



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.

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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	GHG	Greenhouse Gas
CAM	Cathode active material	HEV	Hybrid Electric Vehicle
CHB	Cyclohexylbenzene	LCA	Life cycle assessment
CRM	Critical Raw Material	LCO	Lithium cobalt oxide LiCoO_2
DEC	Diethyl carbonate	LFP	Lithium iron phosphate LiFePO_4
DMC	Dimethyl carbonate	LIB	Lithium-ion battery
EC	Ethylene carbonate	LMO	Lithium manganese oxide LiMn_2O_4
EMC	Ethyl methyl carbonate	NCA	Lithium nickel cobalt aluminum oxide $\text{LiNi}_x\text{Co}_y\text{Al}_{1-x-y}\text{O}_2$
EOL	End-of-life	NiMH	Nickel metal hydride
EV	Electric vehicles	NMC	Lithium nickel manganese cobalt oxide $\text{LiNi}_x\text{Mn}_y\text{Co}_{1-x-y}\text{O}_2$
		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_2 [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.

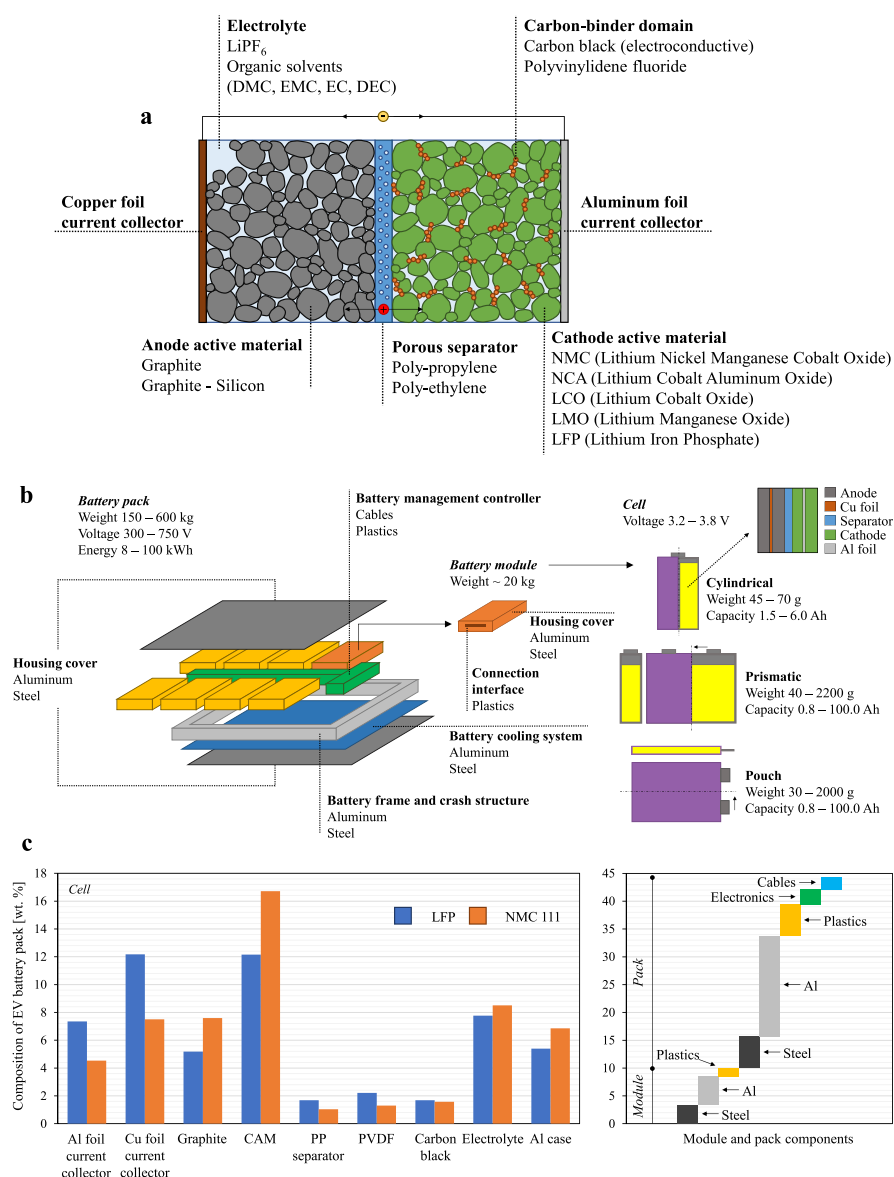
The paper is organized as follows. First, an overview of LIB designs and chemistries is given in Sec. 2 to illustrate the involved materials. Then, LIB recycling routes and processes are discussed in detail in Sec. 3.1, with a further focus on a selected representative pyrometallurgy-based process (Sec. 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_6 salt (ca. 1 mol L^{-1}) 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^{-1} for silicon vs. 372 mAh g^{-1} 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].



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_2 (LCO), lithium manganese oxide LiMn_2O_4 (LMO), lithium nickel manganese cobalt oxide $\text{LiNi}_x\text{Mn}_y\text{Co}_{1-x-y}\text{O}_2$ (NMC), lithium nickel cobalt aluminum oxide $\text{LiNi}_x\text{Co}_y\text{Al}_{1-x-y}\text{O}_2$ (NCA) and lithium iron phosphate LiFePO_4 (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 $\text{LiNi}_{0.33}\text{Co}_{0.33}\text{Mn}_{0.33}\text{O}_2$, named NMC111 or NMC333, and $\text{LiNi}_{0.5}\text{Co}_{0.3}\text{Mn}_{0.2}\text{O}_2$, NMC532) to high ($\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$, NMC622) and very high excess ($\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$, 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., $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$ is more widespread compared to $\text{LiNi}_{0.85}\text{Co}_{0.11}\text{Al}_{0.04}\text{O}_2$ or $\text{LiNi}_{0.84}\text{Co}_{0.12}\text{Al}_{0.04}\text{O}_2$), 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 peripherals 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 peripherals 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 $-65\text{ }^{\circ}\text{C}$ [73] or, as currently explored at laboratory and demonstration levels, electrolyte extraction with super- or sub-critical 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. $1500\text{ }^{\circ}\text{C}$) 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 H_2SO_4 , with H_2O_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

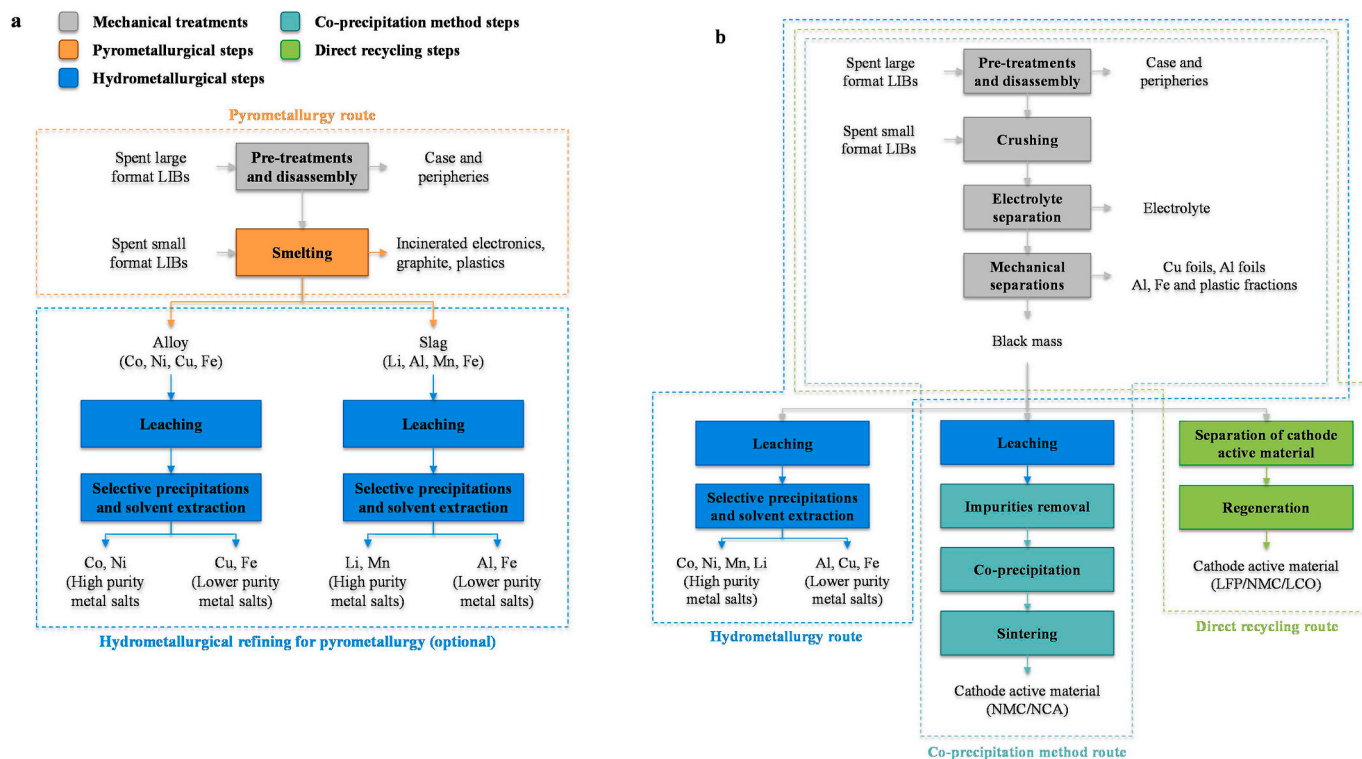


Fig. 2. Simplified block flow diagrams of general LIB recycling processes based on: (a) pyrometallurgical route, with optional hydrometallurgical refining of the alloy and/or the slag, (b) hydrometallurgical/co-precipitation/direct recycling routes, preceded by a mechanical separation section. The colourmap 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"> • Versatile: applicable to several battery chemistries and geometries • High recycling capacity • No sorting or other pre-treatments • High recovery of valuable metals (Co, Ni, Cu) • Established method and industrial know-how 	<ul style="list-style-type: none"> • Graphite, plastics, and electrolyte are necessarily burnt • Downcycling of Li and Al affecting the supply chain costs of the raw material • Expensive off-gas treatments to avoid toxic air emissions • High energy consumption • Low purity of products or further hydrometallurgical refinement
Hydrometallurgy [84, 112]	<ul style="list-style-type: none"> • High recovery rates, which can potentially include the entire electrolyte • High purity of products • Low energy consumption • Low GHG emissions 	<ul style="list-style-type: none"> • High GHG emissions • High sorting efficiencies and mechanical pre-treatments required • Crushing of batteries involves safety problems • Efficient electrolyte separation methods have to be industrially demonstrated • Lower recycling capacity • Contaminated wastewater • High operating costs (e.g., solvents) • Process complexity • High costs for the treatment of the final effluent (neutralization)

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 ⁻¹]	Battery chemistry treated	Industrial reality	Source
Europe					
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)	Li-ion	M	[128,129]
		3000 (2020)	Others		
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]
Batrec 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]
	Sweden	Unknown (2023) up to 125,000 (2030)	Li-ion	M H	
Fortum Oyj	Finland	3000 (2022)	Li-ion	M (H in 2023)	[137]
Stena	Sweden	10,000 (2023)	Li-ion	M H	[138]
North America					
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 ^a (2024)	Cathode scraps	CP (M)	
Lithion Recycling	Canada	200 (2020)	Li-ion	M H	[121,145]
	Canada	2000 (2023)	Li ion	M H	
Asia					
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]
Huayo 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)	Li-ion	M P	[122]
		1200 (2020)	NiCd, NiMH		
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]

^a American Manganese Inc recycling plant will recycle ca. 7700 t y⁻¹ of cathode production scraps from ItalVolt gigafactory, corresponding to 40,000 t y⁻¹ 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 Hofers, Sweden, in 2011 the 7000 t y⁻¹ 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).

