor co-precipitation, Li source addition and sintering	Prototype	[95,225–228]

and mechanical treatments) or to their common presence in a specific recycling route (pyrometallurgy, hydrometallurgy, and direct recycling).

As already mentioned, after collection, sorting, and, optionally, manual dismantling for large battery systems [229], the first treatments in LIB recycling are deactivation steps aimed at reducing the risk due to high voltage and flammability of spent LIBs [230]. One of the first unit operations designed to minimize fire and explosion risks is cryogenic cooling, in which the whole battery is cooled at  $-160^{\circ}\text{C}$  with liquid nitrogen before crushing [82,163,191]. This operation, developed by the former company Toxco [192], is no longer applied due to the high operative costs, although sometimes freezing at ca.  $-60^{\circ}\text{C}$  of spent LIBs is still carried out to reduce fire risks during transportation [191]. High voltage risk is usually handled by an over-discharging step to 0 V

through electronic devices to recovery the remaining electric energy of the LIB, as performed by Duesenfeld [127], Redux [134], and Ascend Elements [231,232]. Otherwise, spent LIBs are effectively and easily discharged by soaking them in a conductive brine, resulting in the loss of the stored energy [67,189,190]. Typically this is coupled to wet crushing [195–197]: Neometals uses an undisclosed brine [135], Retriev uses a recirculating LiOH brine [139], Li-Cycle adopts a solution of Ca(OH)<sub>2</sub> and NaCl [142]. Other wet crushing methods, aimed at electrolyte removal and minimization of fire risk, are carried out by Euro Dieuze Industrie (E.D.I.), which utilizes a water flow [133], and Lithion, which uses recirculating light organic solvents such as DMC followed by thermal evaporation and distillation steps to recover each electrolyte solvent with high purity [145].

The most widespread technique for electrolyte separation is thermal

drying due to its well-known industrial know-how [33,179]. However, the quality of the recovered mixture of electrolyte solvents is affected by impurities generated by electrolyte decomposition, so that the recovered mixture is used only for energy recovery. Several operative conditions are selected by companies, varying not only temperature, pressure, and industrial equipment, but also the position of the operation in the process chain [15]. Accurec, SNAM, Redux, Glencore, and Sumitomo Sony operate thermal treatment to the whole battery at high temperature to evaporate all the electrolyte solvents, to decompose and remove the conductive salt, and to pyrolyze plastics [20,183]. As an example, Accurec operates at 500 °C and 100 mbar in a rotary kiln for 2–3 h to avoid oxidation of valuable metals, thus removing plastics, the whole electrolyte, and decomposing the PVDF binder, which is responsible for the difficult separation of black mass from current collector foils in mechanical treatments [122,183]. Instead, Duesenfeld carries out a thermal treatment on wet battery fragments at 90–140 °C and 100–300 mbar to evaporate only the electrolyte and decompose the conductive salts, thus minimizing energy consumption [179]. Nevertheless, Duesenfeld is also investigating an incineration step at 500 °C aimed at decomposing the PVDF binder to maximize the recovery yield of the black mass in the following mechanical separations [40]. Lithion performs an evaporation step at 90 °C of the liquid organic solvents exiting the wet crushing, then drying the battery wet fragments at 270 °C; heavy organics not evaporated at 90 °C are then burnt at 500 °C [145]. An alternative option for electrolyte separation is the extraction with sub- or super-critical CO<sub>2</sub> [74]. This is an innovative technology, potentially allowing the recovery of the whole electrolyte with enough purity to be re-used in the battery production chain. In the LithoRec projects researchers investigated electrolyte extraction with sub-critical CO<sub>2</sub> (25 °C, 60 bar) and additional polar organic co-solvents operated on battery fragments [179], whereas OnTo patented a process based on super-critical CO<sub>2</sub> (31 °C, 74 bar) performed on the whole battery after perforation of the casing, thus resulting in LIB deactivation which is functional for safe and low-cost transportation [75,76]. However, electrolyte extraction with sub-/super-critical CO<sub>2</sub> is still in its infancy regarding its industrial scale-up, with operative and investment costs which appear to be higher than those required for the established thermal drying.

The starting mechanical treatment operation is crushing, which is carried out in an inert atmosphere, in brine, or using other systems to prevent fire risks. In addition to the already discussed wet crushing, inert crushing is another option [81,194]: Duesenfeld operates with N<sub>2</sub> [179], Batrec uses CO<sub>2</sub> [193], while Recupyl utilizes both CO<sub>2</sub> and Ar [130]. The CO<sub>2</sub> atmosphere avoids any trace of metal Li from strongly react with water by converting it into the less reactive Li<sub>2</sub>CO<sub>3</sub>. Finally, Akkuser carries out the crushing step by exploiting cyclonic air removals, hence preventing the build-up of flammable gases inside the crushing chamber and addressing them to off-gas treatments [18].

Mechanical separations of metal and plastic scraps are realized through specific unit operations arranged in different ways. Crushing and sieving are the only processes always present in the mechanical section. Generally, after crushing the heavy fraction (steel and Al casings) and the light fraction (current collectors, separator, and active materials) are divided via air classification [54,91], as performed by Duesenfeld, Accurec, Redux, and Neometals. The next step, operated by several processes (Duesenfeld, Accurec, Redux, Akkuser, Euro Dieuze Industrie, Ascend Elements, Recupyl, OnTo, Li-Cycle, and Lithion), is the magnetic separation of the heavy fraction or of the whole battery fragments aimed at recovering steel [8,81,198]. Then, a second air classification can be performed to segregate the current collector foils from the separator, as done by Duesenfeld, Accurec, and Redux, or to differentiate Cu and Al foils, as by Lithion [201]. Alternatively, current collectors are divided from plastics through an eddy current separation [199,200,233], adopted by Euro Dieuze Industrie, Li-Cycle, and Lithion; optical separation of Cu and Al scraps is performed by Duesenfeld. An emerging and flexible unit operation is dense media separation, which

splits fragments according to their density by submerging them in a selected liquid with proper density [205,206]. OnTo uses this technique to separate graphite from CAMs by using a liquid with density in the range 2200–3500 kg m<sup>-3</sup> [18], Li-Cycle proposes to separate current collectors from plastics through a liquid with density 2500 kg m<sup>-3</sup> and Cu from Al with another liquid with density 2850 kg m<sup>-3</sup> [207], whereas Ascend Elements uses this technique to remove plastics from Cu scraps [232]. Incidentally, for emerging direct recycling and co-precipitation routes, the separation of graphite from the CAMs is, respectively, necessary and useful: excluding separation due to black mass leaching, the current options are the dense media separation, or the froth floatation, proposed by American Manganese Inc and Li-Cycle [143,208–210], which, however, appears technologically unready. In any case, as already mentioned, the yield of black mass from sieving operations is inversely proportional to the adhesion of electrode active materials to current collector foils [197]. While some companies, such as Duesenfeld and Lithion, employ an additional crushing step of the battery fragments, the detachment of active materials from current collectors can be facilitated by the previously mentioned thermal treatments, specifically designed to decompose the binder, or by binder dissolution in proper organic solvents like N-methyl-2-pyrrolidone, as carried out by Ascend Elements [203,204]. Detailed and recent surveys on pre-treatment technologies for LIB recycling can be found in dedicated studies [78,180,202].

Concerning purely pyrometallurgical steps, common battery smelters like Erasteel Recycling (former Valdi), SNAM, and INMETCO handle the feed including several battery types, also together with other metal scraps; other processes perform the smelting of the whole battery specifically for Li-ion technology, such as Umicore, Glencore, and Sony-Sumitomo [81]. Accurec is different from other pyrometallurgical processes because it performs the smelting only on the black mass recovered after mechanical treatments [18]. In the end, several smelting technologies are used including rotary hearth furnace (INMETCO) [140], electric arc furnace (Erasteel Recycling and Accurec) [211], and shaft furnace (Umicore) [149].

Hydrometallurgical operations are extremely diverse and with operative conditions often undisclosed by the companies. Leaching is the starting point of each hydrometallurgical route and it is currently operated by most of the companies by using sulfuric acid, either without an additional reducing agent, like in the Umicore hydro-refining of the alloy, or by introducing hydrogen peroxide [212], like Ascend Elements, Lithion (optionally substituted by MnO<sub>2</sub> [145]), Li-Cycle, and Accurec, or with SO<sub>2</sub>, as proposed by American Manganese Inc [143].

Fe, Al, and Cu ions in the leachate are treated mainly as by-products [234]. More valuable Cu ions can be selectively separated with Na<sub>2</sub>S solution like Lithion [145], via cementation with Fe shots (e.g., Accurec) [214] or through solvent extraction using for example LIX984 N as Li-Cycle suggests [142,207]. Otherwise, Cu ions are removed together with Fe and Al ions by adjusting the pH of the leachate, generally between 5 and 6.5 via addition of NaOH solution, to precipitate the corresponding hydroxides while keeping in solution Li, Ni, Mn, and Co ions [213]. After that, Ni, Mn, and Co ions are recovered: a practical solution seems to be the co-precipitation step, as carried out by Ascend Elements, American Manganese Inc and Li-Cycle [116]. However, it is undisclosed in these cases if NH<sub>4</sub>OH is used as a chelating agent to obtain a ternary Ni–Mn–Co hydroxide (as it is likely for Ascend Elements), which is the direct precursor of NMC, or if a mixture of single-phase metal hydroxides is recovered (as likely for American Manganese Inc and Li-Cycle) [235,236]. Otherwise, Ni, Co, and Mn are selectively separated by using solvent extraction, especially with Cyanex 272 or similar organic extractants to separate a Co and Mn-rich organic phase from a Ni-rich aqueous phase [175,216]. Co and Mn separation can be then carried out with another solvent extraction or via electrochemical routes [195,217–219]. Finally, Li recovery is realized by most companies by precipitating Li<sub>2</sub>CO<sub>3</sub> through the addition of Na<sub>2</sub>CO<sub>3</sub> solution [99,215] (e.g., Ascend Elements, Accurec, Retrev, Li-Cycle, American Manganese

Inc, Lithion), or through CO<sub>2</sub> bubbling (Recupyl) [130], or again by precipitating Li<sub>3</sub>PO<sub>4</sub> after neutralization with H<sub>3</sub>PO<sub>4</sub> (Recupyl) [130, 237, 238].

As previously explained, direct recycling is focusing on regenerating cathode materials without its destruction, but its development is still at research stage [113, 114, 225]. Among the different methods employed, hydrothermal regeneration has been successfully applied to fully recover LCO capacity [223] or to reinstate usage conditions on spent LFP [222] and LMO [224, 239]. Solid-state sintering is another direct method that proved to effectively restore the capacity of the degraded cathodes [221]. Meng et al. [220] developed an economic viable process that directly regenerates NMC batteries with minimized chemical consumption, while an acidless approach for regenerating LiNi<sub>0.5</sub>Co<sub>0.2</sub>Mn<sub>0.3</sub>O<sub>2</sub> from spent LIBs using LiOH·H<sub>2</sub>O was proposed in another study [26]. Co-precipitating cathode precursors from spent LIBs and re-sintering them by adjusting the temperature to obtain the optimal performance has also been exploited [226, 227]. Finally, direct recycling can be performed by adding external Li sources through different lithiation processes. Yang et al. [228] used a system composed by molten LiOH-KOH-Li<sub>2</sub>CO<sub>3</sub> to compensate the Li<sup>+</sup> loss in the degraded carbonate and repair the damaged structure via a “dissolution-recrystallization” mechanism, while the electrochemical reversibility of FePO<sub>4</sub> as impurity phase was used to regenerate degraded LFP with a graphite pre-lithiation strategy [95].

#### 4. Analysis and comparison of recycling routes in terms of outputs quality

The description of LIB recycling processes provided in the previous sections shows how recycling routes, recovered components and their purity in the process outputs strongly depend on the input and on unit operations. One of the principal aspects of pyrometallurgy-based processes is the capability to treat also small format LIBs for portable devices, since mixed LIB chemistries and battery types are easily processed, whereas one of the main features of the hydrometallurgical recycling route is the necessity of previous mechanical treatments to obtain the black mass, generally carried out in different facilities of the same company or even by different companies. Direct recycling is not mature yet while the co-precipitation method is currently operated at industrial or demonstration scale mainly by companies that treat mostly cathode scraps from battery manufacturing [73, 113].

To better underline how product quality and market potentialities vary among different processes, Table 4 reports a comprehensive list of the outputs and recovered materials of the analyzed battery recycling companies in Europe, North America and Asia as claimed by companies (see Table 2 for sources). The list of companies presented in Table 4 is sensibly shorter than the one in Table 2 (e.g., for Asia), this strictly depends on the availability of data. Concerning the process classification, orange represents pyrometallurgy-based processes (i.e., employing a smelting step), grey marks companies that carry out only mechanical treatments, and blue highlights hydrometallurgy-based processes (i.e., employing the leaching of black mass). Regarding the classification of product quality, materials which are not recycled are marked in red, while yellow refers to recovered materials which require further processing to be considered as recycled; the light-green color denotes materials which are fully recovered with sufficiently high quality, while a dark green color marks recovered materials with a battery-grade quality (or with a quality such that to be addressed to other high-quality markets). In this sense, the dark-green color is linked to the possibility to achieve a closed loop of materials from LIB recycling. Notably, battery-grade purity constraints are extremely strict, and many processes are not able to meet them, even when materials are recovered as high-purity single-phase metal salts [240]. It is also worth mentioning that the same material (for example, the slag) may be classified in Table 4 with different colors for different companies, depending mainly on the legislative framework of the country where the company operates, while

there is little difference in the chemical composition and quality of the material itself. Lastly, white cells mark the materials which are not present in the input of the considered process.