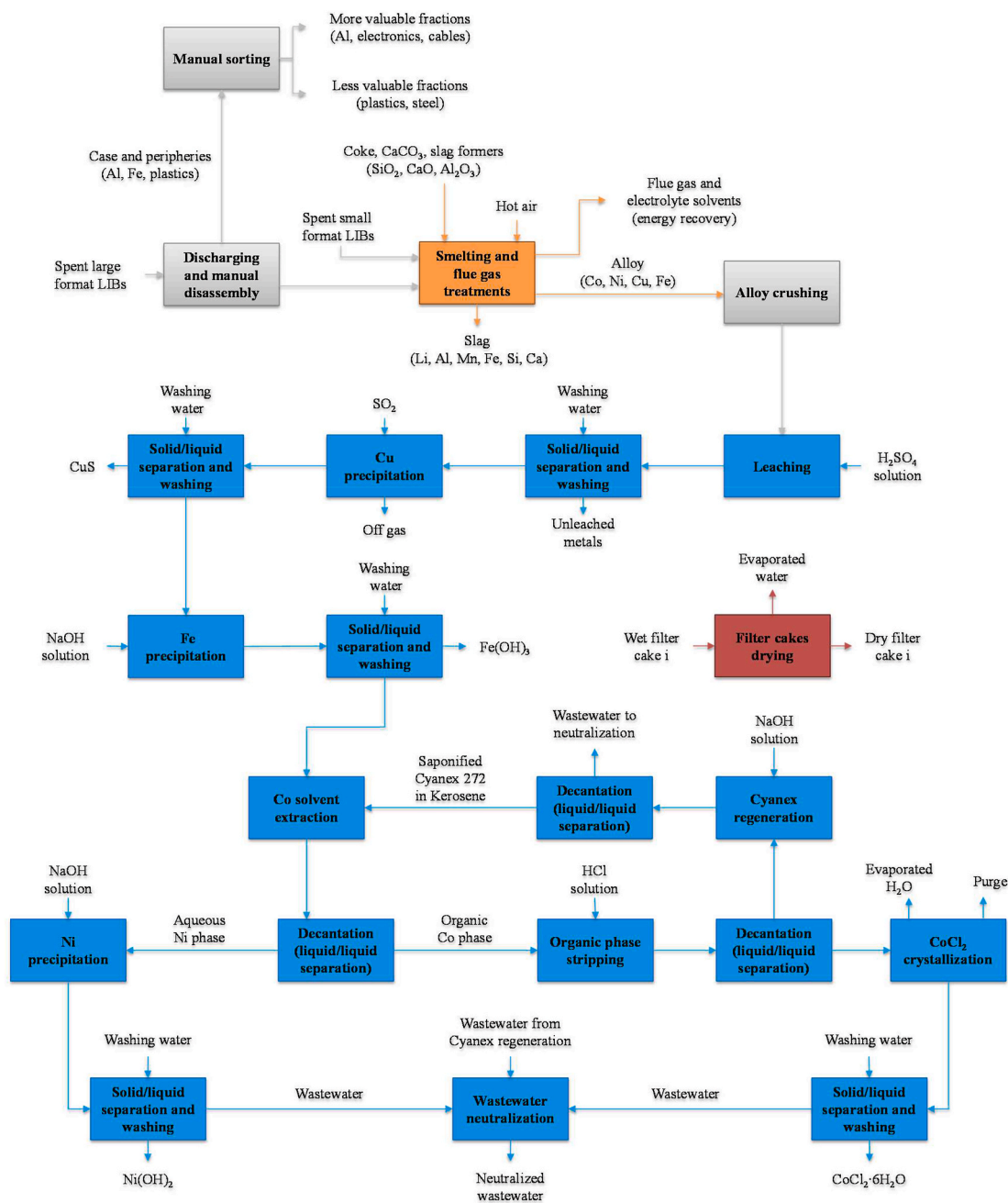


Fig. 3. Simplified flowchart of Umicore process [18,23,24], including pyrometallurgical smelting (orange), mechanical treatments (grey) and hydrometallurgical refining of the alloy (blue); the heat treatments, as the drying of filtered materials, are reported in red and apply to all the humid output 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 (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 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 LiPF_6 and the binder PVDF [153], and volatile organic compounds (VOCs) [154] are captured by injection of Ca, Na, or 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% CaO, 34 wt% SiO_2 , 11 wt% Al_2O_3 , 15 wt% others), 110 kg of SiO_2 , 100 kg of 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 SiO_2 , 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 H_2SO_4 solution, thus forming metal sulfate salts dissolved in aqueous solution [80]. Next, Cu is removed from the solution by precipitation of CuS and Cu_2S using SO_2 at elevated temperatures [54], then Fe is precipitated as hydroxide using 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 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 NaOH is added to the Ni-rich phase to precipitate $\text{Ni}(\text{OH})_2$ at pH ca. 9, leading also to the precipitation of $\text{Co}(\text{OH})_2$ 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 CoCl_2 solution. Such a solution is sent to a crystallization section to obtain solid CoCl_2 , setting the operative condition to avoid the crystallization of NiCl_2 and CuCl_2 , which are more soluble than CoCl_2 [18,39,163]. The organic extractant is regenerated with 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 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 Li_2CO_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 Li_2CO_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

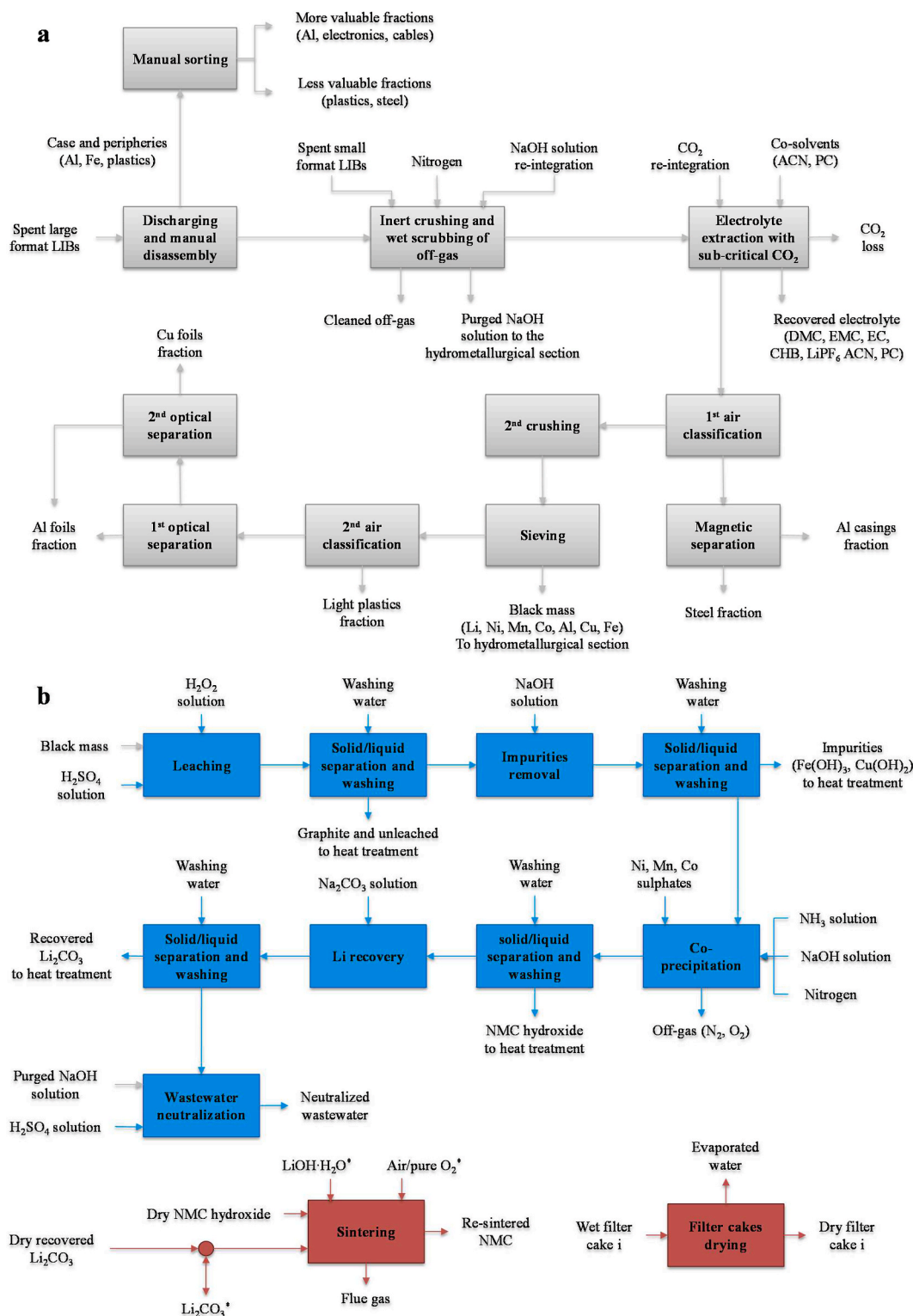


Fig. 4. Simplified flowchart of LithoRec process [18,24,35,54,91,122,164–170], including mechanical treatments (grey), hydrometallurgical steps (blue) and heat treatments (red). (a) First section of the process, comprising all the mechanical treatments. (b) Second part of the process, related to the 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 CO₂, 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 CO₂ 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 CO₂ 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 LiPF₆) 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 CO₂ 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 Li₂CO₃. The applicability of super-critical CO₂ 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 LiPF₆ 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⁻¹ air and an air velocity of 3.34 m s⁻¹ [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 (2nd 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⁻¹ air and an air velocity of 1.1 m s⁻¹ [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 H₂SO₄ 4 mol L⁻¹ and 30–50 wt% H₂O₂ 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 MnSO₄, NiSO₄, and CoSO₄ 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_xMn_yCo_{1-x-y}(OH)₂) is co-precipitated by increasing the pH to ca. 11 by using NaOH in presence of NH₄OH as a chelating agent, which avoids the precipitation of metals as single-phase hydroxides. The operation is realized in a stirred tank under N₂ atmosphere to avoid the oxidation of Mn²⁺ to Mn⁴⁺. 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 Na₂CO₃ solution at 40 °C to precipitate Li₂CO₃. The NMC hydroxide precursor and the Li₂CO₃ are dried (see the red box at the bottom right of Fig. 4b) and mixed with additional virgin Li₂CO₃ 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 (Li₂CO₃ 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_xMn_yCo_{1-x-y}(OH)₂ 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 ₂	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 ₂	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]
	Froth floatation to separate active materials	Prototype	[143,208–210]
Pyrometallurgy	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]
	Off-gas treatments	Industrial	[17–19,79,82,154]
Hydrometallurgy	Leaching of the black mass with H ₂ SO ₄ 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 ₂ CO ₃ precipitation with Na ₂ CO ₃ or CO ₂	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]
	Electrochemical production of Co, Cu and MnO	Prototype	[195,217–219]
Direct recycling	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 $-160\text{ }^{\circ}\text{C}$ 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. $-60\text{ }^{\circ}\text{C}$ 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], Retrie uses a recirculating LiOH brine [139], Li-Cycle adopts a solution of Ca(OH)₂ 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 CO₂ [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 CO₂ (25 °C, 60 bar) and additional polar organic co-solvents operated on battery fragments [179], whereas OnTo patented a process based on super-critical CO₂ (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 CO₂ 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 N₂ [179], Batrec uses CO₂ [193], while Recupyl utilizes both CO₂ and Ar [130]. The CO₂ atmosphere avoids any trace of metal Li from strongly react with water by converting it into the less reactive Li₂CO₃. 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⁻³ [18], Li-Cycle proposes to separate current collectors from plastics through a liquid with density 2500 kg m⁻³ and Cu from Al with another liquid with density 2850 kg m⁻³ [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 MnO₂ [145]), Li-Cycle, and Accurec, or with SO₂, 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 Na₂S 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 NH₄OH 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 Li₂CO₃ through the addition of Na₂CO₃ solution [99,215] (e.g., Ascend Elements, Accurec, Retrie, Li-Cycle, American Manganese

Inc, Lithion), or through CO₂ bubbling (Recupyl) [130], or again by precipitating Li₃PO₄ after neutralization with H₃PO₄ (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_{0.5}Co_{0.2}Mn_{0.3}O₂ from spent LIBs using LiOH-H₂O 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-Li₂CO₃ to compensate the Li⁺ loss in the degraded carbonate and repair the damaged structure via a “dissolution-recrystallization” mechanism, while the electrochemical reversibility of FePO₄ 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 CO₂ extraction introduce impurities coming from the thermal decomposition of LiPF₆

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

¹ slag addressed to the construction industry² the respective material is included in the alloy as an impurity³ black mass recovered in this process also includes Al and Cu current collector foils and plastics in a not negligible amount⁴ 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 (Brunp 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

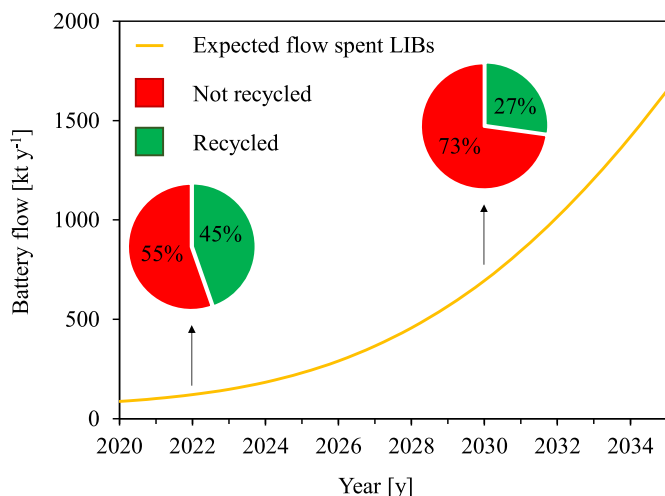


Fig. 5. Comparison of EU total outlooked flow of spent LIBs from 2020 to 2035 (solid yellow line). The insets report the fraction of EOL LIBs which are expected to be recycled (green) or not (red) in 2022 and 2030 considering a conservative situation in which recycling companies keep constant their capacity 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⁻¹ 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⁻¹) and Co (ca. 50 \$ kg⁻¹) [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 manufactures

(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.