Table 4 classifies the output materials as a list of metallic elements, along with graphite, electrolyte, and plastics. Nevertheless, the outputs of a LIB recycling process are quite diverse depending on the recycling route and process chain adopted by the company [39, 241]. This results in a combination of the following ones: housing and peripheral components fractions, magnetic steel scrap, non-magnetic heavy fraction (Al casings and others), current collector foil scraps, plastics light fraction, black mass, electrolyte, slag, alloy, single-phase metal salts to be used as precursors, co-precipitated Ni, Mn and Co salts and even re-sintered cathode active materials, such as NMC or LCO. All these recycling outputs are considered in Table 4 as a destination of the materials.

Some specific considerations on the recycling quality classification adopted in Table 4 are necessary before comparing companies and territories. Scraps, fragments, or disassembled materials fractions, even with low impurities content, usually require further refinements, resulting in purity gain but additional material loss [78, 202]; as such, materials ending up in these fractions are marked in yellow in Table 4. The recycling status of the slag is an open question for LIB recycling, both for economical and policy reasons [15, 18], thus requiring harmonization of classification and definition criteria among different countries. Materials ending up in the slag are downcycled and can be considered recycled only from a legislative point of view depending on the country, e.g., the slag coming from the Umicore process, which has a market as construction material additive (as such, it is marked in light green for that specific company). Considering the elemental composition of the slag, where Li accounts for 2% of its mass, the hydrometallurgical refining can likely produce as outputs mainly Al, Mn, and Fe hydroxides, along with Li carbonate/hydroxide with traces of other metal salts. Broadly speaking, further hydrometallurgical refinement of the slag to recover Li is technologically possible but not economically viable [22, 156, 157], therefore the slag is generally marked as yellow in Table 4, except for battery smelters like INMETCO and Glencore which consider it as a waste of the recycling process. On the other hand, the alloy obtained in pyrometallurgical processes is always accountable as recycled output, albeit not considered as battery-grade quality; in fact, in most cases this type of alloy is not an end product, but it is addressed to Ni, Co or steel primary metallurgical processes.

Graphite is burnt in pyrometallurgical and thermal operations (therefore marked in red in Table 4), while in other processes it is typically recovered as unleached solid, trapped in the black mass and/or including other metal impurities, thus not considered as a high-quality output [210]. However, some companies apply further refinement steps to the unleached graphite, such as an additional leaching and a thermal treatment that removes plastic impurities and restore its morphology, as declared by OnTo and Lithion, whose graphite output can be considered as battery-grade according to company specifications [18, 145]. Nevertheless, graphite is also in the CRMs list [11, 12] in the EU, hence work should be done in Europe to recover such a valuable material, e.g., by investing on companies exploiting the hydrometallurgy-based route. Therefore, the quality and recycling status of a stream exiting from a recycling process depend both on technical considerations as well as on the legislative framework and the end use of such a material output.

Careful judgement is dedicated to assessing the quality of the recovered electrolyte. It has been demonstrated that the electrolyte can be recovered as a mixture of organic solvents, as single compounds or in its entirety including the conductive salts [33, 34]. In the last case, typically the electrolyte composition of a spent LIB is affected by impurities resulting from its aging or from the recycling operations, resulting in a low-quality fraction which cannot be directly addressed to battery-grade applications. For example, electrolyte recycling strategies such as thermal vaporization and sub/super-critical CO<sub>2</sub> extraction introduce impurities coming from the thermal decomposition of LiPF<sub>6</sub>

**Table 4**

Overview of the recycling products and recovered materials of the analyzed battery recycling companies in Europe, North America and Asia. Companies are classified based on the recycling process involved, i.e. pyrometallurgy-based (orange), mechanical treatment (grey), and hydrometallurgy-based (blue) processes. Recycling products and recovered materials are classified based on the recovery rate and quality, specifically recycled battery grade (dark green) and high-quality (light green) materials, recycled materials requiring further processing (yellow), recycled materials lost during the process (red), and materials not included in the input stream (white).

Co	Ni	Li	Mn	Al	Cu	C	Electrol.	Plastics	Fe
<i>Europe</i>									
<b>Umicore</b>	CoCl <sub>2</sub>	Ni(OH) <sub>2</sub>	Slag <sup>1</sup>	Slag <sup>1</sup>	Slag <sup>1</sup>	CuS	-	-	-
<b>Accurec</b>	Alloy	Alloy	Slag	Alloy	Al casings Al-Cu foils Al(OH) <sub>3</sub>	Al-Cu foils Cu	-	-	Steel fract. alloy
<b>Erasteel</b>	Alloy <sup>2</sup>	Alloy	Slag	Slag	Slag	Alloy <sup>2</sup>	-	-	Alloy
<b>SNAM</b>	Alloy	Alloy	Slag	Slag	Slag	Alloy	-	-	Alloy
<b>E.D.I.</b>	Black mass	Black mass	Black mass	Black mass	Al casings Al-Cu foils	Al-Cu foils	Black mass	-	Plastics fract. Steel fract.
<b>Akkuser</b>	Black mass <sup>3</sup>	Black mass <sup>3</sup>	Black mass <sup>3</sup>	Black mass <sup>3</sup>	Black mass <sup>3</sup>	Black mass <sup>3</sup>	Black mass <sup>3</sup>	-	Plastics fract. Steel fract.
<b>Duesenf.</b>	Black mass	Black mass	Black mass	Black mass	Al casings Al foils	Cu foils	Black mass	Organic solvents <sup>4</sup>	Plastics fract. Steel fract.
<b>Redux</b>	Black mass	Black mass	Black mass	Black mass	Al casings Al foils	Cu foils	Black mass	-	Steel fract.
<b>BatRec</b>	Black mass	Black mass	Black mass	Black mass	Al-Cu foils	Al-Cu foils	Black mass	Organic solvents <sup>4</sup>	Steel fract., Fe(OH) <sub>3</sub>
<b>Recupyl</b>	Co(OH) <sub>2</sub> Co	-	Li <sub>2</sub> CO <sub>3</sub> Li <sub>2</sub> PO <sub>4</sub>	-	Al-Cu foils	Al-Cu foils Cu	Graphite	-	Plastics fract. Steel fract. Fe(OH) <sub>3</sub>
<b>Neomet.</b>	CoSO <sub>4</sub>	NiSO <sub>4</sub>	Li <sub>2</sub> SO <sub>4</sub>	MnSO <sub>4</sub>	Al-Cu foils Al(OH) <sub>3</sub>	Al-Cu foils CuSO <sub>4</sub>	Graphite	-	Plastics fract. Steel fract. Fe(OH) <sub>3</sub>
<i>North America</i>									
<b>Inmetco</b>	Alloy	Alloy	Slag	Slag	Slag	Alloy <sup>2</sup>	-	-	-
<b>Glencore</b>	CoCl <sub>2</sub> Co	Alloy	Slag	Slag	Slag	Alloy <sup>2</sup>	-	-	-
<b>Retriev</b>	Black mass	Black mass	Li <sub>2</sub> CO <sub>3</sub>	Black mass	Al-Cu-plastic fract.	Al-Cu-plastic fract.	Black mass	-	Al-Cu-plastics fract. Plastics-steel fract.
<b>Asc. Elem.</b>	NMC and/or its precursor	NMC and/or its precursor	Li <sub>2</sub> CO <sub>3</sub> NMC	NMC and/or its precursor	NaAl(OH) <sub>4</sub>	Cu foils	Graphite	LiPF <sub>6</sub>	Plastic fract. Steel fract.
<b>OnTo</b>	NMC	NMC	NMC	NMC	Al-Cu-plastic fract.	Al-Cu-plastic fract.	Graphite	Electrolyte	Al-Cu-plastics fract. Steel fract.
<b>Li-Cycle</b>	Ni(OH) <sub>2</sub> Mn(OH) <sub>2</sub> Co(OH) <sub>2</sub>	Ni(OH) <sub>2</sub> Mn(OH) <sub>2</sub> Co(OH) <sub>2</sub>	Li <sub>2</sub> CO <sub>3</sub>	Ni(OH) <sub>2</sub> Mn(OH) <sub>2</sub> Co(OH) <sub>2</sub>	Al-Cu foils Al(OH) <sub>3</sub>	Al-Cu foils Cu	Graphite	-	Plastics fract. Steel fract. Fe(OH) <sub>3</sub>
<b>A.M.I.</b>	Ni(OH) <sub>2</sub> Mn(OH) <sub>2</sub> Co(OH) <sub>2</sub>	Ni(OH) <sub>2</sub> Mn(OH) <sub>2</sub> Co(OH) <sub>2</sub>	Li <sub>2</sub> CO <sub>3</sub>	Ni(OH) <sub>2</sub> Mn(OH) <sub>2</sub> Co(OH) <sub>2</sub>	-	-	Graphite	-	-
<b>Lithion</b>	Co	Ni(OH) <sub>2</sub>	Li <sub>2</sub> CO <sub>3</sub>	MnO <sub>2</sub>	Al foil Al(OH) <sub>3</sub>	Cu foil, CuS	Graphite	DMC EMC EC DEC	Plastics fract. Steel fract. Fe(OH) <sub>3</sub>
<i>Asia</i>									
<b>Sumitom.</b>	CoO	NiSO <sub>4</sub>	-	-	-	Cu	-	-	-
<b>Brupn</b>	NMC LCO	NMC	NMC LCO	NMC	NaAl(OH) <sub>4</sub>	-	-	-	-
<b>GEM</b>	Co salt	Ni salt	Li <sub>2</sub> CO <sub>3</sub>	MnO <sub>2</sub>	Al(OH) <sub>3</sub>	-	-	-	-

<sup>1</sup> slag addressed to the construction industry<sup>2</sup> the respective material is included in the alloy as an impurity<sup>3</sup> black mass recovered in this process also includes Al and Cu current collector foils and plastics in a not negligible amount<sup>4</sup> organic solvents of the electrolyte are recovered as a mixture of low quality, likely used for energy recovery.

and partial combustion of light organic solvents for the former, and contamination by extraction organic co-solvents for the latter [33,34]. In these cases, the electrolyte solvents are mostly addressed to energy recovery. Nevertheless, when thermal evaporation is followed by proper distillation, as proposed by Lithion, battery-grade electrolyte solvents may be recovered, despite the technical and economic feasibility of this approach has to be industrially demonstrated [145]. In this sense, Table 4 shows how Europe needs to catch up with North American companies that claim to recover electrolyte components even at battery-grade quality.

The analysis of Table 4 shows that valuable metals, such as Co, Ni, Cu but also Al and Fe, are typically recovered worldwide with sufficient quality, except for companies which perform only mechanical treatments, where these metals end up in the black mass and, as such, their recycling cannot be considered completed. In some cases, cathodic elements as Co and Ni are recovered as battery-grade single-metal salts or in mixtures ready for co-precipitation [80,216], if not as proper CAM as NMC in some American and Asian hydrometallurgical companies. Li and Mn are generally present in the slag output of pyrometallurgical companies, while hydrometallurgical processes typically enable for a battery-grade recovery of these metals. Cu is almost always recovered in the alloy or as foils, which are often mixed with Al; Al is generally recovered from Al casings, except for pyrometallurgical processes, where it ends in the slag [234]. Finally, Fe is basically always recovered in the steel fraction.

Regarding the territorial distribution, it is evident that in North America there is a larger share of recycling companies which provide high-quality and battery-grade recovered fractions compared to Europe. This is attributed to the different recycling routes used in the two continents, with hydrometallurgical routes which are predominant in North America. In fact, as evidenced in both Tables 2 and 4, in Europe several companies such as Akkuser, Duesenfeld, Redux, Euro Dieuze Industrie and BatRec, carry out only pre-treatments and mechanical processes to obtain the black mass, whereas companies that operate the entire LIB recycling process, i.e., Umicore and Accurec, follow the pyrometallurgical route. The only exceptions are the established company Recupyl and the emerging one Neometals, that achieve the whole recycling process using mechanical and hydrometallurgical treatments [130]. In North America several companies as Ascend Elements (former Battery Resources), OnTo Technologies, Li-Cycle and American Manganese Inc (A.M.I.), are focusing on the recycling of LIBs towards achieving a closed loop of CAMs; other two hydrometallurgical companies, i.e., the established Retriev and the emerging Lithion, can recover Li and cathodic elements with battery-grade quality as well. Most EU and US battery smelters like Erasteel Recycling, SNAM and INMETCO do not specifically focus on LIB recycling, but treat several types of batteries, therefore a considerable fraction of LIB materials is lost and the recovered ones are included in alloys. Glencore in Canada and Umicore in Belgium are the only smelters designed for LIB recycling, which operate a further hydrometallurgical refinement of the alloy to recover battery-grade Co and Ni. The Sony-Sumitomo process in Asia works analogously by recovering battery-grade Co, Ni, and high-purity Cu, but completely discarding all the other materials.

Finally, although Table 4 reports only a few Asian companies because of lack of specific information on the quality of their recycling outputs, Table 2 highlights that battery recycling in Asia follows mechanical treatments coupled to hydrometallurgy and even co-precipitation methods, with enormous recycling capacity when compared to similar facilities in Europe. In particular, Chinese LIB recycling companies are often associated to giga-factories which provide them with large amounts of scraps from battery manufacturing [242]. Therefore, as for the two Chinese companies in Table 4 (Brupn and GEM), high-quality recycled outputs are expected from LIB recyclers in Asia.

## 5. Outlook and open research questions

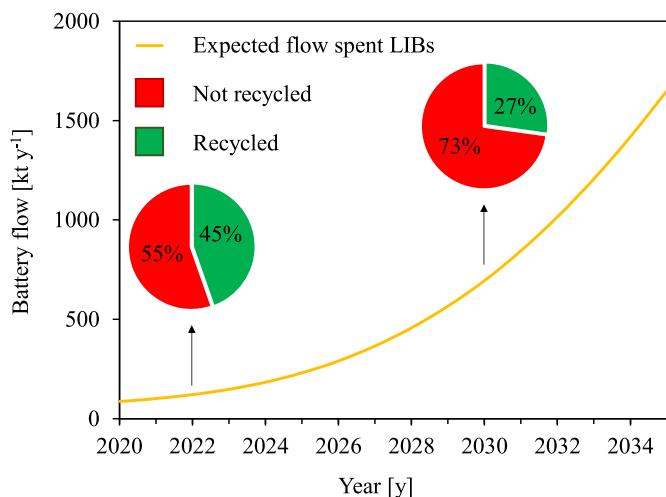
The analysis of recycling processes and quality of the recycled outputs leads to the identification of some criticalities and possible scenarios that are discussed in this section according to technological and management perspectives, evidencing implications for policy, industry, and research levels.

One of the main criticalities for the recovery of CRMs in hydrometallurgy-based and direct recycling processes does not lie in the proper hydrometallurgical or regeneration steps, but rather in the mechanical separations, in particular in the sieving of crashed fragments [78,202]. Sieving efficiency depends on crushing efficiency and thus on battery fragments size distribution; nevertheless, the limiting factor is the strong adhesion of active materials on current collector foils, which is attributable to polymeric organic binders [40,203,243]. Recycling companies are trying to overcome this issue by using additional crushing, thermal incineration, or dissolution of the binder in proper organic solvents, but neither of those options is yet optimal [81]. Hence, the solution does not seem to lie in the recycling steps, but more probably in the design-for-recycling [6,10]: battery makers should aim at replacing polymeric organic with water-soluble binders, which can be in principle easily removed in the recycling operations via a washing step with water; however, taking a life cycle perspective, the impact on LIB durability should be properly assessed since water-soluble binders might not guarantee the same long-lasting adhesion of active materials to metal foils as the current polymeric organic binders.

The problematic recovery of CRMs in the mechanical separation section explains why several hydrometallurgy-based recycling companies, especially those based on co-precipitation method, currently prefer to start from production scraps as input stream. This practice avoids mechanical separations and the consequent introduction of impurities since the composition of production scraps is less fluctuating compared to spent LIBs, thus making the hydrometallurgical steps inherently more efficient and leading to products of higher purity [73]. Thus, such a higher recycling efficiency must be attributed to the input feed rather than to the recycling process itself, showing again that the potential of hydrometallurgy-based processes is undermined by impurities and material losses in mechanical separations when starting from mixed spent batteries.

The variability of the LIB feed entering the recycling process is indeed another important factor. Currently, while pyrometallurgy allows higher versatility and generally can handle feeds of mixed LIB chemistries (provided that a certain amount of Co in the feed is guaranteed to ensure economic profit) [21], most hydrometallurgical, co-precipitation, and direct recycling processes are designed to handle spent LIBs with a few types of chemistries [84,112], thus requiring a sorting phase [205]. Obviously, sorting a ton of small-format LIBs (e.g., 18650-type), summing up to over 20,000 cells, is more complex than sorting a ton of battery systems, i.e., 3–5 battery packs of BEVs. In this regard, all the regulatory efforts in battery labelling, coding, and classification point in this direction [244] since reporting details, such as the chemical composition, in standardized form on shell information labels may realize, or at least facilitate, the automatic sorting [100]. For this reason, from a pure logistics perspective, one may speculate that pyrometallurgical recycling processes could treat small-format LIBs without a pre-sorting step, while hydrometallurgical processes could be dedicated to recycling large battery systems, sorted according to their chemistry. However, such a strict split may be constrained by both legal restrictions and market necessity, resulting in a trade-off that need to be specifically addressed, considering also the evolution of new hydrometallurgy-based processes that do not require the sorting phase [245].

Limiting the discussion to the EU as an example, there is the question of whether the flow of spent LIBs can be effectively recycled and how these batteries will be distributed among the different recycling companies. Based on published data about EV fleets [11], an internal



**Fig. 5.** Comparison of EU total outlooked flow of spent LIBs from 2020 to 2035 (solid yellow line). The insets report the fraction of EOL LIBs which are expected to be recycled (green) or not (red) in 2022 and 2030 considering a conservative situation in which recycling companies keep constant their capacity unless already specifically declared. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

elaboration by the Joint Research Centre estimates the annual flow of EOL LIBs: out of the 121 kt of spent batteries expected for 2022, only 54 kt can be recycled by EU companies based on the declared capacity listed in Table 2 (excluding companies treating mainly other types of batteries, like Erasteel Recycling, as well as the announced American Manganese Inc facility, which will recycle cathode production scraps from the ItalVolt giga-factory). As shown in Fig. 5, currently only 45% of the spent LIBs can be recycled by European companies, albeit with different efficiencies and quality of outputs as shown in Table 4. The current EU recycling capacity is rather equally distributed among pyrometallurgical, hydrometallurgical and mechanical-treatment companies, which contribute for roughly one third each to the total EU recycling capacity. This is arguably an overestimate because mechanical-treatment companies, whose end product is the black mass, do not complete the entire LIB recycling route. The situation is expected to further evolve in 2030, when only 189 kt of LIBs would in principle be recycled in the EU among the 695 kt incoming the EOL stage, resulting in a recycling fraction of only 27%. These figures for 2030 include the declared capacity of the NorthVolt AB new recycling plant, which will shift the share of hydrometallurgy recycling in Europe to 82%, where the remainder will be distributed between pyrometallurgy-based (10%) and mechanical-treatment (8%) companies. In practice, to overcome the forecasted EOL battery flow expected by 2030, each existing recycling company should significantly increase its recycling capacity compared to the actual capacity. Obviously, new recycling companies can be expected to enter the market by 2030 as well. Nevertheless, this analysis indicates that the EU might not be ready to handle a large flow of spent LIBs; moreover, hydrometallurgy does not seem developed enough at industrial scale to guarantee a closed loop on CRMs in the European LIB chain, so that the established pyrometallurgical processes will continue to be a major player in the medium term [11].

In any case, not only the total recycling capacity should be monitored, but also the share of different battery chemistries available for recycling, especially those which entail low economic value, such as LFP. In fact, treating LFP batteries is not economically attractive for pyrometallurgical processes, that are able to technically recover only Fe from this CAM, in addition to the Cu included in current collectors. The situation is even more problematic for hydrometallurgical and co-precipitation method processes because the leaching of LFP produces phosphate ions, which interfere with metal precipitations, causing

undesired loss in metals and impurities in the products [54,170]. As a matter of fact, only direct recycling methods seem to give savings with respect to the virgin materials when treating LFP [110]. Thus, although the new proposal for battery recycling regulation advanced by the European Commission states that all collected waste batteries must undergo proper treatment and recycling [36], proper strategies should be conceived to make LFP recycling effectively practicable, profitable, material efficient, and sustainable. Notably, Ciez and Whitacre [65] estimate that, compared to the mining of Fe precursor materials, the recycling of LFP batteries is more energetically intensive and results in a net increase of GHG emission for all the recycling routes. Therefore, the environmental impacts of LFP batteries must be properly assessed in their whole life cycle and compared to Ni–Co-based chemistries [246].

The final open question refers to the necessity of secondary Li supply. Taking again Europe as an example, while several mining companies are emerging to directly supply raw Li to the upcoming European battery manufacturers, also secondary Li sourcing from spent LIBs should be increased [247]. As shown in the previous sections, only hydrometallurgical recycling processes can recover Li with enough quality to re-enter the battery chain, because in pyrometallurgical processes Li goes in the slag. Actually, recovering Li from the slag is technically possible by using hydrometallurgical operations (even if the corresponding environmental impacts must be carefully evaluated) [248], but currently this seems economically unfeasible due to both the generally low price of Li and the complexity of hydrometallurgical refining. Nowadays the general leaching costs are mostly compatible with the overall total processing cost of 2–6 \$ kg<sup>-1</sup> of battery [249], although this strictly depends on the form, purity and yield of the products from the recovery stage. As a consequence, while the operating costs of hydrometallurgical operations now are mainly compensated by the revenues from Ni (ca. 21 \$ kg<sup>-1</sup>) and Co (ca. 50 \$ kg<sup>-1</sup>) [108] (mostly from NMC batteries), in the future they could be covered also by selling battery-grade Li. In fact, recent increases in Li price (almost by 8 times in the last months) [108], economic incentives, or legislative constraints can push recycling companies toward this solution. For the last aspect, there is certainly the necessity to set up harmonized definitions of what is recycled and what is not to remove differences in the legislative status of the slag depending on the location of the recycling industry (see Section 4). Such considerations go far beyond the slag problem and point in the direction of developing harmonized rules and definitions to assess what can be considered recycled, by establishing recycling indicators and calculation rules to quantify the efficiency of recycling processes to ensure sufficient quantity and quality of recycled outputs to re-enter the battery value chain. In this regard, the recent proposal for European regulation points in these directions [36] by introducing new indicators, such as the material recovery level (article 57 of the regulation proposal), i.e., the fraction of a target element that is present in the recycled outputs compared to its input mass in the spent LIB feed, and the recycled content (article 8 of the regulation proposal), i.e., the percentage of secondary materials coming from battery recycling compared to virgin materials in newly manufactured batteries, along with thresholds on the life cycle carbon footprint (including the EOL phase) for batteries to be put on the market (article 7 of the regulation proposal). Next to these indicators, the recycling efficiency, i.e., a measure of the mass of battery materials which end up in the recycled outputs compared to the battery input material stream, is still in force. These indicators and targets, whose ultimate definitions and ways to calculate them will be available at the publication of the European regulation, when taken together aim at giving a comprehensive perspective on LIB recycling, surely contributing to a more circular and sustainable battery value chain.

Ultimately, any effort should aim at reaching an optimal trade-off between different aspects related to LIB recycling: maximizing the yields of recovered materials (specifically CRMs) while lowering as much as possible the environmental impacts (e.g., carbon footprint) and still being viable from an economic point view both for manufacturers

(who aim at reducing the battery cost by avoiding precious metals) and recyclers (whose revenues come from valuable metals in the spent batteries). In this regard, this review paper represents a starting point, providing a qualitative assessment and classification of LIB recycling routes, unit operations, and outputs quality, opening the way for an in-depth quantitative analysis made with a combination of modeling tools, from process simulation to environmental assessment and economic models. Such models would lead to accurate evaluation of the techno-economic and environmental feasibility of different LIB recycling scenarios and this would greatly support policy makers, industrial investors and researchers.

## Disclaimer

Views expressed are those of the authors and do not reflect an official position of the European Commission. Any mention of commercial companies or products in this paper does not imply recommendation or endorsement by the University of Pisa, the JRC, and the European Commission.

## 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.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jpowsour.2022.231979>.