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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

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SUPPLEMENTARY INFORMATION

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

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Table S1. Characteristics of selected cathode active materials ¹⁻⁶.

Cathode material	Voltage operating range	Nominal voltage	Specific accessible capacity	Specific accessible energy	Cycle life	Cost
	[V]	[V]	[mAh g ⁻¹]	[Wh kg ⁻¹]	[No. cycles]	[€ kg ⁻¹]
LCO	3.00 - 4.20	3.60	145	150 - 200	500 - 1000	29.75
LMO	3.00 - 4.20	3.70 - 3.80	120	100 - 150	300 - 700	8.50
NMC-111	3.00 - 4.20	3.60 - 3.70	160 - 170	150 - 220	1000 - 2000	17.00
NMC-532	3.00 - 4.20	3.60 - 3.70	180	150 - 220	1000 - 2000	16.00
NMC-622	3.00 - 4.20	3.60 - 3.70	180 - 190	150 - 220	1000 - 2000	14.45
NMC-811	3.00 - 4.20	3.60 - 3.70	200	200 - 260	500 - 1000	13.60
NCA	3.00 - 4.20	3.60	180 - 200	200 - 260	500	20.40
LFP	2.50 - 3.65	3.20 - 3.30	150 - 170	90 -120	2000	11.90

Table S2. Overview of some of the most widespread BEV models and their LIBs currently on market ¹.

Cell Chemistry	Battery Manufacturer	Battery Format	Cell Capacity	Cell Voltage	Battery Pack Energy	Battery Pack Range	BEV model
Cathode/Anode			[Ah]	[V]	[kWh]	[km]	
LFP/C	A123	Pouch	20	3.3	21	130	Chevrolet spark EV (2012)
LFP/C	BYD	Prismatic	-	3.3	61	300	BYD e6 (2010)
NCA/C	Panasonic	Cylindrical	3.2	3.6	60-100	330-500	Tesla S (2012)
NCA/Si-C	Panasonic	Cylindrical	3.4	3.6	50-100	330-500	Tesla X (2015)
NCA/Si-C	Panasonic	Cylindrical	4.75	3.6	75-100	350-500	Tesla 3 (2017)
NMC/C	Panasonic/Sanyo	Prismatic	25	3.7	24	190	VW eGolf (2015)
NMC/LTO	Toshiba	Prismatic	20	2.3	20	130	Honda Fit EV (2013)

NMC/C	Li-Tec	Pouch	52	3.65	17	145	Smart Fortwo EV (2013)
NMC/C	SK Innovation	Pouch	38	3.7	27	145	Jia Soul EV (2014)
NMC/C	LG Chem	Pouch	56	3.65	60	383	Chevrolet Bolt (2016)
NMC/C	LG Chem	Pouch	59	3.7	41	400	Renault Zoe (2017)
NMC/C	Samsung SDI	Prismatic	94	3.68	33.2	180	BMW i3 (2016)
NMC-LMO/C	Li energy Japan	Prismatic	50	3.7	16	160	Mitsubishi i-MiEV (2008)
NMC-LMO/C	Samsung SDI	Prismatic	63	3.65	24	140	Fiat 5003 (2013)
NMC-LMO/C	LG Chem	Pouch	16	3.7	35.5	160	Ford Focus EV (2012)
NMC-LMO/C	LG Chem	Pouch	36	3.75	26	150	Renault Zoe (2012)
LMO-NCA/C	AESC	Pouch	33	3.75	24	135	Nissan Leaf (2010)
LMO-NCA/C	AESC	Pouch	40	3.75	30	172	Nissan Leaf (2015)

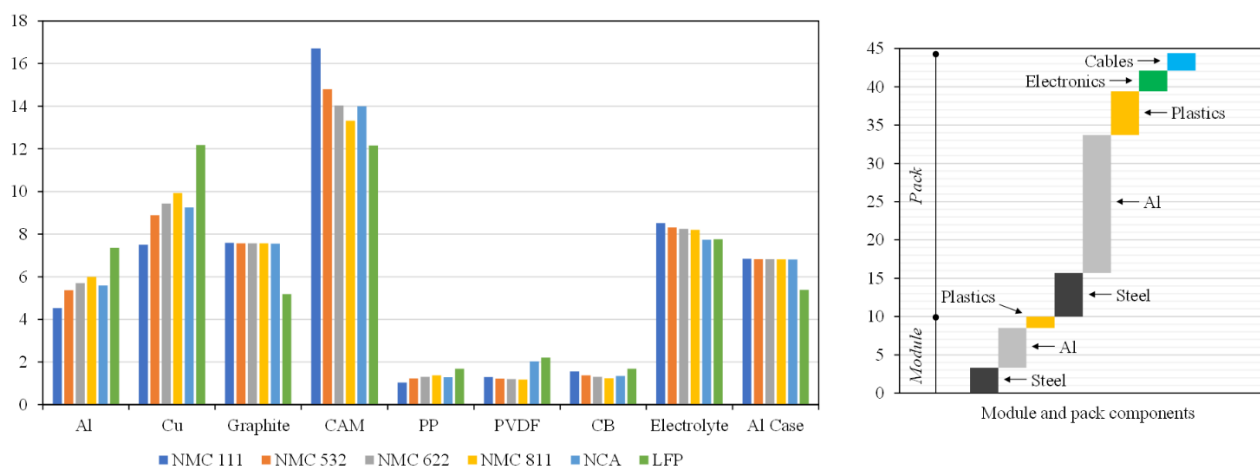


Figure S1. Mass composition of a battery system for EV according to its cathode chemistry.

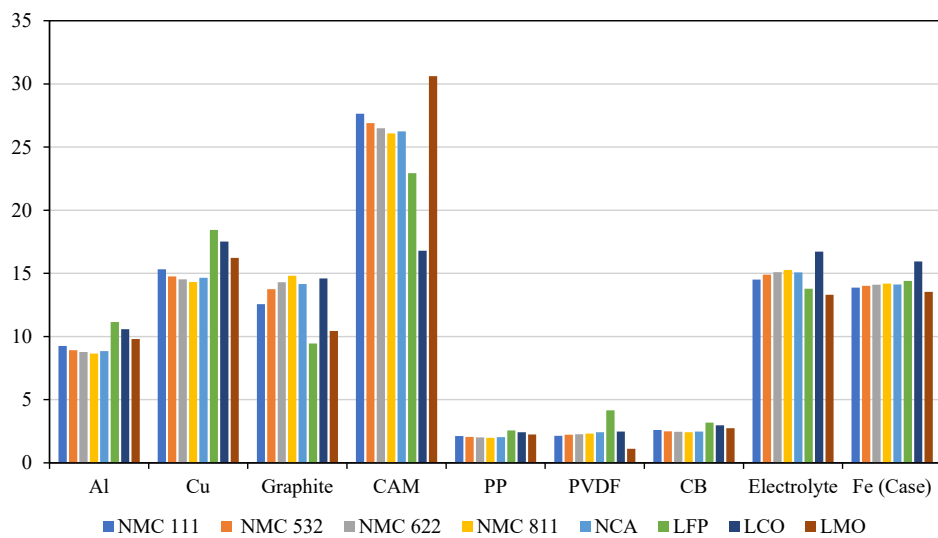


Figure S2. Mass composition of a small format lithium-ion battery according to its cathode chemistry.

S1. Description of other recycling processes in Europe

S1.1 Accurec

Accurec Recycling GmbH is a German battery recycling company founded in 1995⁷⁻¹⁵. In 2016 Accurec opened a new recycling facility in Krefeld, Germany, specific for Li-ion batteries and in 2019 it reached a capacity of 3,000 t y⁻¹. The Accurec process was initially designed only for Ni-metal hydride batteries but it is currently able to recycle also LIBs using a combination of mechanical, pyrometallurgical and hydrometallurgical unit operations in order to recover a Co-Ni-Mn alloy and potentially Li₂CO₃.

LIBs are sorted, cleaned, and manually disassembled. The dismantled battery cells are fed to a vacuum thermal treatment in a rotary kiln at 250-600°C to remove electrolyte, solvents, and volatile hydrocarbons. The temperature is limited to 600°C to avoid the oxidation of valuable metals. In particular, the vacuum thermal pyrolysis is carried out preferably at 500°C and 100 mbar for 2-3 hours. The process eliminates the risk related to flammable organic solvents and toxic decomposition of lithium conductive salt. The off-gas is cleaned by afterburner and the excess energy produced by the incineration of solvents and organic compounds is used to produce high pressure steam, which is used in the process. The flue gases are cooled down with water, then passed through a flue gas scrubber to wash out halogen gaseous compounds, after that dust is removed in a condensation electro-filter and NO_x are removed by a catalyst. Electrolyte solvent could be partially recovered through condensation from the off-gas achieving a recycling efficiency of the electrolyte equal to approximately the 80 wt% with a composition of the recovered solvents mixture equal to 71 wt% of EMC, 10 wt% of EC and 19 wt% others. However, this mixture contains also various decomposition products that make a direct reuse impossible and so this alternative is not yet economically attractive. Treated batteries are milled and grinded using a rotating roll crusher, then fragments undergo a series of mechanical operations including vibrating screen, magnetic separator and zig-zag air classifiers. The mechanical separations produce a heavy fraction containing mostly steel and nickel heavy

magnetic fraction, a plastic and aluminum heavy non-magnetic fraction, an Al and Cu current collector foils light fraction, a plastic separator light fraction and a sieved powder which is the black mass. The black mass, which has a particle size lower than 200 μm , is agglomerated in pellets and undergoes a two-step pyrometallurgical process. The pellets are composed approximately of 30 wt% Co, 3 wt% Li, 30 wt% graphite and the remaining fraction is made of slag formers and binder. The slag formers are Al_2O_3 , CaO, MgO and SiO_2 . The first step at 800°C is in a rotary kiln in order to halve the graphite content and a second step is in an electric arc furnace operating the “reducing meltdown” at 1500-1800°C. In the reducing meltdown graphite is burnt to enhance the recovery of valuable metals like Co and Ni. The slag phase includes Li_2O , CaO, SiO_2 , Al, the remaining graphite, and impurities mainly of Cu, Co and Fe. The alloy is composed of Co, Ni, Fe, and Mn and is sold to metal industry. In the reducing meltdown process most of the lithium is volatilized as flue dust and oxidized as Li_2O .

In the past, efforts were made to further process flue dust and slag with hydrometallurgical treatments to recover Li_2CO_3 ; however, such a recovery is not operated anymore⁹. During such research studies, the slag was crushed to obtain a particle size lower than 100 μm , then it was mixed with the flue dust and leached in 1 mol L⁻¹ H_2SO_4 for 2 hours at 60-80°C with possible addition of H_2O_2 . The solution was refined to remove impurities mainly of Cu, Fe, Al. First copper ions were cemented adding steel shots achieving a precipitation of more than 95 wt% of Cu in 30 minutes. Then Al and Fe ions were precipitated at 40°C by adjusting the pH to 4.4 with NaOH solution to precipitate hydroxides with a removal efficiency of Al and Fe higher than 97 wt%. Finally, the refined solution was boiled at 90°C to concentrate Li in the solution, 200 g L⁻¹ Na_2CO_3 solution were added and Li_2CO_3 was precipitated keeping the temperature at 95°C, achieving a claimed Li_2CO_3 recovery efficiency of 90 wt% with battery grade quality and a declared overall recycling efficiency of 65.2 wt%. However, Li recovery does not appear to be currently operated anymore by the company.