## References

- [1] ECONOMIC and SOCIAL COMMITTEE and the COMMITTEE of the REGIONS the European Green Deal, (n.d.).
- [2] European Commission, Horizon Europe: Strategic Plan 2021–2024, 2021.
- [3] H. Ritchie, M. Roser, CO<sub>2</sub> and Greenhouse Gas Emissions, OurWorldInData.Org., 2020. <https://ourworldindata.org/co2-and-other-greenhouse-gas-emissions>.
- [4] H. Fink, Li-ion Batteries for Automotive Applications – Quo Vadis?, 2016, pp. 69–81, [https://doi.org/10.1007/978-3-658-13255-2\\_7](https://doi.org/10.1007/978-3-658-13255-2_7).
- [5] A. Masias, J. Marcicki, W.A. Paxton, Opportunities and challenges of lithium ion batteries in automotive applications, ACS Energy Lett. 6 (2021) 621–630, <https://doi.org/10.1021/acsenergylett.0c02584>.
- [6] D.L. Thompson, J.M. Hartley, S.M. Lambert, M. Shiref, G.D.J. Harper, E. Kendrick, P. Anderson, K.S. Ryder, L. Gaines, A.P. Abbott, The importance of design in lithium ion battery recycling—a critical review, Green Chem. 22 (2020) 7585–7603, <https://doi.org/10.1039/D0GC02745F>.
- [7] Y. Bai, N. Muralidharan, Y.K. Sun, S. Passerini, M. Stanley Whittingham, I. Belharouak, Energy and environmental aspects in recycling lithium-ion batteries: concept of battery identity global passport, Mater. Today 41 (2020) 304–315.
- [8] P. Meshram, A. Mishra, Abhilash, R. Sahu, Environmental impact of spent lithium ion batteries and green recycling perspectives by organic acids – a review, Chemosphere 242 (2020).
- [9] L. An, Recycling of Spent Lithium-Ion Batteries: Processing Methods and Environmental Impacts, Recycling of Spent Lithium-Ion Batteries: Processing Methods and Environmental Impacts, 2019, pp. 1–217, <https://doi.org/10.1007/978-3-030-31834-5>.
- [10] G. Harper, R. Sommerville, E. Kendrick, L. Driscoll, P. Slater, R. Stolkin, A. Walton, P. Christensen, O. Heidrich, S. Lambert, A. Abbott, K. Ryder, L. Gaines, P. Anderson, Recycling lithium-ion batteries from electric vehicles, Nature 575 (2019) 75–86, <https://doi.org/10.1038/S41586-019-1682-5>.
- [11] S. Bobba, S. Carrara, J. Huisman, F. Mathieu, C. Pavel, I. European Commission. Directorate-General for Internal Market, Critical Raw Materials for Strategic Technologies and Sectors in the EU : a Foresight Study., n.d.
- [12] B. Vidal-Legaz, G.A. Blengini, F. Mathieu, C. Latunussa, L. Mancini, V. Nita, T. Hamor, F. Ardente, P. Nuss, C. Torres de Matos, D. Wittmer, L. Talens Peiró, E. Garbossa, C. Pavel, P. Alves Dias, D. Blagoeva, S. Bobba, J. Huisman, U. Eynard, F. di Persio, H. dos Santos Gervasio, C. Ciupagea, D. Pennington, Raw Materials Scoreboard, 2018, <https://doi.org/10.2873/08258>. Luxembourg.
- [13] J. Huisman, T. Ciuta, F. Mathieu, S. Bobba, K. Georgitzikis, D. Pennington, European Commission. Joint Research Centre., RMIS, Raw Materials in the Battery Value Chain : Final Content for the Raw Materials Information System : Strategic Value Chains : Batteries Section, 2020.
- [14] E.A. Olivetti, G. Ceder, G.G. Gaustad, X. Fu, Lithium-ion battery supply chain considerations: analysis of potential bottlenecks in critical metals, Joule 1 (2017) 229–243, <https://doi.org/10.1016/j.joule.2017.08.019>.
- [15] S. Windisch-Kern, E. Gerold, T. Nigl, A. Jandric, M. Altendorfer, B. Rutrecht, S. Scherhaufner, H. Raupenstrauch, R. Pomberger, H. Antrekowitsch, F. Part, Recycling chains for lithium-ion batteries: a critical examination of current challenges, opportunities and process dependencies, Waste Manag. 138 (2022) 125–139, <https://doi.org/10.1016/J.WASMAN.2021.11.038>.
- [16] A. Beaudet, F. Larouche, K. Amouzegar, P. Bouchard, K. Zaghib, Key challenges and opportunities for recycling electric vehicle battery materials, Sustainability (2020) 12, <https://doi.org/10.3390/su12145837>.
- [17] W. Lv, Z. Wang, H. Cao, Y. Sun, Y. Zhang, Z. Sun, A critical review and analysis on the recycling of spent lithium-ion batteries, ACS Sustain. Chem. Eng. 6 (2018) 1504–1521, <https://doi.org/10.1021/ACSSUSCHEMENG.7B03811>.
- [18] O. Velázquez-Martínez, J. Valio, A. Santasalo-Aarnio, M. Reuter, R. Serna-Guerrero, A critical review of lithium-ion battery recycling processes from a circular economy perspective, Batteries (2019) 5, <https://doi.org/10.3390/batteries5040068>.
- [19] C. Liu, J. Lin, H. Cao, Y. Zhang, Z. Sun, Recycling of spent lithium-ion batteries in view of lithium recovery: a critical review, J. Clean. Prod. 228 (2019) 801–813.
- [20] B. Makuzza, Q. Tian, X. Guo, K. Chattopadhyay, D. Yu, Pyrometallurgical options for recycling spent lithium-ion batteries: a comprehensive review, J. Power Sources 491 (2021), 229622, <https://doi.org/10.1016/j.jpowsour.2021.229622>.
- [21] M. Assefi, S. Maroufi, Y. Yamauchi, V. Sahajwalla, Pyrometallurgical recycling of Li-ion, Ni-Cd and Ni-MH batteries: a minireview, Curr. Opin. Green Sustain. Chem. 24 (2020) 26–31, <https://doi.org/10.1016/j.cogsc.2020.01.005>.
- [22] N. Li, J. Guo, Z. Chang, H. Dang, X. Zhao, S. Ali, W. Li, H. Zhou, C. Sun, Aqueous Leaching of Lithium from Simulated Pyrometallurgical Slag by Sodium Sulfate Roasting †, 2019, <https://doi.org/10.1039/c9ra03754c>.
- [23] A. Vezzini, Manufacturers, Materials and Recycling Technologies, Lithium-Ion Batteries: Advances and Applications, 2014, pp. 529–551, <https://doi.org/10.1016/B978-0-444-59513-3.00023-6>.
- [24] C. Hanisch, J. Diekmann, A. Steiger, W. Haselrieder, A. Kwade, Recycling of lithium-ion batteries, Handb. Clean Energy Syst. (2015) 1–24, <https://doi.org/10.1002/9781118991978.HCES221>.
- [25] L. Gaines, Lithium-ion battery recycling processes: research towards a sustainable course, Sustain. Mater. Technol. (2018) 17, <https://doi.org/10.1016/J.SUSMAT.2018.E00068>.
- [26] J. Li, L. Hu, H. Zhou, L. Wang, B. Zhai, S. Yang, P. Meng, R. Hu, Regenerating of LiNi0.5Co0.2Mn0.3O2 cathode materials from spent lithium-ion batteries, J. Mater. Sci. Mater. Electron. 29 (2018) 17661–17669, <https://doi.org/10.1007/S10854-018-9870-X/TABLES/3>.
- [27] C. Yi, L. Zhou, X. Wu, W. Sun, L. Yi, Y. Yang, Technology for recycling and regenerating graphite from spent lithium-ion batteries, Chin. J. Chem. Eng. 39 (2021) 37–50, <https://doi.org/10.1016/j.cjche.2021.09.014>.
- [28] M. Sethurajan, S. Gaydardzhiev, Bioprocessing of spent lithium ion batteries for critical metals recovery – a review, Resour. Conserv. Recycl. 165 (2021), 105225, <https://doi.org/10.1016/J.RESCONREC.2020.105225>.
- [29] J. Jegann Roy, S. Rarotra, V. Krikstolaityte, K. Wu Zhuoran, Y. Dja-la Cindy, X. Yi Tan, M. Carboni, D. Meyer, Q. Yan, M. Srinivasan, J.J. Roy, S. Rarotra, V. Krikstolaityte, K.W. Zhuoran, Y.D.-I. Cindy, Q. Yan, M. Srinivasan, X.Y. Tan, M. Carboni, D. Meyer, Green recycling methods to treat lithium-ion batteries E-waste: a circular approach to sustainability, Adv. Mater. 34 (2022), 2103346, <https://doi.org/10.1002/ADMA.202103346>.
- [30] H. Gao, D. Tran, Z. Chen, Seeking direct cathode regeneration for more efficient lithium-ion battery recycling, Opin. Electrochem. 31 (2022), 100875, <https://doi.org/10.1016/J.COELC.2021.100875>.
- [31] R. Sommerville, J. Shaw-Stewart, V. Goodship, N. Rowson, E. Kendrick, A review of physical processes used in the safe recycling of lithium ion batteries, Sustain. Mater. Technol. 25 (2020), <https://doi.org/10.1016/j.susmat.2020.e00197>.
- [32] R. Sommerville, P. Zhu, M.A. Rajaeifar, O. Heidrich, V. Goodship, E. Kendrick, A qualitative assessment of lithium ion battery recycling processes, Resour. Conserv. Recycl. 165 (2021), <https://doi.org/10.1016/j.resconrec.2020.105219>.
- [33] F. Stehmann, C. Bradtmöller, S. Scholl, Separation of the Electrolyte—Thermal Drying, Sustainable Production, Life Cycle Engineering and Management, 2018, pp. 139–153, [https://doi.org/10.1007/978-3-319-70572-9\\_8](https://doi.org/10.1007/978-3-319-70572-9_8).
- [34] P. Haas, S. Pfeifer, J. Müller, C. Bradtmöller, S. Scholl, Separation of the Electrolyte—Solvent Extraction BT - Recycling of Lithium-Ion Batteries: the LithoRec Way, in: A. Kwade, J. Diekmann (Eds.), Springer International Publishing, Cham, 2018, pp. 155–176, [https://doi.org/10.1007/978-3-319-70572-9\\_9](https://doi.org/10.1007/978-3-319-70572-9_9).
- [35] M. Grützke, V. Kraft, W. Weber, C. Wendt, A. Friesen, S. Klarmor, M. Winter, S. Nowak, Supercritical carbon dioxide extraction of lithium-ion battery

- electrolytes, *J. Supercrit. Fluids* 94 (2014) 216–222, <https://doi.org/10.1016/j.supflu.2014.07.014>.
- [36] European Commission, Proposal for a REGULATION of the EUROPEAN PARLIAMENT and of the COUNCIL Concerning Batteries and Waste Batteries, Repealing Directive 2006/66/EC and Amending Regulation (EU) No 2019/1020, 2020, <https://www.eib.org/en/press/all/2020-121-eib-reaffirms-commitment-to-a-european-battery-industry->.
- [37] J. Neumann, M. Petranikova, M. Meeus, J.D. Gamarra, R. Younesi, M. Winter, S. Nowak, Recycling of lithium-ion batteries—current state of the art, circular economy, and next generation recycling, *Adv. Energy Mater.* (2022), 2102917, <https://doi.org/10.1002/AENM.2020102917>.
- [38] E. Mossali, N. Picone, L. Gentilini, O. Rodriguez, J.M. Pérez, M. Colledani, Lithium-ion batteries towards circular economy: a literature review of opportunities and issues of recycling treatments, *J. Environ. Manag.* 264 (2020), <https://doi.org/10.1016/j.jenvman.2020.110500>.
- [39] X. Duan, W. Zhu, Z. Ruan, M. Xie, J. Chen, X. Ren, Recycling of lithium batteries—a review, *Energies* 15 (2022) 1611, <https://doi.org/10.3390/EN15051611>.
- [40] M. Mohr, M. Weil, J. Peters, Z. Wang, Recycling of lithium-ion batteries, *Encyclop. Electrochem.* (2020) 1–33, <https://doi.org/10.1002/9783527610426.bard110009>.
- [41] W. Mrozik, M.A. Rajaeifar, O. Heidrich, P. Christensen, Environmental impacts, pollution sources and pathways of spent lithium-ion batteries, *Energy Environ. Sci.* 14 (2021) 6099–6121, <https://doi.org/10.1039/DIEE00691F>.
- [42] Y.-L.L. Ding, Z.P. Cano, A.P. Yu, J. Lu, Z.W. Chen, Y.-L.L. Ding, Z.P. Cano, A.P. Yu, Z.W. Chen, J. Lu, *Automotive Li-Ion Batteries: Current Status and Future Perspectives*, 2019.
- [43] G. Zubi, R. Dufo-López, M. Carvalho, G. Pasaoglu, The lithium-ion battery: state of the art and future perspectives, *Renew. Sustain. Energy Rev.* 89 (2018) 292–308, <https://doi.org/10.1016/j.rser.2018.03.002>.
- [44] S. Doose, J.K. Mayer, P. Michalowski, A. Kwade, Challenges in ecofriendly battery recycling and closed material cycles: a perspective on future lithium battery generations, *Metals* 11 (2021) 1–17.
- [45] B. Huang, Z. Pan, X. Su, L. An, Recycling of lithium-ion batteries: recent advances and perspectives, *J. Power Sources* 399 (2018) 274–286.
- [46] N. Nitta, F. Wu, J.T. Lee, G. Yushin, Li-ion battery materials: present and future, *Mater. Today* 18 (2015) 252–264, <https://doi.org/10.1016/j.mattod.2014.10.040>.
- [47] J. (John T.) Warner, *The Handbook of Lithium-Ion Battery Pack Design : Chemistry, Components, Types and Terminology*, 2015.
- [48] J. Asenbauer, T. Eisemann, M. Kuenzel, A. Kazzazi, Z. Chen, D. Bresser, The success story of graphite as a lithium-ion anode material—fundamentals, remaining challenges, and recent developments including silicon (oxide) composites, *Sustain. Energy Fuels* 4 (2020) 5387–5416, <https://doi.org/10.1039/dose00175a>.
- [49] M.F. Lagadec, R. Zahn, V. Wood, Characterization and performance evaluation of lithium-ion battery separators, *Nat. Energy* 4 (2019) 16–25, <https://doi.org/10.1038/s41560-018-0295-9>.
- [50] P. Rozier, J.M. Tarascon, Review—Li-Rich layered oxide cathodes for next-generation Li-ion batteries: chances and challenges, *J. Electrochem. Soc.* 162 (2015) A2490–A2499, <https://doi.org/10.1149/2.01111514jes>.
- [51] B.L. Ellis, K.T. Lee, L.F. Nazar, Positive electrode materials for Li-Ion and Li-batteries, *Chem. Mater.* 22 (2010) 691–714, <https://doi.org/10.1021/cm902696j>.
- [52] J. Xu, F. Lin, M.M. Doeff, W. Tong, A review of Ni-based layered oxides for rechargeable Li-ion batteries, *J. Mater. Chem. B* 5 (2017) 874–901, <https://doi.org/10.1039/C6TA07991A>.
- [53] M. Marcinek, J. Syzdek, M. Marczewski, M. Piszcz, L. Niedzicki, M. Kalita, A. Plewa-Marczewska, A. Bitner, P. Wieczorek, T. Trzeciak, M. Kasprzyk, P. Łęzak, Z. Zukowska, A. Zalewska, W. Wieczorek, Electrolytes for Li-ion transport - review, *Solid State Ionics* 276 (2015) 107–126, <https://doi.org/10.1016/j.ssi.2015.02.006>.
- [54] C. Herrmann, S. Kara, A. Kwade, J. Diekmann, Sustainable Production, Life Cycle Engineering and Management, 2018. <http://www.springer.com/series/10615>.
- [55] R. Schröder, M. Aydemir, G. Seliger, Comparatively assessing different shapes of lithium-ion battery cells, *Procedia Manuf.* 8 (2017) 104–111, <https://doi.org/10.1016/j.promfg.2017.02.013>.
- [56] F.B.I. CRC, Li-ion Battery Cathode Manufacture in Australia A SCENE SETTING PROJECT FUTURE BATTERY INDUSTRIES CRC, 2020. <https://fbicrc.com.au/wp-content/uploads/2020/07/Li-ion-Battery-Cathode-Manufacturing-in-Aust-1.pdf>.
- [57] G. Zhao, *Reuse and Recycling of Lithium-Ion Power Batteries*, 2017.
- [58] Q. Dai, J. Spangenberger, S. Ahmed, L. Gaines, J.C. Kelly, M. Wang, EverBatt: A Closed-Loop Battery Recycling Cost and Environmental Impacts Model Energy Systems Division, 2019. [www.anl.gov](http://www.anl.gov).
- [59] A.W. Golubkov, S. Scheikl, R. Planteau, G. Voitig, H. Wiltsche, C. Stangl, G. Fauler, A. Thaler, V. Hacker, Thermal runaway of commercial 18650 Li-ion batteries with LFP and NCA cathodes - impact of state of charge and overcharge, *RSC Adv.* 5 (2015) 57171–57186, <https://doi.org/10.1039/C5RA05897J>.
- [60] K. Turcheniuk, D. Bondarev, G.G. Amatucci, G. Yushin, Battery materials for low-cost electric transportation, *Mater. Today* 42 (2021) 57–72, <https://doi.org/10.1016/j.mattod.2020.09.027>.
- [61] M. Herrmann, Packaging - materials review, in: AIP Conference Proceedings, American Institute of Physics Inc., 2014, pp. 121–133, <https://doi.org/10.1063/1.4878483>.
- [62] S.F. Tie, C.W. Tan, A review of energy sources and energy management system in electric vehicles, *Renew. Sustain. Energy Rev.* 20 (2013) 82–102, <https://doi.org/10.1016/j.rser.2012.11.077>.
- [63] J. Kim, J. Oh, H. Lee, Review on battery thermal management system for electric vehicles, *Appl. Therm. Eng.* 149 (2019) 192–212, <https://doi.org/10.1016/j.applthermaleng.2018.12.020>.
- [64] M.A. Hannan, M.S.H. Lipu, A. Hussain, A. Mohamed, A review of lithium-ion battery state of charge estimation and management system in electric vehicle applications: challenges and recommendations, *Renew. Sustain. Energy Rev.* 78 (2017) 834–854, <https://doi.org/10.1016/j.rser.2017.05.001>.
- [65] R.E. Ciez, J.F. Whitacre, Examining different recycling processes for lithium-ion batteries, *Nat. Sustain.* 2 (2019) 148–156.
- [66] J. Shaw-Stewart, A. Alvarez-Reguera, A. Greszta, J. Marco, M. Masood, R. Sommerville, E. Kendrick, Aqueous solution discharge of cylindrical lithium-ion cells, *Sustain. Mater. Technol.* 22 (2019), <https://doi.org/10.1016/j.susmat.2019.e00110>.
- [67] S. Ojanen, M. Lundström, A. Santasalo-Aarnio, R. Serna-Guerrero, Challenging the concept of electrochemical discharge using salt solutions for lithium-ion batteries recycling, *Waste Manag.* 76 (2018) 242–249, <https://doi.org/10.1016/J.WASMAN.2018.03.045>.
- [68] J. Xiao, J. Guo, L. Zhan, Z. Xu, A cleaner approach to the discharge process of spent lithium ion batteries in different solutions, *J. Clean. Prod.* 255 (2020), <https://doi.org/10.1016/j.jclepro.2020.120064>.
- [69] J. Nan, D. Han, X. Zuo, Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction, *J. Power Sources* 152 (2005) 278–284.
- [70] N.S. Nemehard, Safe, Sustainable Discharge of Electric Vehicle Batteries as a Pretreatment Step to Crushing in the Recycling Process, (n.d.).
- [71] G. Lombardo, B. Ebin, M.R. Mark, B.M. Steenari, M. Petranikova, Incineration of EV Lithium-ion batteries as a pretreatment for recycling – determination of the potential formation of hazardous by-products and effects on metal compounds, *J. Hazard Mater.* 393 (2020), <https://doi.org/10.1016/J.JHAZMAT.2020.122372>.
- [72] G. Lombardo, B. Ebin, M.R.J. St Foreman, B.M. Steenari, M. Petranikova, Chemical transformations in Li-ion battery electrode materials by carbothermic reduction, *ACS Sustain. Chem. Eng.* 7 (2019) 13668–13679, <https://doi.org/10.1021/ACSSUSCHEMENG.8B06540>.
- [73] F. Larouche, F. Tedjar, K. Amouzegar, G. Houlachi, P. Bouchard, G. P. Demopoulos, K. Zaghib, Progress and status of hydrometallurgical and direct recycling of Li-Ion batteries and beyond, *Materials* 13 (2020), <https://doi.org/10.3390/ma13030801>.
- [74] S. Nowak, M. Winter, The role of sub- and supercritical CO<sub>2</sub> as “processing solvent” for the recycling and sample preparation of lithium ion battery electrolytes, *Molecules* 22 (2017).
- [75] S.E. Sloop, Recycling Positive-Electrode Material of a Lithium-Ion Battery, US 2016/0043450 A1, 2016.
- [76] S.E. Sloop, System and Method for Removing an Electrolyte from an Energy Storage And/or Conversion Device Using a Supercritical Fluid, US 7198865 B2, 185, 2007, pp. 103–105.
- [77] S.E. Sloop, R. Parker, System and method for processing an end-of-life or reduced performance energy storage and/or conversion device using a supercritical fluid, U. S. Jpn. Outlook 8067107 B2 (2011).
- [78] S. Kim, J. Bang, J. Yoo, Y. Shin, J. Bae, J. Jeong, K. Kim, P. Dong, K. Kwon, A comprehensive review on the pretreatment process in lithium-ion battery recycling, *J. Clean. Prod.* (2021) 294, <https://doi.org/10.1016/J.JCLEPRO.2021.126329>.
- [79] L. Brückner, J. Frank, T. Elwert, Industrial recycling of lithium-ion batteries—a critical review of metallurgical process routes, *Metals* 10 (2020) 1–29, <https://doi.org/10.3390/MET10081107>.
- [80] J. Zhang, J. Hu, W. Zhang, Y. Chen, C. Wang, Efficient and economical recovery of lithium, cobalt, nickel, manganese from cathode scrap of spent lithium-ion batteries, *J. Clean. Prod.* 204 (2018) 437–446, <https://doi.org/10.1016/J.JCLEPRO.2018.09.033>.
- [81] H. Pinegar, Y.R. Smith, Recycling of end-of-life lithium ion batteries, Part I: commercial processes, *J. Sustain. Metal.* 5 (2019) 402–416, <https://doi.org/10.1007/S40831-019-00235-9>.
- [82] A. Sonoc, J. Jeswiet, V.K. Soo, Opportunities to improve recycling of automotive lithium ion batteries, in: *Procedia CIRP*, Elsevier B.V., 2015, pp. 752–757, <https://doi.org/10.1016/j.procir.2015.02.039>.
- [83] F. Saloojee, J. Lloyd, CRUNDWELL MANAGEMENT SOLUTIONS (PTY) LTD, T/a CM SOLUTIONS (PTY) LTD LITHIUM BATTERY RECYCLING PROCESS Desktop Study, 2014.
- [84] J.C.Y. Jung, P.C. Sui, J. Zhang, A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments, *J. Energy Storage* 35 (2021), 102217. <https://www.sciencedirect.com/science/article/pii/S2352152X20302405>.
- [85] L. Sun, K. Qiu, Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries, *J. Hazard Mater.* 194 (2011) 378–384, <https://doi.org/10.1016/J.JHAZMAT.2011.07.114>.
- [86] A.H. Kaksomen, N.J. Boxall, Y. Gumulya, H.N. Khaleque, C. Morris, T. Bohu, K. Y. Cheng, K.M. Usher, A.M. Lakaniemi, Recent progress in biohydrometallurgy and microbial characterisation, *Hydrometallurgy* 180 (2018) 7–25, <https://doi.org/10.1016/J.HYDROMET.2018.06.018>.
- [87] B.R. Khatri, D.R. Tipre, S.R. Dave, Comparison of hydro- and biohydrometallurgical extraction of metals from waste Li-ion batteries of cell