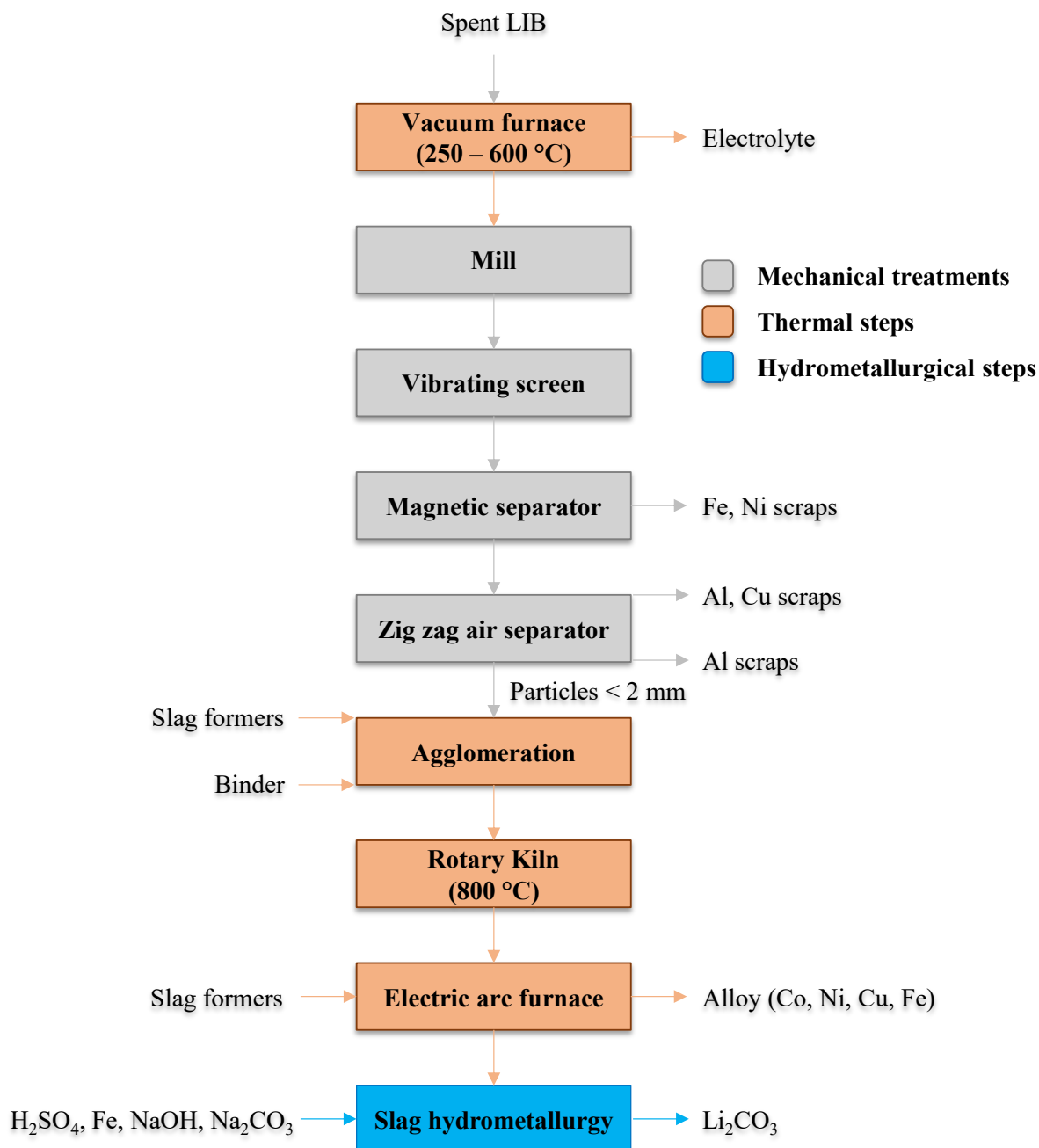


Figure S3. Simplified flowchart of Accurec process¹³; the Li_2CO_3 recovery is no longer operated⁹.

S1.2 Erasteel Recycling (former Valdi)¹⁶

Valdi was a battery recycling French company, which was acquired by the French mining and metallurgical group Eramet in 2009. In 2021 Valdi trademark was replaced by Erasteel Recycling as the Eramet group became Erasteel in 1992. The battery recycling process is focused on the recycling of a mixture of alkaline, saline and Ni-metal hydride batteries, with only small amount of Li-ion

optionally treated. Batteries are fed in a continuous arc furnace that produces a metal alloy of Ni-Fe and a manganese slag. The alloy is sold to stainless steel market and it contains 20-25 wt% of Ni, whereas the Mn slag composed by MnO for up to the 47 wt% is addressed to producers of silico-manganese alloy. The flue gas of the furnace is specifically cleaned and the flue dust is recovered and dedicated to a process unit for refinement and production of ZnO fraction with a purity up to 82 wt%. The recycling efficiency claimed by Erasteel is around the 80 wt%.

SI.3 SNAM

SNAM (Societe Nouvelle d’Affinage des Metaux) is a battery recycling company founded in 1977 in Lyon, France⁹. It recycles Ni-Cd, Ni-metal hydride, alkaline and Li-ion batteries. The recycling activities for Li-ion batteries started in 2000. Li-ion batteries are treated as Ni-Cd ones with a process including pyrolysis and refinements. The pyrolysis is a pre-treatment step, in which flammable solvents and other organic and plastic compounds are decomposed and incinerated in a following afterburner. The remaining battery scraps are sent to primary metallurgy processors to recover Co, Ni and Cu. The refinement treatments are mainly pyrometallurgy-based, but a hydrometallurgical section is under development.

SI.4 Euro Dieuze Industrie

Euro Dieuze Industrie (E.D.I.) is a French company located in the Metz-Nancy area^{9,17}. It is a subsidiary of SARP Industries and of the Veolia Environment Waste Management Division. E.D.I. treats 5,000 t y⁻¹ of spent batteries including alkaline, Ni-Cd, primary and secondary lithium batteries. The process of lithium-ion batteries recycling is mainly mechanical, including manual sorting, disassembly, crushing and other separations. Crushing for Li-ion batteries is carried out with a specially designed process using water flow to catch the electrolyte solvents. Crushed fragments are sieved to separate the black mass. The oversized fractions are fed to an electromagnetic conveyor belt

to separate steel and then, using an eddy current disk, paramagnetic metals such Al and Cu are recovered. Black mass is not currently refined.

SI.5 Akkuser Oy

Akkuser Ltd is a battery recycling company founded in 2006 in Nivala, Finland ^{7,11,12,18,19}. It handles ca. 4000 t y⁻¹ of mixed LIBs with several chemistries using its own method named Dry-Technology, which is based on low temperature mechanical treatments and pre-processing. According to Akkuser data, the recycling rate is higher than 90 wt% and energy consumption is only 0.3 kWh kg⁻¹ ¹⁹.

The mixed spent LIBs are pre-sorted by type and then crushed by two continuous cutting mills. The first one operates at 40-50°C with a rotor speed of 100-400 rpm, resulting in battery fragments size ranging from 1.25 to 2.5 cm. During crushing gases such as H₂ and O₂ are released, thus the chambers are equipped with a cyclonic air removal. The off-gas is filtered a first time to remove plastic fragments with attached metals, then it is filtered further to produce a flue gas free of particulate, that is released to the atmosphere. An air-tight cooling tube transfers the crushed fragments to the second crusher, then operates at 1000-1200 rpm achieving a fragment size lower than 6 mm. Also, this equipment is provided with a cyclonic air removal and the same off-gas cleaning system previously described. The crushed batteries are fed to a magnetic separation step, resulting in the recovery of iron. The remained powder is composed mostly by electrodic materials and current collector foils and is sold as product to other recycling processes.

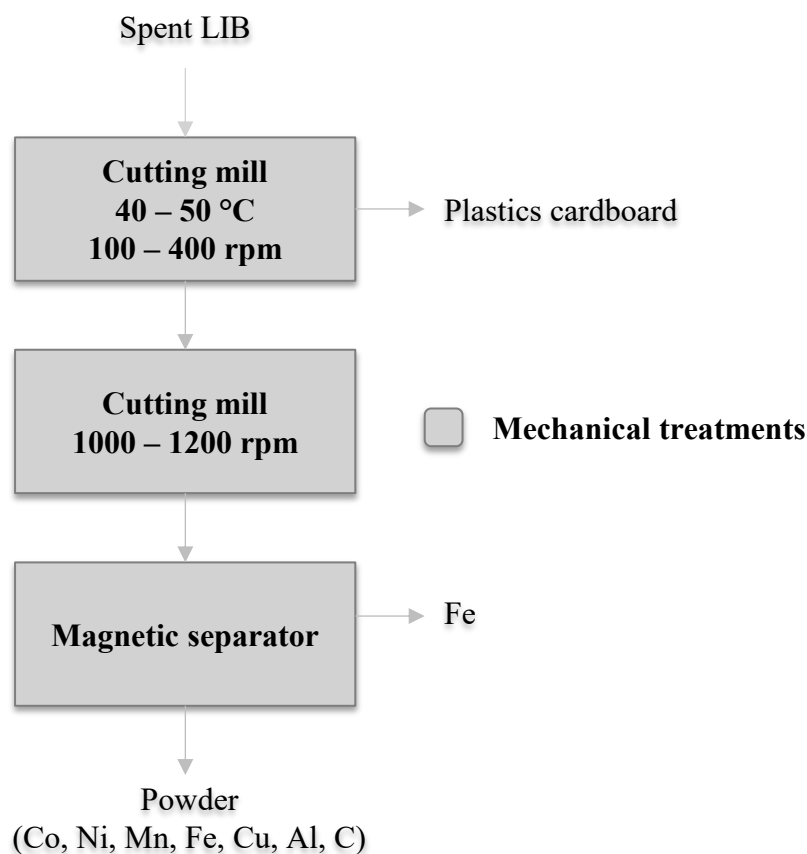


Figure S4. Simplified flowchart of Akkuser process ¹¹.

SI.6 Redux

Redux Recycling GmbH is a battery recycling company founded in 1997 in Dietzenbach, Germany ⁹. In 2016 it was acquired by the Austrian Saubermacher AG, which is a waste management and recycling specialist. In 2016 Redux opened its new hi-tech facility at Offenbach on the Main (Germany), which is able to recycle nearly 80% of all major portable batteries including alkaline, Ni-Cd and others. In 2018 Redux opened a new hi-tech plant for LIBs in Bremerhaven, which can recycle 10,000 t y⁻¹ of all types of Li-ion batteries, achieving a recycling efficiency of ca. 60-70%. The process for recycling Li-ion batteries is mainly mechanical.

The process starts with a sorting step, followed by a discharging step achieved with specific electronic devices that allow the recovery of the remaining energy stored in the batteries. The batteries are then dismantled to separate steel and plastic casings of the modules. A thermal pyrolysis treatment

is carried out in a rotary kiln to deactivate batteries by removing flammable electrolyte. A crushing step follows, along with undisclosed mechanical separations leading to the production of aluminum, copper and active materials fractions.

SI.7 BatRec

The BatRec Industries AG is a company based in Wimmis, Switzerland, specialized in hazardous waste treatment/recovery since 1989 ^{7,11,12,20}. The spent LIBs recycling process is mainly based on mechanical treatments.

Batteries are fed to a crusher, where they are comminuted under CO₂ atmosphere, which reduces flammable risks and reacts with elementary metallic lithium creating a passivation layer of Li₂CO₃. Crushed fragments are neutralized by feeding a flow of moist air to the crusher. Then the protective atmosphere is released and the electrolyte evaporated during the crushing is condensed in the off-gas treatment section, but it is not considered for further use due to the presence of decomposition impurities. After that undisclosed mechanical separation and leaching are carried out to obtain four different fractions: a magnetic metal fraction including mostly steel, a non-magnetic metal fraction composed by Al and Cu, a plastics fraction, and a fine fraction containing active materials. The black mass is sold to companies producing cobalt and nickel such as Glencore and Umicore.

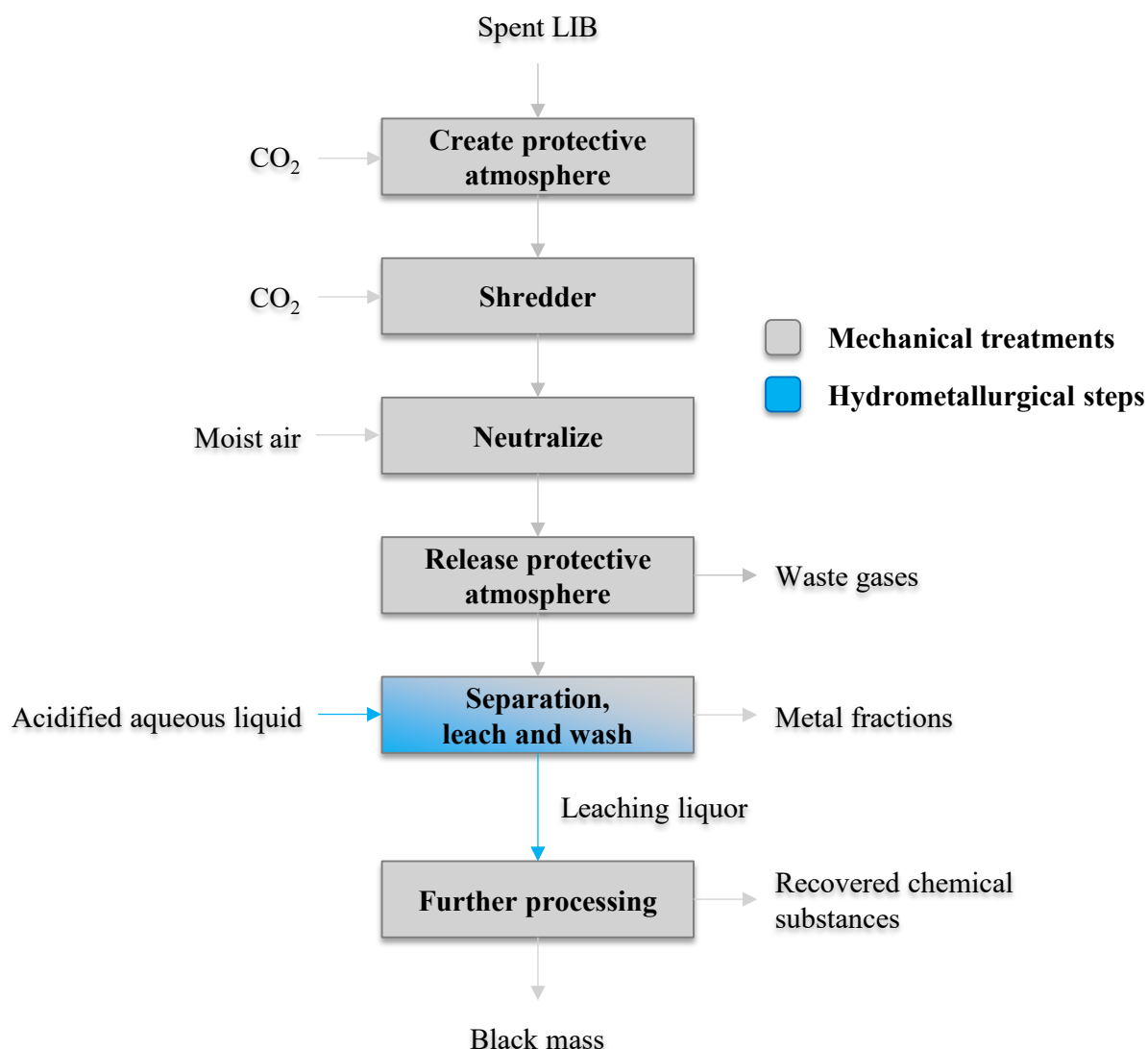


Figure S5. Simplified flowchart of BatRec process ¹².

S1.8 Recupyl Valibat

Recupyl is a French company founded in 1993 with focus on battery recycling ^{7-12,21-24}. The current process was developed during the VALIBAT project within the “European Commission’s Fifth Framework Programme” (FP5) between 2000 and 2003. The Recupyl process is a low temperature LIBs recycling technology based on mechanical treatments and hydrometallurgy.

First LIBs are crushed in a low-speed rotary shear at 11 rpm, then fragments undergo a second comminution step in an impact mill at 90 rpm to reduce particles to a size lower than 3 mm. Both

crushing steps are carried out under inert atmosphere made of CO₂ with 10-35% of Ar. Crushed fragments are sieved using a vibrating screen separating an oversized fraction (> 3mm) and an undersized one (< 3 mm). The oversized fraction, including Al, steel, and plastics from the housings and the major fraction of current collector foils and separator, undergoes a magnetic separation to remove ferrous metals and the remaining non-magnetic materials are processed with a densiometric table. Thanks to the relative difference in density, fragments are divided in a heavy fraction, containing mostly Cu and Al foils, and a light fraction, mostly made of plastics. The undersized fraction contains the anode and cathode active materials and traces of current collector foils, so it is further sieved at mesh size of 500 μm separating impurities of Cu. The sieved powder consists of cathode and anode active materials with less than 0.3 wt% of Cu impurities. The electrodic powder is mixed with water and steel shots with a steel-to-powder weight ratio of 0.15 and the pH is adjusted to 12-13 using LiOH alkaline solution. The hydrolysis of Li produces gaseous explosive hydrogen, so the tank is blanketed with CO₂ or Ar coming from the flue gas of inert crushing. Li from the cathode material is dissolved in aqueous phase whereas other metals oxide and graphite are separated by filtration. Some of the lithium is precipitated as Li₂CO₃ using part of the flue gas of inert crushing rich of CO₂ and the remaining lithium is precipitated as lithium phosphate in a neutralization step with 85 wt% H₃PO₄ solution. The precipitation of lithium with the neutralization step is carried out at pH 8.5 and achieves a recovery efficiency of Li up to 99 wt%. The precipitation occurs at pH around 9, then the precipitate is washed with CO₂ saturated solution and dried at 105°C.

The remaining solid fraction is leached in 1 mol L⁻¹ H₂SO₄ solution at 80°C and pH lower than 3, which is able to dissolve in solution almost all the metals, whereas the insoluble graphite is filtered out. Then remaining traces of Cu ions are cemented with steel shots cooling down the solution at 60°C, resulting in the precipitation of elementary metallic Cu, which is separated by filtration. Then 20 wt% NaOH solution is added to increase the pH to 3.85 to precipitate iron hydroxides. Finally, Co is recovered from the solution through crystallization as Co(OH)₃ in presence of NaClO or through electrolysis as elemental Co. The electrolysis is performed at 55°C, current density of 400-

600 A m⁻² by using stainless steel cathode and antimony-lead alloy anode. The process solution incoming the electrolysis is neutralized at pH 5.8 and after the process is recycled to the leaching step. The part of the solution which is sent to the Co precipitation process is adjusted to pH 2.3-2.8 using NaClO, thus Co hydroxide is precipitated and filtered out whereas the solution containing lithium sulphate traces is sent to the lithium precipitation step.

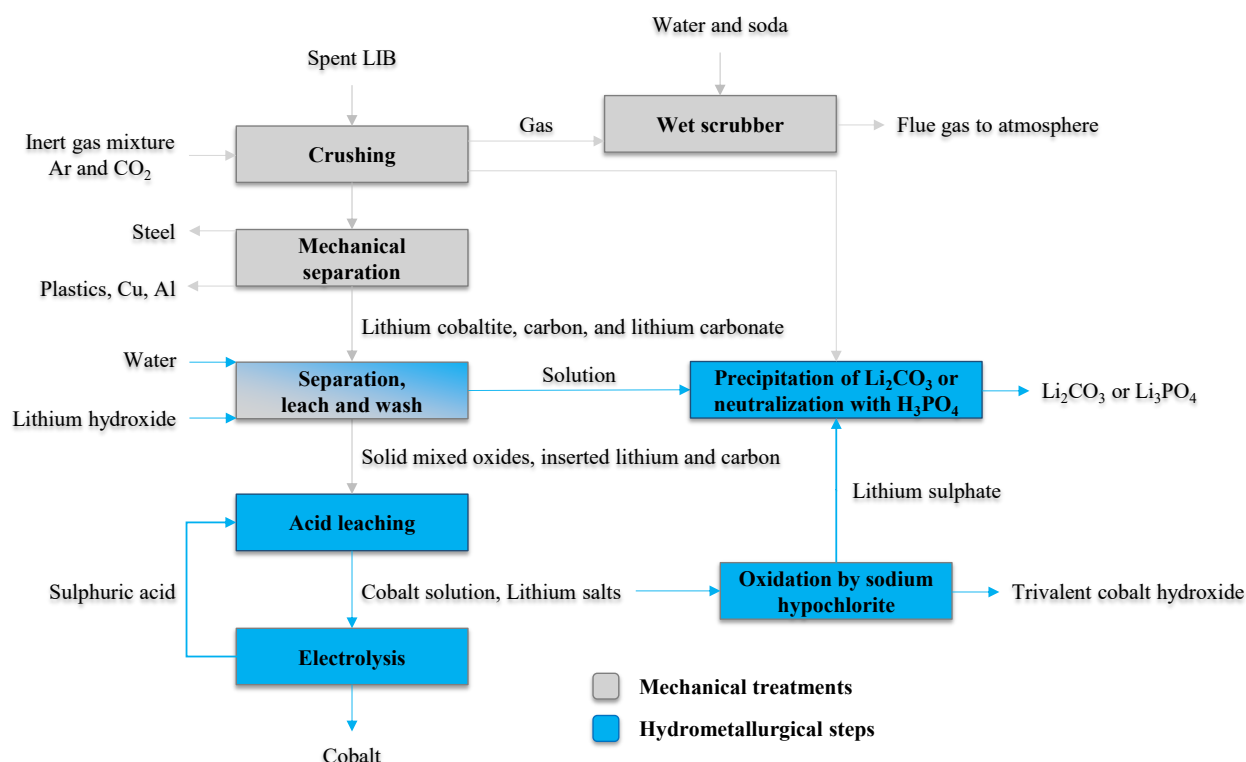


Figure S6. Simplified flowchart of Recupyl process ¹².

S1.9 Neometals

Neometals Ltd. is an Australian lithium mining and refining company which entered in the lithium battery recycling market ²⁵⁻²⁷. After three years of development including bench and pilot trials, Neometals developed a recycling joint venture called “Primobius GmbH” with the German Company SMS group. First a pilot plant was tested in Lakefield, Canada, then in 2020 the joint venture built a LIB recycling demonstration plant in Germany and in 2021 started carrying out feasibility analysis and market evaluation in order to make a decision on commercial deployment. A

commercial plant with a capacity of 20,000 t y⁻¹ is under investigation. Primobius has also announced that its German demonstration plant will treat 1 t of black mass per day in 2022, thus achieving an annual capacity of recycled spent LIBs corresponding to ca. 10,000 t. The company is also studying a business model with several decentralized plants operating the mechanical treatments and a single centralized plant that carries out the hydrometallurgical treatments.

The Neometals process is a combination of mechanical and hydrometallurgical treatments designed for portable electronics LIBs mostly with LCO chemistry, and large format LIBs with NMC and eventually NCA chemistry coming from EVs and stationary energy storage. The key minerals targeted for recovery are lithium, cobalt, nickel, copper, iron, aluminum, graphite, and manganese, but also plastics, aluminum foil, and copper foil are mechanically separated in the first part of the process. Metals from cathode active material and lithium are recovered with a battery grade purity to create a closed loop of recycling. Neometals claims an overall recycling efficiency higher than 90 wt%.

The process starts with a sorting, followed by a two-stage wet shredding. The crushed fragments are sieved to separate a slurry containing electrolyte, black mass, and the brine. The oversized fragments are fed to a zig-zag sifter, which separates a light plastic fraction and a heavy fraction including steel, aluminum, and copper. The heavy fraction could be further refined to separate metals. The black mass slurry is fed to a plate and frame filter to obtain a black mass solid cake and a solution containing diluted electrolyte. A vacuum dryer follows, which removes the remaining electrolyte and brine. The dried powder is then crushed with a mill and sieved. The resulting black mass can be stored and sent to the hydrometallurgical section of the plant, where it is leached; the unleached graphite is filtered out through a filter press. The leached solution is sent to a crystallization step to remove copper sulphate, which is then filtered out. Another impurities removal step follows, where Al and Fe hydroxides are precipitated by adjusting the pH with NaOH solution. Then, undisclosed solvent extraction and precipitation phases are carried out to obtain in

chronological order manganese sulphate, cobalt sulphate, nickel sulphate, lithium sulphate and ammonium sulphate.

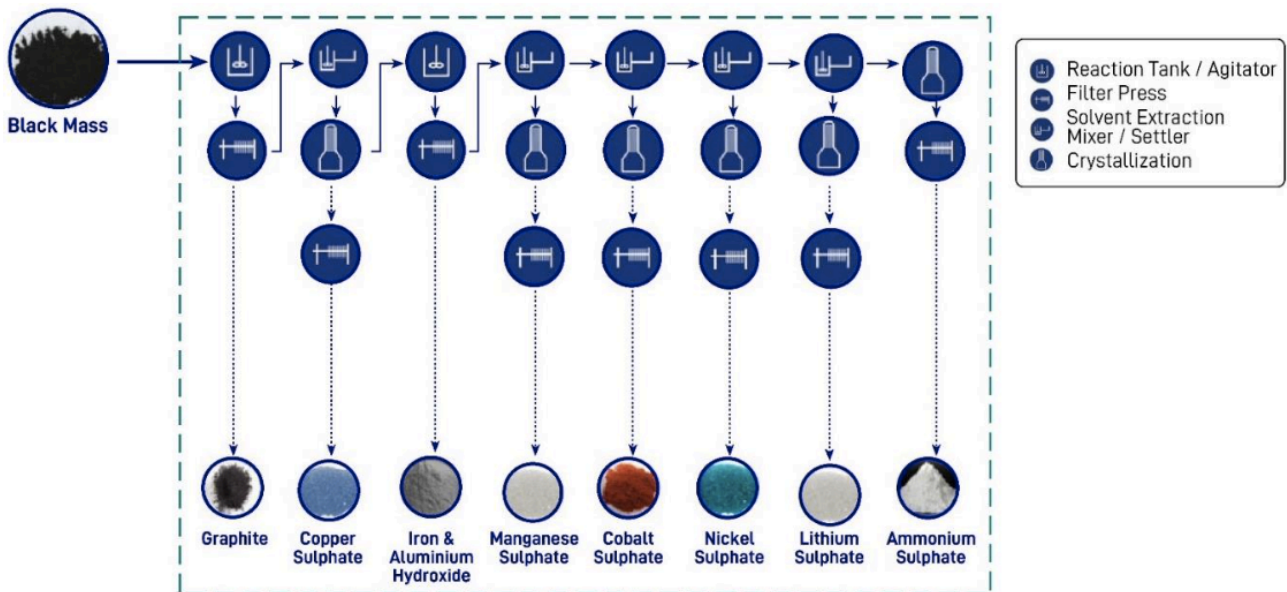


Figure S7. Hydrometallurgical steps in Neometals process²⁶.