- phone, J. Sustain. Metal. 5 (2019) 250–261, <https://doi.org/10.1007/S40831-019-00223-Z/FIGURES/6>.
- [88] R. Tao, P. Xing, H. Li, Z. Sun, Y. Wu, Recovery of spent LiCoO<sub>2</sub> lithium-ion battery via environmentally friendly pyrolysis and hydrometallurgical leaching, Resour. Conserv. Recycl. (2022) 176, <https://doi.org/10.1016/J.RESCONREC.2021.105921>.
- [89] T. Huang, L. Liu, S. Zhang, Recovery of cobalt, lithium, and manganese from the cathode active materials of spent lithium-ion batteries in a bio-electro-hydrometallurgical process, Hydrometallurgy 188 (2019) 101–111, <https://doi.org/10.1016/j.hydromet.2019.06.011>.
- [90] J.T. Marcos, C. Scheller, R. Godina, T.S. Spengler, H. Carvalho, Sources of uncertainty in the closed-loop supply chain of lithium-ion batteries for electric vehicles, Clean. Log. Supply Chain 1 (2021), 100006, <https://doi.org/10.1016/J.CLSCN.2021.100006>.
- [91] J. Diekmann, C. Hanisch, L. Froböse, G. Schälicke, T. Loellhoeffel, A.-S. Fölster, A. Kwade, Ecological recycling of lithium-ion batteries from electric vehicles with focus on mechanical processes, J. Electrochem. Soc. 164 (2017) A6184–A6191, <https://doi.org/10.1149/2.0271701JES>.
- [92] X. Song, T. Hu, C. Liang, H.L. Long, L. Zhou, W. Song, L. You, Z.S. Wu, J.W. Liu, Direct regeneration of cathode materials from spent lithium iron phosphate batteries using a solid phase sintering method, RSC Adv. 7 (2017) 4783–4790, <https://doi.org/10.1039/C6RA27210J>.
- [93] X. Li, J. Zhang, D. Song, J. Song, L. Zhang, Direct regeneration of recycled cathode material mixture from scrapped LiFePO<sub>4</sub> batteries, J. Power Sources 345 (2017) 78–84, <https://doi.org/10.1016/j.jpowsour.2017.01.118>.
- [94] J. Li, Y. Wang, L. Wang, B. Liu, H. Zhou, A facile recycling and regeneration process for spent LiFePO<sub>4</sub> batteries, J. Mater. Sci. Mater. Electron. 30 (2019) 14580–14588, <https://doi.org/10.1007/S10854-019-01830-Y>.
- [95] T. Wang, X. Yu, M. Fan, Q. Meng, Y. Xiao, Y.X. Yin, H. Li, Y.G. Guo, Direct regeneration of spent LiFePO<sub>4</sub> via a graphite prelithiation strategy, Chem. Commun. 56 (2019) 245–248, <https://doi.org/10.1039/C9CC08155K>.
- [96] C. Wu, J. Hu, L. Ye, Z. Su, X. Fang, X. Zhu, L. Zhuang, X. Ai, H. Yang, J. Qian, Direct regeneration of spent Li-ion battery cathodes via chemical relithiation reaction, ACS Sustain. Chem. Eng. 9 (2021) 16384–16393, [https://doi.org/10.1021/ACSSUSCHEMENG.1C06278/ASSET/IMAGES/LARGE/SC1C06278\\_0007.JPG](https://doi.org/10.1021/ACSSUSCHEMENG.1C06278/ASSET/IMAGES/LARGE/SC1C06278_0007.JPG).
- [97] L. Zhang, Z. Xu, Z. He, Electrochemical relithiation for direct regeneration of LiCoO<sub>2</sub>Materials from spent lithium-ion battery electrodes, ACS Sustain. Chem. Eng. 8 (2020) 11596–11605, [https://doi.org/10.1021/ACSSUSCHEMENG.0C02854/ASSET/IMAGES/LARGE/SC0C02854\\_0008.JPG](https://doi.org/10.1021/ACSSUSCHEMENG.0C02854/ASSET/IMAGES/LARGE/SC0C02854_0008.JPG).
- [98] Y. Yang, S. Xu, Y. He, Lithium recycling and cathode material regeneration from acid leach liquor of spent lithium-ion battery via facile co-extraction and co-precipitation processes, Waste Manag. 64 (2017) 219–227, <https://doi.org/10.1016/j.wasman.2017.03.018>.
- [99] Y. Yang, S. Lei, S. Song, W. Sun, L. Wang, Stepwise recycling of valuable metals from Ni-rich cathode material of spent lithium-ion batteries, Waste Manag. 102 (2020) 131–138, <https://doi.org/10.1016/J.WASMAN.2019.09.044>.
- [100] S. Jin, D. Mu, Z. Lu, R. Li, Z. Liu, Y. Wang, S. Tian, C. Dai, A comprehensive review on the recycling of spent lithium-ion batteries: urgent status and technology advances, J. Clean. Prod. 340 (2022), 130535, <https://doi.org/10.1016/J.JCLEPRO.2022.130535>.
- [101] E. Fan, L. Li, Z. Wang, J. Lin, Y. Huang, Y. Yao, R. Chen, F. Wu, Sustainable recycling technology for Li-ion batteries and beyond: challenges and future prospects, Chem. Rev. 120 (2020) 7020–7063, <https://doi.org/10.1021/acs.chemrev.9b00535>.
- [102] M.A. Cusenza, S. Bobba, F. Ardente, M. Cellura, F. di Persio, Energy and environmental assessment of a traction lithium-ion battery pack for plug-in hybrid electric vehicles, J. Clean. Prod. 215 (2019) 634–649, <https://doi.org/10.1016/j.jclepro.2019.01.056>.
- [103] A. Boyden, V.K. Soo, M. Doolan, The environmental impacts of recycling portable lithium-ion batteries, in: Procedia CIRP, Elsevier B.V., 2016, pp. 188–193, <https://doi.org/10.1016/j.procir.2016.03.100>.
- [104] T.P. Hendrickson, O. Kavvada, N. Shah, R. Sathre, C. D Scown, Life-cycle implications and supply chain logistics of electric vehicle battery recycling in California, Environ. Res. Lett. 10 (2015), 014011, <https://doi.org/10.1088/1748-9326/10/1/014011>.
- [105] K. Richa, C.W. Babbitt, G. Gaustad, Eco-efficiency analysis of a lithium-ion battery waste hierarchy inspired by circular economy, J. Ind. Ecol. 21 (2017) 715–730, <https://doi.org/10.1111/JIEC.12607>.
- [106] R. Sattar, S. Ilyas, H.N. Bhatti, A. Ghaffar, Resource recovery of critically-rare metals by hydrometallurgical recycling of spent lithium ion batteries, Separ. Purif. Technol. 209 (2019) 725–733, <https://doi.org/10.1016/j.seppur.2018.09.019>.
- [107] E. Asadi Dalini, G. Karimi, S. Zandevakili, M. Goodarzi, A Review on Environmental, Economic and Hydrometallurgical Processes of Recycling Spent Lithium-Ion Batteries, 2020, pp. 1–22, <https://doi.org/10.1080/08827508.2020.1781628>, <https://doi.org/10.1080/08827508.2020.1781628>.
- [108] International Energy Agency, Global supply chains of EV batteries, n.d. [www.iea.org/t&c/](http://www.iea.org/t&c/). (Accessed 20 July 2022).
- [109] M.C.C. Lima, L.P. Pontes, A.S.M. Vasconcelos, W. de Araujo Silva Junior, K. Wu, Economic aspects for recycling of used lithium-ion batteries from electric vehicles, Energies 15 (2022) 2203, <https://doi.org/10.3390/EN15062203>, 15 (2022) 2203.
- [110] D. Steward, A. Mayyas, M. Mann, Economics and challenges of Li-ion battery recycling from end-of-life vehicles, Procedia Manuf. 33 (2019) 272–279, <https://doi.org/10.1016/j.promfg.2019.04.033>.
- [111] J. Spangenberger, L. Gaines, Q. Dai, Comparison of Lithium-Ion Battery Recycling Processes Using the ReCell Model, 2018. Fort Lauderdale, FL.
- [112] Y. Yao, M. Zhu, Z. Zhao, B. Tong, Y. Fan, Z. Hua, Hydrometallurgical processes for recycling spent lithium-ion batteries: a critical review, ACS Sustain. Chem. Eng. 6 (2018) 13611–13627, <https://doi.org/10.1021/ACSSUSCHEMENG.8B03545>.
- [113] S.E. Sloop, J.E. Trevey, L. Gaines, M.M. Lerner, W. Xu, Advances in direct recycling of lithium-ion electrode materials, ECS Trans. 85 (2018) 397–403, <https://doi.org/10.1149/08513.0397ECST>.
- [114] S. Sloop, L. Crandon, M. Allen, K. Koetje, L. Reed, L. Gaines, W. Sirisaksoontorn, M. Lerner, A direct recycling case study from a lithium-ion battery recall, Sustain. Mater. Technol. 25 (2020), <https://doi.org/10.1016/j.susmat.2020.e00152>.
- [115] P. Xu, D.H.S. Tan, Z. Chen, Emerging trends in sustainable battery chemistries, Trend Chem. 3 (2021) 620–630, <https://doi.org/10.1016/J.TRECHM.2021.04.007>.
- [116] E. Gerold, S. Luidold, H. Antrekowitsch, Selective precipitation of metal oxalates from lithium ion battery leach solutions, Metals 10 (2020) 1–15, <https://doi.org/10.3390/MET10111435>.
- [117] M. Jo, H. Ku, S. Park, J. Song, K. Kwon, Effects of Residual Lithium in the precursors of Li[Ni<sub>1/3</sub>Co<sub>1/3</sub>Mn<sub>1/3</sub>]O<sub>2</sub> on their lithium-ion battery performance, J. Phys. Chem. Solid. 118 (2018) 47–52, <https://doi.org/10.1016/j.jpc.2018.02.043>.
- [118] M. Chen, X. Ma, B. Chen, R. Arsenault, P. Karlson, N. Simon, Y. Wang, Recycling end-of-life electric vehicle lithium-ion batteries, Joule 3 (2019) 2622–2646, <https://doi.org/10.1016/j.joule.2019.09.014>.
- [119] R. Danino-Perraud, The Recycling of liThium-ion BaTTeRies A Strategic Pillar for the European Battery Alliance études de l'Ifri Raphaël Danino-Perraud Center for Energy, 2020. <https://www.ifri.org/en/publications/etudes-de-lifri/recycling-lithium-ion-batteries-strategic-pillar-european-battery>.
- [120] D. Naberezhnykh, Recycling of Tracked Li-Ion EV Batteries Final Project Report, Transport Research Laboratory, 2013.
- [121] K. Environmetal, G. INC, M. Environmental, Research Study on Reuse and Recycling of Batteries Employed in Electric Vehicles: the Technical, Environmental, Economic, Energy and Cost Implications of Reusing and Recycling EV Batteries Project Report, 2019. [https://www.api.org/~media/File/s/Oil-and-Natural-Gas/Fuels/Kelleher\\_Final\\_EV\\_Battery\\_Reuse\\_and\\_Recycling\\_Report\\_to\\_API\\_18Sept2019\\_edits\\_18Dec2019.pdf](https://www.api.org/~media/File/s/Oil-and-Natural-Gas/Fuels/Kelleher_Final_EV_Battery_Reuse_and_Recycling_Report_to_API_18Sept2019_edits_18Dec2019.pdf).
- [122] R. Sojka, Q. Pan, L. Billmann, Comparative Study of Li-Ion Battery Recycling Processes, 2020. <https://accurec.de/wp-content/uploads/2021/04/Accurec-Co-mparative-study.pdf>.
- [123] A. Sattar, D. Greenwood, M. Dowson, P. Unadkat, Automotive Lithium Ion Battery Recycling in the UK Based on a Feasibility Study, REPORT SUMMARY, 2020.
- [124] L. Bravo Diaz, X. He, Z. Hu, F. Restuccia, M. Marinescu, J.V. Barreras, Y. Patel, G. Offer, G. Rein, Review of fire safety of lithium-ion batteries: industry challenges and research contributions, J. Electrochem. Soc. 167 (2020), 90559, <https://doi.org/10.1149/1945-7111/ABA8B9>.
- [125] L. Gaines, K. Richa, J. Spangenberger, Key issues for Li-ion battery recycling, MRS Energy Sustain. 5 (2018), <https://doi.org/10.1557/MRE.2018.13>.
- [126] Umicore Group, Umicore - Our recycling process, in: <https://csm.uminicore.com/en/battery-recycling/our-recycling-process/>, 2019.
- [127] Duesenfeld GmbH, Ecofriendly recycling of lithium-ion batteries. [https://www.duesenfeld.com/recycling\\_en.html](https://www.duesenfeld.com/recycling_en.html), 2022.
- [128] J. Pudas, AkkuSer Oy mobile phone and battery recycling services. [https://ec.europa.eu/environment/archives/ecoinnovation2011/2nd\\_forum/presentations/Session\\_2011](https://ec.europa.eu/environment/archives/ecoinnovation2011/2nd_forum/presentations/Session_2011).
- [129] Akkuser Patient US8979006 Copia, (n.d.).
- [130] F. Tedjar, J.C. Foudraz, Method for the Mixed Recycling of Lithium-Based Anode Batteries and Cells, US 7820317 B2, 2010.
- [131] SNAM, Collection and recycling of batteries, hybrid and electric vehicle sector (n.d.), <https://www.snam.com/>. (Accessed 26 May 2022).
- [132] Recycling - Erasteel (n.d.), <https://www.erasteel.com/technologies/recycling/>. (Accessed 26 May 2022).
- [133] E.D. Industrie, Euro Dieuze Industrie report. [https://elibaba.files.wordpress.com/2013/01/euro\\_dieuze\\_industrie.pdf](https://elibaba.files.wordpress.com/2013/01/euro_dieuze_industrie.pdf), 2013.
- [134] Redux GmbH, Redux smart battery recycling. <https://www.redox-recycling.com/de>, 2022.
- [135] Neometals, Neometals company announcement. 7 May 2021. <https://www.criticalmetals.eu/reports/132-210507-NMT-LiB-Recycling—Outstanding-Cost>, 2021.
- [136] Northvolt - the future of energy | Northvolt (n.d.), <https://northvolt.com/>. (Accessed 26 May 2022).
- [137] For a cleaner world | Fortum (n.d.), <https://www.fortum.com/>. (Accessed 26 May 2022).
- [138] Resource management and recycling solutions | Stena Recycling (n.d.), <http://www.stenarecycling.com/>. (Accessed 26 May 2022).
- [139] W.N. Smith, S. Swaffer, Process for Recovering and Regenerating Lithium-Ion Cathode Material from Lithium-Ion Batteries, US 8882007 B1, 2014.
- [140] A.M. Bernardes, D.C.R. Espinosa, J.A.S. Tenório, Recycling of batteries: a review of current processes and technologies, J. Power Sources 130 (2004) 291–298, <https://doi.org/10.1016/J.JPOWSOUR.2003.12.026>.
- [141] A. Kwade, J. Diekmann, Recycling of Lithium-Ion Batteries. The LithoRec Way, Springer International Publishing, Cham, 2018, <https://doi.org/10.1007/978-3-319-70572-9>.
- [142] B. Davis, K. Watson, A. Roy, A. Kochhar, D. Tait, Li-cycle—a case study in integrated process development, Minerals, Metals and Materials Series (2019) 247–260, [https://doi.org/10.1007/978-3-030-10386-6\\_29](https://doi.org/10.1007/978-3-030-10386-6_29).