S1.10 NorthVolt AB, Fortum Oyj and Stena

NorthVolt AB is a Swedish leading battery company in Europe, which has decided also to invest in LIBs recycling. HydroVolt is owned by NorthVolt and the Norwegian aluminum producer Norsk Hydro. The plant in Fredrikstad, Norway, has been operating since the end of 2021 and has a capacity of ca. 8,000 t y⁻¹ of spent batteries. This process is mainly mechanical with the aim to separate aluminum, addressed to Norsk Hydro, and black mass, sent to NorthVolt battery production. In addition, NorthVolt developed its personal recycling process called ReVolt, which will be implemented in a new recycling facility situated near Northvolt Ett gigafactory in Skellefteå, Sweden. The plant will be operative in 2023 and the process is claimed to recycle 97% of the LIB. The ReVolt process starts with a discharging step for energy recovery followed by a robot-assisted dismantling step. Batteries are shredded and several undisclosed mechanical separations are carried out to recover metal scraps, black mass and also the electrolyte. Finally, the black mass is fed to a hydrometallurgy-based section to recover battery grade metal salts, that will be re-introduced in NorthVolt battery

production chain. According to the business plans, by 2030 the ReVolt recycling facility will treat ca. 125,000 t y⁻¹ of spent batteries, allowing NorthVolt to introduce 50% of recycled materials in its battery production.

Fortum Oyj is a Finnish company already carrying out a LIB recycling process based only on mechanical treatments to recover the black mass. The stated annual capacity of its mechanical process is ca. 3,000 tons of spent LIBs. Fortum Oyj has announced a new hydrometallurgy-based LIBs recycling plant, that will be ready 2023 in Harjavalta, Finland, to treat the black mass coming from the former established mechanical process. As a whole, LIBs are first disassembled and treated at Fortum's mechanical separation plant in Ikaalinen, Finland, allowing the recovery of plastics, aluminum, copper, and black mass. Black mass will be addressed to Fortum's hydrometallurgical plant in Harjavalta to obtain battery-grade metal salts. Fortum declares that 80% of the battery will be recycled and the 95% of metals contained in the black mass will be recovered.

Stena is a Swedish company currently operating collection and sorting especially of small format LIBs coming from portable electronics, summing up to ca. 1,500 t y⁻¹, which account for about half of Sweden small format spent LIBs. Stena has announced the development of a new hydrometallurgical facility in Halmstad in collaboration with the multinational company Johnson Matthey, claiming that 95% of the LIB will be recycled from starting from 2023.

S2. Description of recycling processes in North America

S2.1 Retrieval Technologies

Retrieval Technologies, called "Toxco" until 2013, commercialized one of the first lithium-ion batteries recycling processes in 1992^{7-9,11-13,21,22,25,28,29}. It is an established recycling route focused on LIBs and based on hydrometallurgy without any pyrometallurgical step.

EV battery packs are manually disassembled. In the former Toxco process the reactive potential of LIBs was reduced by cryogenic cooling to -160°C with liquid nitrogen, which avoided short circuits due to the frozen state of the electrolyte. Currently this step is not carried out anymore for LIBs but only for batteries that could contain large amount of flammable metal Li like primary lithium batteries. So, nowadays dismantled LIBs are shredded and wet milled using first a shredder and then a hammer mill both operating under brine solution consisting of lithium brine, which is re-circulated from a subsequent process step. A light floating fraction composed of plastic and steel fragments from outer housing is separated. If the content of Fe is high enough (65 wt% or more) the fraction is sold to metal industries otherwise it is sent to disposal. After milling, shredded fragments with an average dimension lower than 2.5 cm are screened at mesh size of $707\ \mu\text{m}$ and fed to a shaker table, which separates a solid mixture of copper, aluminum, remaining plastics and active materials inclusions and a liquid slurry containing lithium, cobalt and graphite. The solid metal fraction is dried and sold to other primary metals producers. The cold brine solution containing active materials is transferred into a tank, where LiOH could be added to keep the pH constant at 10 avoiding the use of NaOH, that contaminates the brine. Afterwards, the solution is filtered by a filter press, whereby the active materials are separated as a mixture in form of a filter cake and are sold to other recycling processes, such as the pyrometallurgical facility of Glencore. The filtered lithium brine solution is transferred to a downstream tank where it is partially re-circulated to the previous crushing process or transferred to a following mixing tank. In this tank the solution is heated and concentrated through water evaporation, then the solution is transferred to a stirred tank where lithium is precipitated as Li_2CO_3 by addition of Na_2CO_3 . The insoluble product is dewatered in a further filter press achieving a filter cake with 28 wt% moisture. The press has an electrolytic membrane selective for Li ions and, for further refining of Li_2CO_3 , a subsequent dissolution in sulfuric acid is carried out, resulting in the dissolution of metals. Only Li ions pass through the membrane and then precipitate as LiOH, which is converted to Li_2CO_3 via CO_2 bubbling. The produced Li_2CO_3 has a 99 wt% purity but is currently sold to steel manufacturers. Retrieval carries out a preliminary analysis to determine the contained

metal values of each cell and, depending on metal content and current market values, it sets a gate fee to be paid by the battery owner or a credit to be assigned.

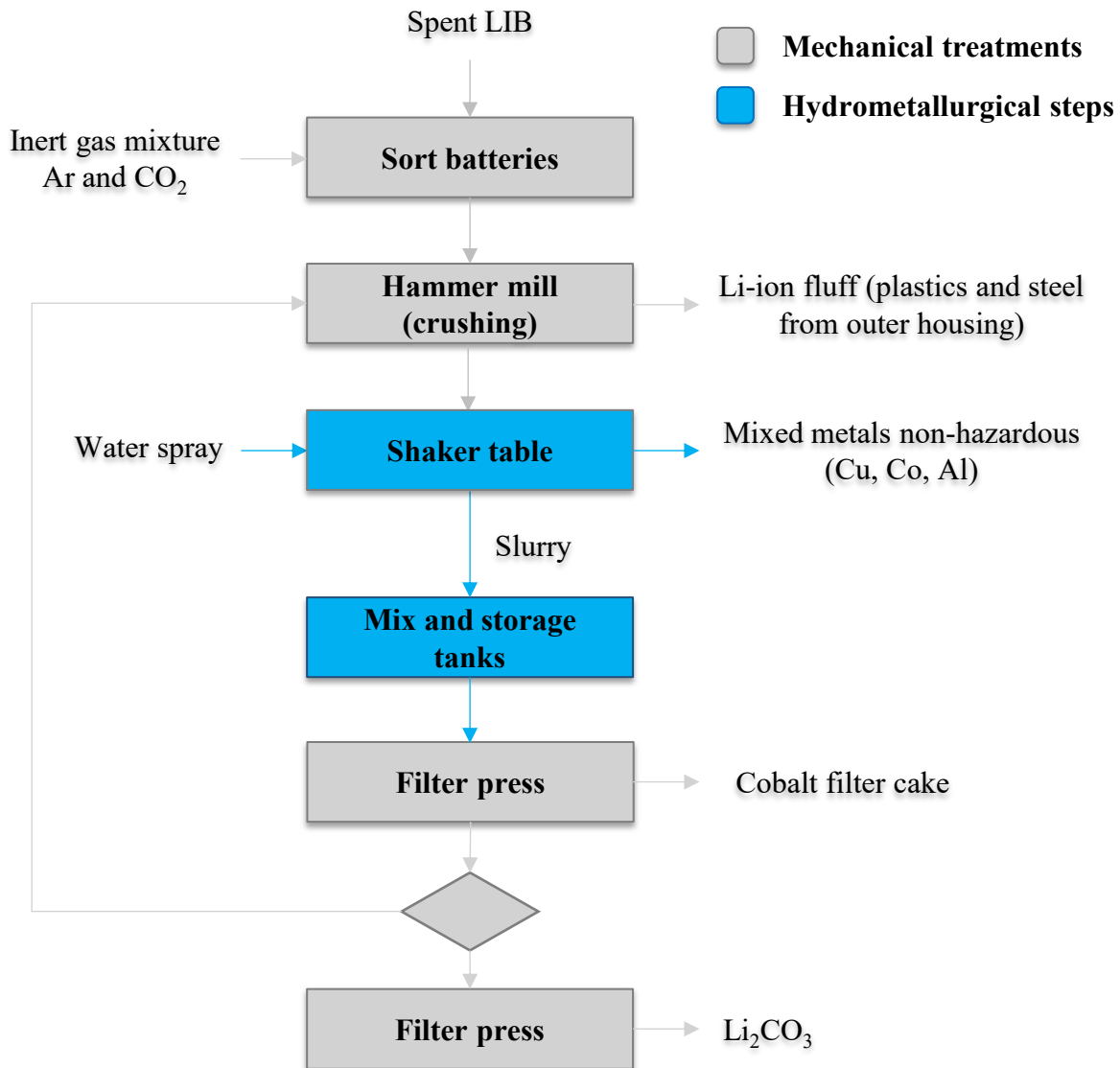


Figure S8. Simplified flowchart of Retrieval process¹².

S2.2 INMETCO

The International Metals Reclamation Company (INMETCO) is a metal recycling company located near Pittsburgh (USA) that handles common industrial metal wastes such as electric arc furnace dust, mill scale, swarf, and flue dust from stainless steel manufacturers^{7,11,25,30}. It was established in 1978 and in 2009 became a subsidiary of American Zinc Recycling. INMETCO in

1995 opened a recycling unit specifically designed for Ni-Cd batteries. Since 2005 the unit recycled also zinc-carbon batteries, Ni-metal hydride batteries and also Li-ion batteries containing Ni, for a total of 6000 t y⁻¹ of treated materials. Ni-Cd batteries continue to have their own unit whereas other batteries are fed to the primary unit treating also common metal scraps.

The feed is treated in a rotary hearth furnace at 1350°C together with carbon-based reducing pellets. Pellets are 12 mm in diameter and the load corresponds to three layers of pellets. The molten material is refined in an electric arc furnace recovering a Co-Ni-Fe alloy, whereas other metals such as Li are lost in the slag, while organic materials like the electrolyte solvents are burned. In the unit treating only Ni-Cd batteries the flue dust containing volatile Cd is further refined obtaining Cd with 99.95 wt% purity.

S2.3 Glencore

Glencore merged with Xstrata in 2013 and acquired also Sudbury Integrated Nickel Operations (INO)^{7,11,25}. Xstrata was a Swiss mining company, which started recycling batteries by adding battery scrap as secondary feedstock material in the extractive cobalt, nickel, and copper process routes. An advantage of this method is the reduction of energy consumption regarding the waste gas treatment. Because of the added sulfur-free battery scrap, the content of sulfur in the waste gases is reduced noticeably compared to the pure ore, that is mainly composed of sulphates of Co and Ni. There are also energy savings of about 75% because of the added secondary cobalt compared to the processing of the pure ore. Since 2008, Xstrata implemented a pretreatment of the LIBs in a rotary kiln to dry out the electrolyte. Similar to Inmetco, the Xstrata process targets the recovery of cobalt and nickel from the LIBs. Other contained materials just serve as reductants and are slagged or leave the process as waste gases.

Sudbury INO is a battery recycling company that in the past treated mostly small portable batteries, while currently it also handles EV batteries. After the pre-treatment in a rotary kiln to

remove electrolyte and plastics, battery scraps are fed to a furnace operating at 1300°C. Li is lost in the slag, whereas Ni and Co are collected in the alloy. Co is extracted from the alloy using HCl as leaching agent and then is addressed to Glencore Refinery in Norway. Sudbury INO treats only Ni and Co-containing Li-ion batteries, whereas LFP batteries are not accepted unless after payment of a treatment charge.

S2.4 Ascend Elements (former Battery Resourcers)

The Battery Resourcers process is considered a “closed loop” process because it recovers materials from LIBs suitable for LIB production^{11,13,25,31–34}. It was founded in 2015 and it is a spin-off of Worcester Polytechnic Institute (WPI) based in Worcester, MA, which has developed a recycling process with an exclusive license from WPI that recovers cathode materials from a mixture of EOL LIBs. In 2018 the bench scale (50 kg of EOL batteries per batch) was upgraded to the pilot scale to process daily 500 kg of spent LIBs from major vehicles as well as production scrap from leading battery manufacturers. In 2020 Battery Resourcers opened a facility with a capacity of 5 t per day of spent EV batteries. The process is based on the co-precipitation method, which involves mechanical and hydrometallurgical treatments with a final single sintering step to produce cathode materials. It is specifically designed to treat LIBs with NMC chemistry but can accept also other chemistries as impurities. In January 2022 it changed its name into Ascend Elements, but the following description refers to the recycling process up to such a rebrand.