- [143] N. Chow, J.-C.-Y. Jung, A.M. Nacu, D.D. Warkentin, Processing of Cobaltous Sulphate/dithionate Liquors Derived from Cobalt Resource. US 10308523 B1, 183, 2019, p. 343.
- [144] A.M. Inc, American Manganese Inc Presentation June 2021, 2021.
- [145] D. Morin, C. Gagnebourque, E. Nadeau, B. Couture, Lithium-ion Batteries Recycling Process. WO 2019/060996 A1, 2019.
- [146] 성길하이메탈 - SungEel HiMetal (n.d.), <http://www.sungeel.com/>. (Accessed 26 May 2022).
- [147] Huayou Cobalt Co., Ltd., Chemical products (n.d.), <http://en.huayou.com/>. (Accessed 26 May 2022).
- [148] Highpower Technology (n.d.), <https://www.highpowertech.com/>. (Accessed 26 May 2022).
- [149] D. Chéret, S. Santen, Battery Recycling. US 7169206 B2, 2007.
- [150] T. Or, S.W.D.D. Gourley, K. Kaliyappan, A. Yu, Z. Chen, Recycling of mixed cathode lithium-ion batteries for electric vehicles: current status and future outlook, Carbon Energy 2 (2020) 6–43, <https://doi.org/10.1002/cey.229>.
- [151] J. Kang, G. Senanayake, J. Sohn, S.M. Shin, Recovery of cobalt sulfate from spent lithium ion batteries by reductive leaching and solvent extraction with Cyanex 272, Hydrometallurgy 100 (2010) 168–171, <https://doi.org/10.1016/j.hydromet.2009.10.010>.
- [152] K. Tanong, J.-F. Blais, G. Mercier, Metal recycling technologies for battery waste, Recent Pat. Eng. 8 (2014) 13–23.
- [153] K.M. Winslow, S.J. Laux, T.G. Townsend, A review on the growing concern and potential management strategies of waste lithium-ion batteries, Resour. Conserv. Recycl. 129 (2018) 263–277, <https://doi.org/10.1016/j.resconrec.2017.11.001>.
- [154] J. Li, G. Wang, Z. Xu, Generation and detection of metal ions and volatile organic compounds (VOCs) emissions from the pretreatment processes for recycling spent lithium-ion batteries, Waste Manag. 52 (2016) 221–227, <https://doi.org/10.1016/j.wasman.2016.03.011>.
- [155] G. Ren, S. Xiao, M. Xie, B. Pan, J. Chen, F. Wang, X. Xia, Recovery of valuable metals from spent lithium ion batteries by smelting reduction process based on FeO-SiO<sub>2</sub> – Al 2 O 3 slag system, Trans. Nonferrous Metals Soc. China 27 (2017) 450–456, [https://doi.org/10.1016/S1003-6326\(17\)60051-7](https://doi.org/10.1016/S1003-6326(17)60051-7).
- [156] A. Wittkowski, T. Schirmer, H. Qiu, D. Goldmann, U.E.A. Fittschen, Speciation of manganese in a synthetic recycling slag relevant for lithium recycling from lithium-ion batteries, Metals 11 (2021) 188, <https://doi.org/10.3390/met11020188>.
- [157] J. Klímková, D. Oráč, A. Miskufová, C. Vonderstein, C. Dertmann, M. Sommerfeld, B. Friedrich, T. Havlík, A combined pyro- and hydrometallurgical approach to recycle pyrolyzed lithium-ion battery black mass Part 2: lithium recovery from Li enriched slag—thermodynamic study, kinetic study, and dry digestion, Metals 10 (2020) 1558, <https://doi.org/10.3390/met10111558>.
- [158] Q. Cheng, Y. Han, Y. You, C. Hou, P.T. Wulandari, R.A. Amiliana, M. Huda, F. A. Kusumadewi, IOP conference series: earth and environmental science you may also like effect of different reductants on leaching lithium and cobalt from lithium ion batteries in tartaric acid solution cathode materials from spent power lithium-ion batteries acquisition of Co metal from spent lithium-ion battery with emulsion liquid membrane technology using cyanex 272 as extractant. <https://doi.org/10.1088/1755-1315/105/1/012044>, 2018, 105.
- [159] X. Jing, Z. Wu, D. Zhao, S. Li, F. Kong, Y. Chu, Environmentally Friendly Extraction and Recovery of Cobalt from Simulated Solution of Spent Ternary Lithium Batteries Using the Novel Ionic Liquids of [C 8 H 17 NH 2 ] [Cyanex 272], 2021, <https://doi.org/10.1021/acssuschemeng.0c07470>.
- [160] V.N.H. Nguyen, T.H. Nguyen, M.S. Lee, Review on the comparison of the chemical reactivity of cyanex 272, Cyanex 301 and Cyanex 302 for Their Application to Metal Separation from Acid Media, Metals 10 (2020) 1105, <https://doi.org/10.3390/MET10081105>, 10 (2020) 1105.
- [161] G. Alval-Hein, H. Mahendra, A. Ghahreman, Separation and recovery of cobalt and nickel from end of life products via solvent extraction technique: a review, J. Clean. Prod. 297 (2021), <https://doi.org/10.1016/J.JCLEPRO.2021.126592>.
- [162] F. Arshad, L. Li, K. Amin, E. Fan, N. Manurkar, A. Ahmad, J. Yang, F. Wu, R. Chen, A comprehensive review of the advancement in recycling the anode and electrolyte from spent lithium ion batteries, ACS Sustain. Chem. Eng. 8 (2020) 13527–13554, <https://doi.org/10.1021/acssuschemeng.0c04940>.
- [163] S. Al-Thyabat, T. Nakamura, E. Shibata, A. Iizuka, Adaptation of minerals processing operations for lithium-ion (LiBs) and nickel metal hydride (NiMH) batteries recycling: critical review, Miner. Eng. 45 (2013) 4–17, <https://doi.org/10.1016/j.mineng.2012.12.005>.
- [164] M. Grützke, X. Mönnighoff, F. Horsthemke, V. Kraft, M. Winter, S. Nowak, Extraction of lithium-ion battery electrolytes with liquid and supercritical carbon dioxide and additional solvents, RSC Adv. 5 (2015) 43209–43217, <https://doi.org/10.1039/c5ra04451k>.
- [165] Hanisch, Recycling Method for Treating Used Batteries in Particular Rechargeable Batteries, and Battery Processing Installation. US 2019/0260101 A1, 2019.
- [166] C. Hanisch, W. Haselrieder, A. Kwade, Method for Reclaiming Active Material from a Galvanic Cell, and an Active Material Separation Installation, Particularly an Active Metal Separation Installation. US 9780419 B2, 2017.
- [167] C. Hanisch, B. Westphal, W. Haselrieder, M. Schoenitz, Method for Treatment of Used Batteries in Particular Rechargeable Batteries, and Battery Processing Installation. US 2018/0301769 A1, 2018.
- [168] E. Gratz, Q. Sa, D. Apelian, Y. Wang, A closed loop process for recycling spent lithium ion batteries, J. Power Sources 262 (2014) 255–262, <https://doi.org/10.1016/j.jpowsour.2014.03.126>.
- [169] H. Zou, E. Gratz, D. Apelian, Y. Wang, A novel method to recycle mixed cathode materials for lithium ion batteries, Green Chem. 15 (2013) 1183–1191, <https://doi.org/10.1039/c3cg40182k>.
- [170] Q. Sa, E. Gratz, M. He, W. Lu, D. Apelian, Y. Wang, Synthesis of high performance LiNi<sub>1/3</sub>Mn<sub>1/3</sub>Co<sub>1/3</sub>O<sub>2</sub> from lithium ion battery recovery stream, J. Power Sources 282 (2015) 140–145, <https://doi.org/10.1016/j.jpowsour.2015.02.046>.
- [171] H. Rouhi, E. Karola, R. Serna-Guerrero, A. Santasalo-Aarnio, Voltage behavior in lithium-ion batteries after electrochemical discharge and its implications on the safety of recycling processes, J. Energy Storage 35 (2021), 102323, <https://doi.org/10.1016/j.est.2021.102323>.
- [172] J. Shu, M. Shui, D. Xu, D. Wang, Y. Ren, S. Gao, A comparative study of overdischarge behaviors of cathode materials for lithium-ion batteries, J. Solid State Electrochem. 16 (2012) 819–824, <https://doi.org/10.1007/S10008-011-1484-7>.
- [173] J. Piątek, S. Afyon, T.M. Budnyak, S. Budnyk, M.H. Sipponen, A. Slabon, Sustainable Li-ion batteries: chemistry and recycling, Adv. Energy Mater. 11 (2021), <https://doi.org/10.1002/AENM.202003456>.
- [174] Z. Liang, C. Cai, G. Peng, J. Hu, H. Hou, B. Liu, S. Liang, K. Xiao, S. Yuan, J. Yang, Hydrometallurgical recovery of spent lithium ion batteries: environmental strategies and sustainability evaluation, ACS Sustain. Chem. Eng. 9 (2021) 5750–5767, <https://doi.org/10.1021/acssuschemeng.1c00942>.
- [175] S. Lei, W. Sun, Y. Yang, Solvent extraction for recycling of spent lithium-ion batteries, J. Hazard Mater. 424 (2022), 127654, <https://doi.org/10.1016/j.jhazmat.2021.127654>.
- [176] L. Schwich, T. Schubert, B. Friedrich, Early-stage recovery of lithium from tailored thermal conditioned black mass part ii: mobilizing lithium via supercritical co2-carbonation, Metals 11 (2021) 1–30, <https://doi.org/10.3390/MET11020177>.
- [177] Y. Liu, D. Mu, R. Zheng, C. Dai, Supercritical CO<sub>2</sub> extraction of organic carbonate-based electrolytes of lithium-ion batteries, RSC Adv. 4 (2014) 54525–54531, <https://doi.org/10.1039/C4RA10530C>.
- [178] Y. Liu, D. Mu, Y. Dai, Q. Ma, R. Zheng, C. Dai, Analysis on extraction behaviour of lithium-ion battery electrolyte solvents in supercritical CO<sub>2</sub> by gas chromatography, Int. J. Electrochem. Sci. 11 (2016) 7594–7604, <https://doi.org/10.20964/2016.09.03>.
- [179] C. Hanisch, Recycling Method for Treating Used Batteries, in Particular Rechargeable Batteries, and Battery Processing Installation. US 20210175556 A1, 2021.
- [180] G. Zhang, X. Yuan, Y. He, H. Wang, T. Zhang, W. Xie, Recent advances in pretreating technology for recycling valuable metals from spent lithium-ion batteries, J. Hazard Mater. 406 (2021), <https://doi.org/10.1016/j.jhazmat.2020.124332>.
- [181] S. Krüger, C. Hanisch, A. Kwade, M. Winter, S. Nowak, Effect of impurities caused by a recycling process on the electrochemical performance of Li [Ni<sub>0.33</sub>Co<sub>0.33</sub>Mn<sub>0.33</sub>]O<sub>2</sub>, J. Electroanal. Chem. 726 (2014) 91–96, <https://doi.org/10.1016/j.jelechem.2014.05.017>.
- [182] C. Hanisch, T. Loellhoeffel, J. Diekmann, K.J. Markley, W. Haselrieder, A. Kwade, Recycling of lithium-ion batteries: a novel method to separate coating and foil of electrodes, J. Clean. Prod. 108 (2015) 301–311, <https://doi.org/10.1016/j.jclepro.2015.08.026>.
- [183] T. Georgi-Maschler, B. Friedrich, R. Weyhe, H. Heegn, M. Rutz, Development of a recycling process for Li-ion batteries, J. Power Sources 207 (2012) 173–182, <https://doi.org/10.1016/j.jpowsour.2012.01.152>.
- [184] N. Vieceli, R. Casasola, G. Lombardo, B. Ebin, M. Petranikova, C.A. Nogueira, C. Guimarães, M.F.C. Pereira, F.O. Durão, F. Margarido, Hydrometallurgical recycling of lithium-ion batteries by reductive leaching with sodium metabisulphite, Waste Manag. 71 (2018) 350–361, <https://doi.org/10.1016/j.wasman.2017.09.032>.
- [185] J. Kumar, R.R. Neiber, J. Park, R. Ali Soomro, G.W. Greene, S. Ali Mazari, H. Young Seo, J. Hong Lee, M. Shon, D. Wook Chang, K. Yong Cho, Recent progress in sustainable recycling of LiFePO<sub>4</sub>-type lithium-ion batteries: strategies for highly selective lithium recovery, Chem. Eng. J. 431 (2022), <https://doi.org/10.1016/j.cej.2021.133993>.
- [186] S. Dhiman, B. Gupta, Partition studies on cobalt and recycling of valuable metals from waste Li-ion batteries via solvent extraction and chemical precipitation, J. Clean. Prod. 225 (2019) 820–832.
- [187] L. Chen, X. Tang, Y.Y. Zhang, L. Li, Z. Zeng, Y.Y. Zhang, Process for the recovery of cobalt oxalate from spent lithium-ion batteries, Hydrometallurgy 108 (2011) 80–86, <https://doi.org/10.1016/j.hydromet.2011.02.010>.
- [188] X. Chen, Y. Chen, T. Zhou, D. Liu, H. Hu, S. Fan, Hydrometallurgical recovery of metal values from sulfuric acid leaching liquor of spent lithium-ion batteries, Waste Manag. 38 (2015) 349–356, <https://doi.org/10.1016/j.wasman.2014.12.023>.
- [189] M. Jafari, M.M. Torabian, A. Bazargan, A facile chemical-free cathode powder separation method for lithium ion battery resource recovery, J. Energy Storage 31 (2020), 101564, <https://doi.org/10.1016/J.EST.2020.101564>.
- [190] M.M. Torabian, M. Jafari, A. Bazargan, Discharge of lithium-ion batteries in salt solutions for safer storage, transport, and resource recovery, Waste Manag. Res.: The Journal for a Sustainable Circular Economy 40 (2022) 402–409, <https://doi.org/10.1177/0734242X211022658>.
- [191] T.R.B. Grandjean, J. Groenewald, J. Marco, The experimental evaluation of lithium ion batteries after flash cryogenic freezing, J. Energy Storage 21 (2019) 202–215, <https://doi.org/10.1016/j.est.2018.11.027>.
- [192] F. Cardarelli, J. Dube, Method for Recycling Spent Lithium Metal Polymer Rechargeable Batteries and Related Materials. US7192564B2, 2007.
- [193] J. Pudas, A. Erkkila, J. Viljamaa, Battery Recycling Method. US8979006 B2, 2015.
- [194] H. Pinegar, Y.R. Smith, Recycling of end-of-life lithium-ion batteries, Part II: laboratory-scale research developments in mechanical, thermal, and leaching