Spent LIBs are first over-discharged and then shredded with a hammer mill producing fragments with particle size lower than 6.3 mm. Wet fragments are soaked in propylene carbonate (PC) to remove the lithium conductive salt LiPF_6 , which can be recovered by evaporating the solvent at low temperature and pressure. Next, the undissolved material is filtered out and immersed in 1-methyl-2-pyrrolidone (NMP) at 70°C to dissolve the PVDF binder and thus separate current collector foils from active materials. Fragments are treated by a magnetic separation to recover a magnetic

fraction composed mostly by steel. The non-magnetic fraction is composed by current collectors foils and electrodic material and is mixed with NaOH solution to extract Al as NaAl(OH)_4 . The slurry is then filtered and dried at 60°C . The dried fragments containing mostly plastics, Cu foils, graphite, cathode material are sieved at mesh size of $250\ \mu\text{m}$. The coarser fraction undergoes a dense media separation to obtain a Cu fraction and a plastic one. The sieved powder composed by graphite, cathode materials, and impurities are sent to the hydrometallurgical refinement. A leaching step with H_2SO_4 $4\ \text{mol L}^{-1}$ and 30-50 wt% H_2O_2 at $70\text{-}80^\circ\text{C}$ for 2-3 hours is carried out to dissolve the cathode material with NMC chemistries and separate unleached LFP cathode material, graphite, and plastics impurities. The remaining solution contains Co, Ni, Mn, Li Al, Cu and Fe ions, so that an impurities removal step is performed by adding NaOH solution. At pH 6.5 Fe, Al, and Cu ions precipitate as hydroxides, whereas Co, Ni, Mn, and Li ions remain in solution. After filtering out the precipitated salts, a stoichiometry adjustment is carried out by adding MnSO_4 , NiSO_4 , and CoSO_4 to obtain the desired molar ratio (e.g., 1:1:1, 5:3:2 or 6:2:2 of Ni, Mn, and Co, while the production of NMC811 is currently under development). Then a transition metal hydroxide precursor ($\text{Ni}_x\text{Mn}_y\text{Co}_{1-x-y}(\text{OH})_2$) is co-precipitated by increasing the pH to 11 (range 10.5-13) with NaOH and in presence of NH_4OH solution as chelating agent. Each hydrometallurgical operation is realized in a stirred tank blanketed with N_2 to avoid the oxidation of Mn^{2+} to Mn^{4+} , that can easily happen in presence of oxygen. After filtering out the precursor, the Li rich solution is mixed with Na_2CO_3 solution at 40°C to precipitate Li_2CO_3 . The transition metal hydroxide precursor and the Li_2CO_3 are dried and mixed with some additional virgin Li_2CO_3 necessary to reach the right stoichiometry, that is a Li-to-metals precursor molar ratio of 1.1. The blended powders are ball milled for 48 hours and compressed into pellets which are sintered at 900°C for 12-15 hours to produce cathode material with the chosen NMC chemistry. The most valuable products offered by Battery Resourcers are battery grade Li_2CO_3 , NMC hydroxide precursors and NMC sintered materials with molar ratio 111, 532 and 622 and with d_{50} equal to $5\text{-}6\ \mu\text{m}$ or $10\text{-}12\ \mu\text{m}$. The recovery efficiency of Co, Ni and Mn is respectively 94, 96 and

91 wt%³¹, whereas the Li recovery efficiency ranges between 65 and 80 wt% according to the mixed LIB chemistry feed.

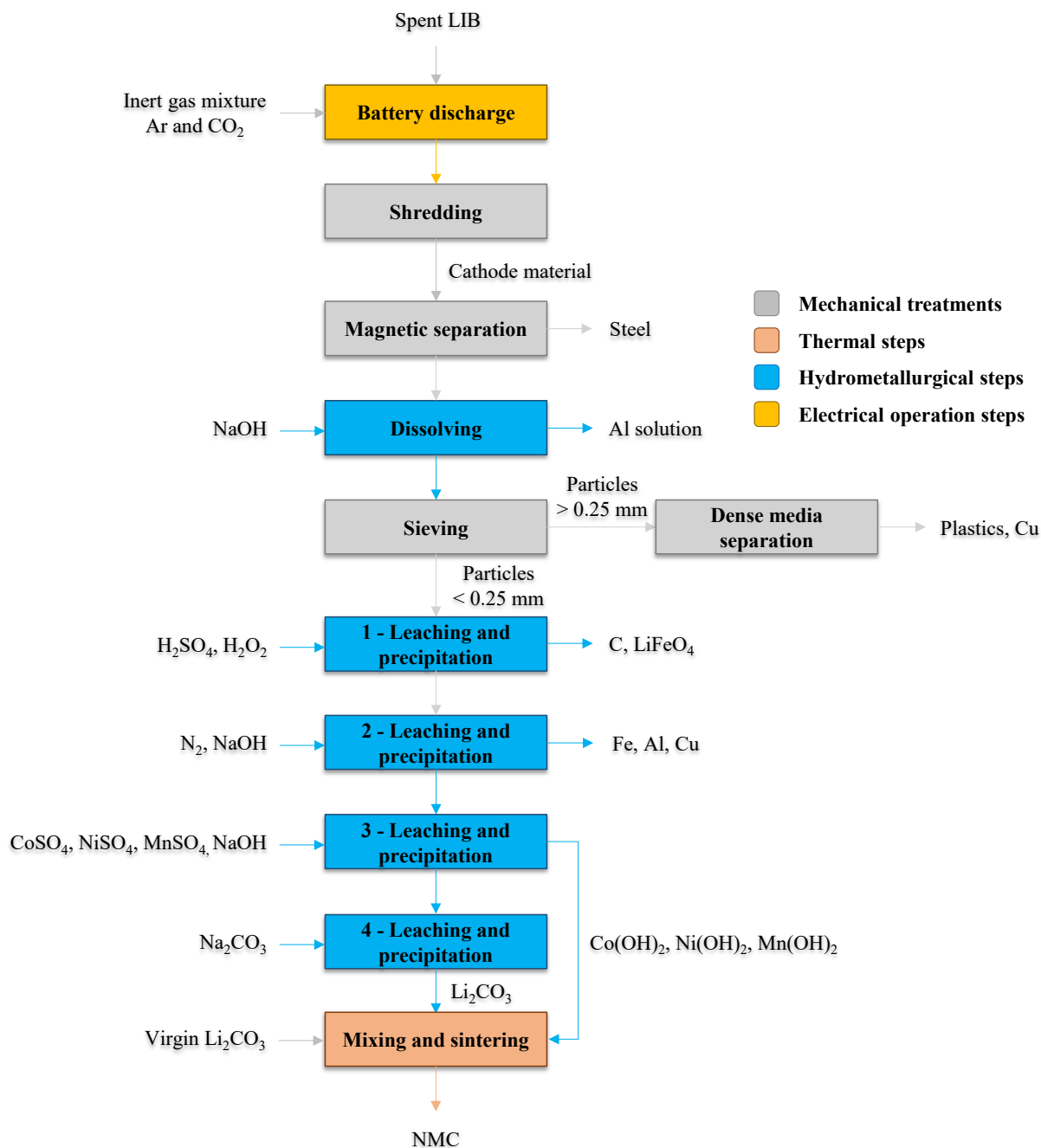


Figure S9. Simplified flowchart of Battery Resources process¹¹.

S2.5 OnTo Technologies

OnTo is a company founded in 2004 which develops and patents advanced battery recycling innovations that produce manufacturing quality electrode materials from recycled LIBs^{11,25,35–42}. These breakthrough technologies are available to serve the battery and environmental services industries. In 2020 OnTo won the Phase II of the “Lithium-Ion Battery Recycling Prize” by the U.S. Department of Energy (DOE).

The OnTo recycling process, also called EcoBat, is based on the direct recycling concept, patented and implemented at laboratory scale. It starts with an over-discharging step, followed by disassembly to obtain single battery cells or small modules. Then cells undergo a perforation stage and an electrolyte separation step is carried out in a chamber using super-critical CO₂ (74 bar and 31°C) and additional polar co-solvents. This operation allows the extraction and recovery of the whole electrolyte including LiPF₆ salt. Then, dried cells are shredded at ambient condition using a ball mill, crushed fragments are sieved obtaining an over-sized fraction composed of Al and Cu foils, plastics and steel, along with an under-sized fraction including active materials and impurities. The coarser fraction undergoes a magnetic separation to separate steel. The sieved powder is fed to a dense media separation (DMS) to separate graphite from cathode active material. The DMS is carried out in media of about 2.2-3.5 g cm⁻³. The obtained active material powders are addressed to the hydrometallurgical section, where they are purified first: the graphite fraction undergoes a low-pH treatment to dissolve cathode active material impurities and a high-pH treatment to remove remaining Cu and Al; on the other hand, the cathode fraction purification occurs in a basic solution of LiOH at pH 11-11.5 to dissolve the PVDF binder, then the solid is filtered out and washed using acetone or ethanol. Both purifications are able to produce streams with a purity of 99%. The purified graphite is then fed to a furnace to remove PVDF and remaining plastic impurities, thus producing battery grade graphite. The cathode material is dried at 140°C in a vacuum oven, ball milled and sieved at mesh size of 45 μm or preferably 38 μm. The cathodic powder is mixed with LiOH solution and a

hydrothermal regeneration is carried out in order to restore the lithium lost during long cycling and degradation. One liter of 24 wt% LiOH is added for each kilogram of active material. The slurry in the autoclave is heated up to 250-275°C at rate of 5°C min⁻¹ and then temperature is kept constant for 12-14 hours. Next the solid is filtered out and washed with demineralized water or LiOH solution until pH drops under 11-11.5 due to NMC hydrolysis. The regenerated cathodic material is dried at 150-160°C and grounded. The powder is sintered under air or oxygen flow in a tube furnace. The temperature is increased at a rate of 2-3°C min⁻¹, kept constant at 900°C for 8 hours and rapidly decreased at ambient temperature at about 50°C min⁻¹. Also, graphite undergoes a high temperature treatment to remove impurities and intercalated lithium. These treatments allow to produce battery grade active materials at costs below 10 \$ kg⁻¹ using less energy than in the original manufacturing.

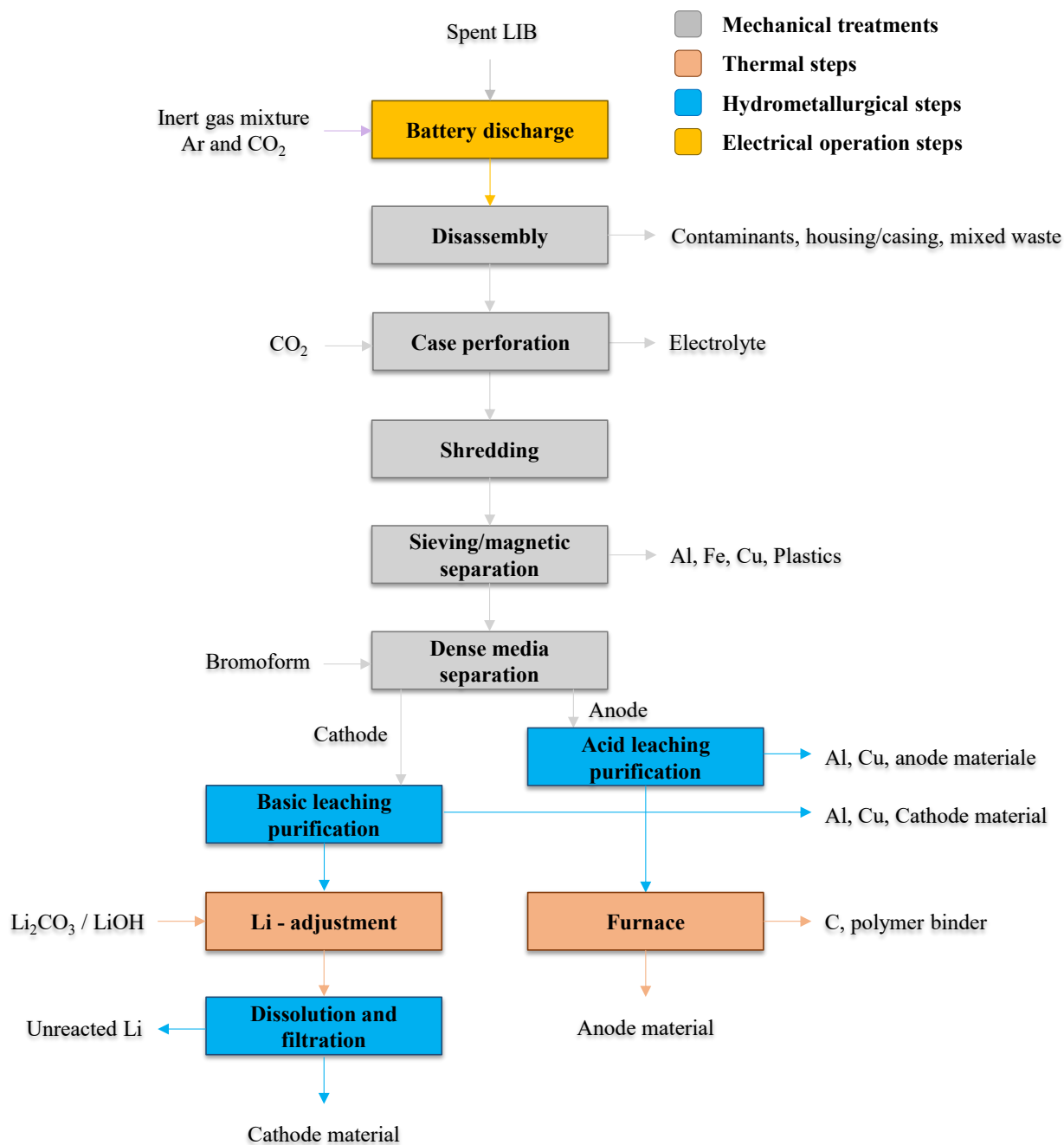


Figure S10. Simplified flowchart of OnTo process ¹¹.