- treatments, *J. Sustain. Metal.* 6 (2020) 142–160, <https://doi.org/10.1007/s40831-020-00265-8>.
- [195] G. Prabaharan, S.P. Barik, N. Kumar, L. Kumar, Electrochemical process for electrode material of spent lithium ion batteries, *Waste Manag.* 68 (2017) 527–533.
- [196] S.A. Silvester, I.S. Lowndes, S.W. Kingman, A. Arroussi, Improved dust capture methods for crushing plant, *Appl. Math. Model.* 31 (2007) 311–331, <https://doi.org/10.1016/J.APM.2005.11.005>.
- [197] S.C. Chelgani, M. Parian, P.S. Parapari, Y. Ghorbani, J. Rosenkranz, A comparative study on the effects of dry and wet grinding on mineral flotation separation—a review, *J. Mater. Res. Technol.* 8 (2019) 5004–5011, <https://doi.org/10.1016/J.JMRT.2019.07.053>.
- [198] S.M. Shin, N.H. Kim, J.S. Sohn, D.H. Yang, Y.H. Kim, Development of a metal recovery process from Li-ion battery wastes, *Hydrometallurgy* 79 (2005) 172–181.
- [199] J. Li, Y. Jiang, Z. Xu, Eddy current separation technology for recycling printed circuit boards from crushed cell phones, *J. Clean. Prod.* 141 (2017) 1316–1323, <https://doi.org/10.1016/J.JCLEPRO.2016.09.144>.
- [200] H. Bi, H. Zhu, L. Zu, Y. Gao, S. Gao, Z. Wu, Eddy current separation for recovering aluminum and lithium–iron phosphate components of spent lithium–iron phosphate batteries, *Waste Manag. Res.* 37 (2019) 1217–1228, <https://doi.org/10.1177/0734242X19871610>.
- [201] J. Diekmann, M. Grützke, T. Loellhoeffel, M. Petermann, S. Rothermel, M. Winter, S. Nowak, A. Kwade, Potential Dangers during the Handling of Lithium-Ion Batteries, Sustainable Production, Life Cycle Engineering and Management, 2018, pp. 39–51, [https://doi.org/10.1007/978-3-319-70572-9\\_3](https://doi.org/10.1007/978-3-319-70572-9_3).
- [202] D. Yu, Z. Huang, B. Makuza, X. Guo, Q. Tian, Pretreatment options for the recycling of spent lithium-ion batteries: a comprehensive review, *Miner. Eng.* 173 (2021), 107218, <https://doi.org/10.1016/J.MINENG.2021.107218>.
- [203] A. Sarkar, R. May, S. Ramesh, W. Chang, L.E. Marbella, Recovery and Reuse of Composite Cathode Binder in Lithium Ion Batteries, 2021, <https://doi.org/10.1002/open.202100060>.
- [204] J. Li, Y. Lu, T. Yang, D. Ge, D.L. Wood, Z. Li, Water-based electrode manufacturing and direct recycling of lithium-ion battery electrodes—a green and sustainable manufacturing system, *iScience* 23 (2020), 101081, <https://doi.org/10.1016/j.isci.2020.101081>.
- [205] W.L. Dalmijn, T.P.R. de Jong, The development of vehicle recycling in Europe: sorting, shredding, and separation, *J. Occup. Med.* 59 (2007) 11, <https://doi.org/10.1007/S11837-007-0141-1>, 59 (2007) 52–56.
- [206] H. Wang, B. Friedrich, Innovative recycling of Li-based electric vehicle batteries Innovatives Recycling von Li-basierten Batterien für Elektrofahrzeuge, 2013.
- [207] A. Kochhar, T.G. Johnston, Process, Apparatus and System for Recovering Materials from Batteries. US 10919046 B2, 2021.
- [208] R. Zhan, Z. Oldenburg, L. Pan, Recovery of active cathode materials from lithium-ion batteries using froth flotation, *Sustain. Mater. Technol.* 17 (2018), <https://doi.org/10.1016/j.susmat.2018.e00062>.
- [209] H. Shin, R. Zhan, K.S. Dhindsa, L. Pan, T. Han, Electrochemical performance of recycled cathode active materials using froth flotation-based separation process, *J. Electrochim. Soc.* 167 (2020) 20504, <https://doi.org/10.1149/1945-7111/AB6280>.
- [210] J. Liu, H. Wang, T. Hu, X. Bai, S. Wang, W. Xie, J. Hao, Y. He, Recovery of LiCoO<sub>2</sub> and graphite from spent lithium-ion batteries by cryogenic grinding and froth flotation, *Miner. Eng.* (2020) 148.
- [211] J. Wang, Y. Zhang, K. Cui, T. Fu, J. Gao, S. Hussain, T.S. AlGarni, Pyrometallurgical recovery of zinc and valuable metals from electric arc furnace dust – a review, *J. Clean. Prod.* 298 (2021), 126788, <https://doi.org/10.1016/J.JCLEPRO.2021.126788>.
- [212] M. Esmaeili, S.O. Rastegar, R. Beigzadeh, T. Gu, Ultrasound-assisted leaching of spent lithium ion batteries by natural organic acids and H2O2, *Chemosphere* (2020) 254, <https://doi.org/10.1016/j.chemosphere.2020.126670>.
- [213] Z. Takacova, T. Havlik, F. Kukurugya, D. Orac, Cobalt and lithium recovery from active mass of spent Li-ion batteries: theoretical and experimental approach, *Hydrometallurgy* 163 (2016) 9–17, <https://doi.org/10.1016/j.hydromet.2016.03.007>.
- [214] H. Wang, B. Friedrich, Development of a highly efficient hydrometallurgical recycling process for automotive Li-ion batteries, *J. Sustain. Metal.* 1 (2015) 168–178, <https://doi.org/10.1007/S40831-015-0016-6>.
- [215] Y. Yang, X. Meng, H. Cao, X. Lin, C. Liu, Y. Sun, Y. Zhang, Z. Sun, Selective recovery of lithium from spent lithium iron phosphate batteries: a sustainable process, *Green Chem.* 20 (2018) 3121–3133, <https://doi.org/10.1039/C7GC03376A>.
- [216] G. Zante, A. Braun, A. Masmoudi, R. Barillon, D. Trébouet, M. Boltoeva, Solvent extraction fractionation of manganese, cobalt, nickel and lithium using ionic liquids and deep eutectic solvents, *Miner. Eng.* 156 (2020), <https://doi.org/10.1016/j.mineng.2020.106512>.
- [217] C. Lupi, M. Pasquali, A. Dell'Era, Nickel and cobalt recycling from lithium-ion batteries by electrochemical processes, in: Waste Management, Elsevier Ltd, 2005, pp. 215–220, <https://doi.org/10.1016/j.wasman.2004.12.012>.
- [218] M.B.J.G. Freitas, E.M. Garcia, Electrochemical recycling of cobalt from cathodes of spent lithium-ion batteries, *J. Power Sources* 171 (2007) 953–959, <https://doi.org/10.1016/j.jpowsour.2007.07.002>.
- [219] Q. Meng, Y. Zhang, P. Dong, Use of electrochemical cathode-reduction method for leaching of cobalt from spent lithium-ion batteries, *J. Clean. Prod.* 180 (2018) 64–70, <https://doi.org/10.1016/j.jclepro.2018.01.101>.
- [220] X. Meng, J. Hao, H. Cao, X. Lin, P. Ning, X. Zheng, J. Chang, X. Zhang, B. Wang, Z. Sun, Recycling of LiNi<sub>1/3</sub>Co<sub>1/3</sub>Mn<sub>1/3</sub>O<sub>2</sub> cathode materials from spent lithium-ion batteries using mechanochemical activation and solid-state sintering, *Waste Manag.* 48 (2019) 54–63, <https://doi.org/10.1016/J.WASMAN.2018.11.034>.
- [221] H. Nie, L. Xu, D. Song, J. Song, X. Shi, X. Wang, L. Zhang, Z. Yuan, LiCoO<sub>2</sub>: recycling from spent batteries and regeneration with solid state synthesis, *Green Chem.* 17 (2015) 1276–1280, <https://doi.org/10.1039/C4GC01951B>.
- [222] Q. Jing, J. Zhang, Y. Liu, W. Zhang, Y. Chen, C. Wang, Direct regeneration of spent LiFePO<sub>4</sub>Cathode material by a green and efficient one-step hydrothermal method, *ACS Sustain. Chem. Eng.* 8 (2020) 17622–17628, [https://doi.org/10.1021/ACSSUSCHEMENG.0C07166/ASSET/IMAGES/LARGE/SC0C07166\\_0004.JPG](https://doi.org/10.1021/ACSSUSCHEMENG.0C07166/ASSET/IMAGES/LARGE/SC0C07166_0004.JPG).
- [223] Y. Shi, G. Chen, Z. Chen, Effective regeneration of LiCoO<sub>2</sub> from spent lithium-ion batteries: a direct approach towards high-performance active particles, *Green Chem.* 20 (2018) 851–862, <https://doi.org/10.1039/C7GC02831H>.
- [224] H. Gao, Q. Yan, P. Xu, H. Liu, M. Li, P. Liu, J. Luo, Z. Chen, Efficient direct recycling of degraded LiMn<sub>2</sub>O<sub>4</sub>Cathodes by one-step hydrothermal relithiation, *ACS Appl. Mater. Interfaces* 12 (2020) 51546–51554, [https://doi.org/10.1021/ACSA.MI.0C15704/ASSET/IMAGES/LARGE/AM0C15704\\_0007.JPG](https://doi.org/10.1021/ACSA.MI.0C15704/ASSET/IMAGES/LARGE/AM0C15704_0007.JPG).
- [225] M. Fan, X. Chang, Q. Meng, L. Wan, Y. Guo, Progress in the sustainable recycling of spent lithium-ion batteries, *SusMat* 1 (2021) 241–254, <https://doi.org/10.1002/sus.216>.
- [226] X. Chen, C. Yang, Y. Yang, H. Ji, G. Yang, Co-precipitation preparation of Ni-Co-Mn ternary cathode materials by using the sources extracting directly from spent lithium-ion batteries, *J. Alloys Compd.* 909 (2022), 164691, <https://doi.org/10.1016/J.JALLCOM.2022.164691>.
- [227] L.P. He, S.Y. Sun, J.G. Yu, Performance of LiNi<sub>1/3</sub>Co<sub>1/3</sub>Mn<sub>1/3</sub>O<sub>2</sub> prepared from spent lithium-ion batteries by a carbonate co-precipitation method, *Ceram. Int.* 44 (2018) 351–357, <https://doi.org/10.1016/J.CERAMINT.2017.09.180>.
- [228] J. Yang, W. Wang, H. Yang, D. Wang, One-pot compositional and structural regeneration of degraded LiCoO<sub>2</sub> for directly reusing it as a high-performance lithium-ion battery cathode, *Green Chem.* 22 (2020) 6489–6496, <https://doi.org/10.1039/D0GC02662J>.
- [229] A. Garg, L. Zhou, J. Zheng, L. Gao, Qualitative framework based on intelligent robotics for safe and efficient disassembly of battery modules for recycling purposes, *IOP Conf. Ser. Earth Environ. Sci.* 463 (2020), 12159, <https://doi.org/10.1088/1755-1315/463/1/012159>.
- [230] O. Sung Kwon, I. Sohn, Fundamental thermokinetic study of a sustainable lithium-ion battery pyrometallurgical recycling process, *Resour. Conserv. Recycl.* (2020) 158, <https://doi.org/10.1016/J.RESCONREC.2020.104809>.
- [231] Ascend Elements, Sustainable lithium-ion battery materials and recycling — elevated (n.d.), <https://ascendelements.com/>.
- [232] Y. Wang, D. Apelian, H. Zou, Method and Apparatus for Recycling Lithium-Ion Batteries. US 2013/0302226 A1, 2013.
- [233] H. Bi, H. Zhu, L. Zu, Y. Bai, S. Gao, Y. Gao, A new model of trajectory in eddy current separation for recovering spent lithium iron phosphate batteries, *Waste Manag.* 100 (2019) 1–9, <https://doi.org/10.1016/j.wasman.2019.08.041>.
- [234] S. Virolainen, T. Wesselsborg, A. Kaukinen, T. Sainio, Removal of iron, aluminium, manganese and copper from leach solutions of lithium-ion battery waste using ion exchange, *Hydrometallurgy* 202 (2021), 105602, <https://doi.org/10.1016/j.hydromet.2021.105602>.
- [235] H. Ku, Y. Jung, M. Jo, S. Park, S. Kim, D. Yang, K. Rhee, E.M. An, J. Sohn, K. Kwon, Recycling of spent lithium-ion battery cathode materials by ammoniacal leaching, *J. Hazard Mater.* 313 (2016) 138–146, <https://doi.org/10.1016/j.jhazmat.2016.03.062>.
- [236] X. Liu, K. Huang, H. Xiong, H. Dong, Ammoniacal leaching process for the selective recovery of value metals from waste lithium-ion batteries, *Environ. Technol.* (2021) 1–15, <https://doi.org/10.1080/09593330.2021.1968505>.
- [237] L. Zhuang, C. Sun, T. Zhou, H. Li, A. Dai, Recovery of valuable metals from LiNi<sub>0.5</sub>Co<sub>0.2</sub>Mn<sub>0.3</sub>O<sub>2</sub> cathode materials of spent Li-ion batteries using mild mixed acid as leachant, *Waste Manag.* 85 (2019) 175–185, <https://doi.org/10.1016/j.wasman.2018.12.034>.
- [238] X. Chen, H. Ma, C. Luo, T. Zhou, Recovery of valuable metals from waste cathode materials of spent lithium-ion batteries using mild phosphoric acid, *J. Hazard Mater.* 326 (2017) 77–86, <https://doi.org/10.1016/j.jhazmat.2016.12.021>.
- [239] D. Gao, Y. Zhou, T. Wang, Y. Wang, A method for predicting the remaining useful life of lithium-ion batteries based on particle filter using kendall rank correlation coefficient, *Energies* 13 (2020), <https://doi.org/10.3390/EN13164183>.
- [240] O.A. Nasser, M. Petranikova, Review of achieved purities after li-ion batteries hydrometallurgical treatment and impurities effects on the cathode performance, *Batteries* 7 (2021), <https://doi.org/10.3390/batteries0703006>.
- [241] F. Duarte Castro, M. Vaccari, L. Cutaia, Valorization of resources from end-of-life lithium-ion batteries: a review, *Crit. Rev. Environ. Sci. Technol.* (2021), <https://doi.org/10.1080/10643389.2021.1874854>.
- [242] F. Gu, J. Guo, X. Yao, P.A. Summers, S.D. Widjatmoko, P. Hall, An investigation of the current status of recycling spent lithium-ion batteries from consumer electronics in China, *J. Clean. Prod.* 161 (2017) 765–780, <https://doi.org/10.1016/j.jclepro.2017.05.181>.
- [243] L. Wuschke, H.G. Jäckel, T. Leißner, U.A. Peuker, Crushing of large Li-ion battery cells, *Waste Manag.* 85 (2019) 317–326, <https://doi.org/10.1016/j.wasman.2018.12.042>.
- [244] Jaco Huisman, Silvia. Bobba, European commission. Joint research Centre., “Available for Collection” study on alternative collection targets for portable and light means of transport batteries., (n.d.).
- [245] C. Yang, J. Zhang, G. Liang, H. Jin, Y. Chen, C. Wang, An advanced strategy of “metallurgy before sorting” for recycling spent entire ternary lithium-ion

- batteries, *J. Clean. Prod.* 361 (2022), 132268, <https://doi.org/10.1016/J.JCLEPRO.2022.132268>.
- [246] J. Quan, S. Zhao, D. Song, T. Wang, W. He, G. Li, Comparative life cycle assessment of LFP and NCM batteries including the secondary use and different recycling technologies, *Sci. Total Environ.* (2022) 819, <https://doi.org/10.1016/j.scitotenv.2022.153105>.
- [247] A. Amato, A. Becci, M. Villen-Guzman, C. Vereda-Alonso, F. Beolchini, Challenges for sustainable lithium supply: a critical review, *J. Clean. Prod.* 300 (2021), 126954, <https://doi.org/10.1016/j.jclepro.2021.126954>.
- [248] H. Dang, B. Wang, Z. Chang, X. Wu, J. Feng, H. Zhou, W. Li, C. Sun, Recycled lithium from simulated pyrometallurgical slag by chlorination roasting, *ACS Sustain. Chem. Eng.* 6 (2018) 13160–13167, <https://doi.org/10.1021/ACSSUSCHEMENG.8B02713>.
- [249] D. Thompson, C. Hyde, J.M. Hartley, A.P. Abbott, P.A. Anderson, G.D.J. Harper, To shred or not to shred: a comparative techno-economic assessment of lithium ion battery hydrometallurgical recycling retaining value and improving circularity in LIB supply chains, *Resour. Conserv. Recycl.* 175 (2021), 105741, <https://doi.org/10.1016/J.RESCONREC.2021.105741>.