S2.6 Li-Cycle

Li-Cycle is a LIBs recycling company based in Mississauga, Ontario, since 2016 ^{25,43-45}. The company is looking for achieving an overall recycling efficiency between 80% to 100% of material from both portable and EV batteries. First in 2017 a mini pilot plant with a recycling capacity of 50 t

y^{-1} was built in Canada, then Li-Cycle has received funding from Sustainable Development Technology Canada (SDTC) to build a demonstration plant in Kingston, Ontario, which included the Spoke and the Hub and was built in 2018. The following steps were the opening of commercial Spoke with a capacity of 5,000 t y^{-1} , the first one was opened in 2019 in Canada, whereas the second one was built in New York State in 2020. In 2022 another commercial Spoke is expected to open in Arizona and it will be able to recycle 10,000 t y^{-1} of LIBs, in addition in 2023 Li-Cycle will build the first commercial Hub, that will be set in Rochester, New York. and will treat black mass coming from the equivalent of 60,000 t y^{-1} of LIBs.

The company is proprietary of two different patented technologies, that are the Spoke and the Hub. The Spoke is a mechanical process including wet crushing, magnetic separations, sieving and other material separations to obtain mainly black mass, but also an aluminum and copper foils fraction, a plastic stream and a steel fraction.

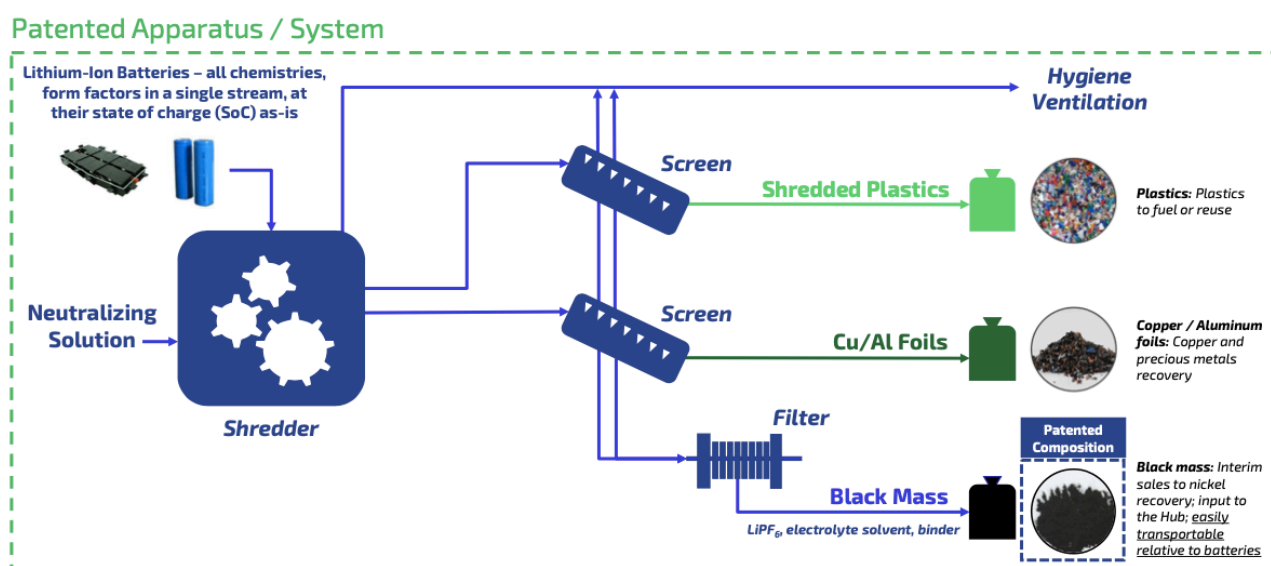


Figure S11. Spoke mechanical process⁴³.

The Hub is a hydrometallurgical process (still under development) which treats the black mass from several Spoke facilities to obtain as main products Li_2CO_3 and Ni-Co-Mn hydroxide cake, which will be sent to South Korea for battery production. A future development will be the creation of

further hydrometallurgical steps to produce separated streams of nickel sulphate, cobalt sulphate, and manganese carbonate. A graphite fraction is separated but it will be unlikely used in battery industry due to very stringent quality constraints. Li-Cycle supposes that the best and suitable strategy for market commercialization is the decentralization of tailored Spoke plants, that are close to the highest volume points of consolidation of LIBs, and the centralization of the Hub hydrometallurgical process. The main advantage is the generation of non-hazardous intermediate products close to the point of consolidation/generation of spent LIBs, which leads to the minimization of transportation liability and cost.



Figure S12. Hub hydrometallurgical future process, including stages which are currently under development ⁴³.

In the process proposed by Li-Cycle spent LIBs are optionally discharged to approximately 0 V to recover electricity. Then LIBs are fed to a wet crusher with two different feed lines: large format LIBs coming from EVs or industrial applications enter the equipment in a dedicated opening and undergo a preliminary crushing at 10-20 rpm to obtain scraps with a size lower than 400 mm; portable LIBs are fed in the other opening and, together with the scraps from the previous pre-crushing, are fed to a second shredder operating at 30-50 rpm to produce fragments with a size lower than 100 mm.

The whole equipment is submerged adding 2 m³ of a solution containing Ca(OH)₂ and NaCl at 60°C for each m³ of LIBs: Ca(OH)₂ solution is used to create an alkaline ambient, which neutralizes and removes all the HF toxic gas produced by the hydrolysis and decomposition of LiPF₆, while NaCl is added as conductive medium to dissipate the residual charge of the batteries. The crushed slurry is screened and undersized fragments under 10 mm pass whereas oversized fragments are sent another time to the crusher. The undersized fraction undergoes a solid-liquid separation using a belt filter and then is sent to a magnetic separation, which separates steel and ferrous product with a recovery efficiency of 95 wt%. The magnetic fraction is then dewatered using a dewatering screen that leaves fragments with only the 1 wt% of moisture. The non-magnetic stream is stripped by mixing at 10 wt% with NMP at 80°C for 6 hours. This step is necessary to dissolve PVDF binder to enhance the detachment of active materials from current collector foils. Then the slurry is sieved at mesh size of 500 μm producing an undersized fraction containing the liquid solution and the black mass and an oversized fraction containing current collectors and plastics. This oversized stream is sent to an eddy current separation, which separates almost all aluminum and some copper. Alternatively, plastics are separated by using a liquid with density of 2.5 kg L⁻¹ and aluminum is separated from copper using a liquid with density of 2.85 kg L⁻¹. The densiometric separations have an efficiency beyond 95 wt%. The undersized fraction undergoes a solid-liquid separation to remove the organic solvent. The recovered black mass is dried and sent to the Hub to carry out the hydrometallurgical process.

The black mass is leached with 0.5 mol L⁻¹ H₂SO₄ and 20-30 g L⁻¹ H₂O₂ for 6 hours at 80°C: H₂O₂ is added stoichiometrically, whereas H₂SO₄ is added with a 10% excess. The slurry contains 10 wt% of solid and the pH is kept at 2.5 by adding H₂SO₄ during the reaction time. The leaching results in the recovery of 95% of all the metals. A solid-liquid separation step is carried out to separate the unleached solid from the leachate. The residual solid is mixed with water, the pH is adjusted to 5 and through a floatation step the hydrophobic graphite is separated from hydrophilic metals with a recovery efficiency of 80 wt% or more. The leachate is added with 50 wt% NaOH adjusting its pH to 2. Cu is removed using solvent extraction with organic LIX 984N at 30 vol.% in kerosene. The Cu

extraction efficiency is higher than 95 wt%. The extracted Cu is removed from the organic phase using 1 mol L⁻¹ H₂SO₄ and the aqueous solution rich in Cu (10 g L⁻¹) is sent to electrowinning to produce copper plating with a conversion efficiency higher than 85 wt% and achieving a purity of 99.9 wt%. The electrowinning is carried out with a current density of 250 A m⁻², a cathode voltage of 0.34 V and an anode voltage of -1.23 V. The raffinate aqueous process solution is adjusted to pH 4.5 at 50°C using 50 wt% NaOH, which causes the precipitation of Al and Fe hydroxides. A solid-liquid separation follows to remove the precipitated impurities from the process solution. Then the solution is adjusted to pH 9.5 at 50°C adding 50 wt% NaOH to precipitate single-phase Co, Ni and Mn hydroxides. The solution is stirred for 1 hour. The precipitated salts are filtered out and dried in an oven at 80°C. The filtrate is evaporated at 95°C and 0.85-1 kPa to increase the concentration of Na ions up to 70 g L⁻¹. Then the pH is adjusted to 9.5 by adding 50 wt% NaOH and the solution is cooled down to 10°C causing the precipitation of Na₂SO₄·10H₂O. The salt is filtered out and dried under vacuum to produce Na₂SO₄. The process solution is then evaporated to reach a concentration of Li equal to 11 g L⁻¹ and a saturated solution of Na₂CO₃ at 90°C with concentration of 430 g L⁻¹ is added such that the carbonate is 1.25 times the stoichiometric requirement to precipitate Li. The mixture is stirred for 6 hours at 95°C and the precipitated Li₂CO₃ is then filtered out, washed twice with hot water at 70°C and dried in an oven at 80°C. The lithium recovery step has an efficiency of 90 wt% and the Li₂CO₃ has a purity of 89 wt%, thus it needs to be purified to comply with battery industry constraint of 99.5 wt% purity. Finally, the step of Na₂SO₄ precipitation can be repeated to recover the remaining sulphates. A simplified flowchart of Li-Cycle process can be found in ⁴⁵.

S2.7 American Manganese Inc

American Manganese Inc is a Canadian company founded in 1987 in British Columbia, Canada ^{25,46,47}. The company owns manganese mining operations in Arizona and started the recycling business as a result of testing new ways to recover more manganese from low concentration sources.

The company has developed the RecycLiCo patented process for recycling Li-ion batteries in an environmentally friendly way achieving a closed loop of the cathode materials. This process is able to recover cathode metals such as Li, Co, Ni, Mn and Al using hydrometallurgical treatments. The process input consists of black mass or cathode scraps from battery manufacturers with NMC, LCO, NCA and also LMO chemistry, whereas the output includes cathodic material metal precursors, lithium carbonate, aluminum foils and others recycled streams such as graphite depending on the feed. The recovered cathodic material precursor and lithium carbonate are battery-grade and the material recovery efficiency is extremely high. Currently the process is carried out at laboratory scale at a rate of 1 kg h⁻¹ of treated black mass, while the target is a demonstration plant with a cathode waste processing capacity of 500 kg per day. After the demonstration stage, the company will build a commercial recycling plant with a daily capacity of 5 t, that will be located near a battery manufacturing Gigafactory, such as the commercial partner Italtvolt, to treat cathode manufacturing scraps to produce cathode precursors to be reintegrated in the battery manufacturing process. Italtvolt Gigafactory in Scarmagno, Italy, is planned to start its production in 2024 with a full capacity production of 45 GWh, corresponding to more than 75,000 t y⁻¹ of cathode active material. Battery manufacturing leads to generation of an amount of scraps equal to near the 10 wt% of production, thus the recycling facility of American Manganese Inc will handle more than 7,500 t y⁻¹ of cathodic scraps.

The RecycLiCo process is a hydrometallurgical process based on the co-precipitation method. The process accepts as input black mass from mechanically processed spent batteries with cathode chemistry principally of LCO, NMC and NCA, but not mixed together, and also cathode active material scraps from battery manufacturers. Concerning cathode active material scraps, the first step is leaching with 1.2-2 mol L⁻¹ H₂SO₄ with SO₂ as reducing agent to produce sulphates and dithionate of Co, Ni, Mn, Li and Al. The leaching is carried out for 2 hours at pH around 1.5 with a solid concentration of 10 wt% in the slurry and almost 100 wt% of Li, Ni, Mn, Co and Al are leached. Then the solution is slowly mixed with 10 mol L⁻¹ NaOH to raise the pH to 10.7 to precipitate Ni,

Mn and Co hydroxides with an efficiency of metal recovery equal to almost 100 wt% for these metals. Precipitated mixed Ni, Mn and Co hydroxides are filtered out and dried. Then an amount of solid Na_2CO_3 equal to 1.2 times the stoichiometric quantity required to precipitate all Li is added to the solution to precipitate Li_2CO_3 . The mixed solution is stirred for 15-30 min at 95°C achieving up to 68 wt% lithium recovery. The precipitated solid is filtered out and washed with saturated Li_2CO_3 solution at 95°C . Then the solution containing Na_2SO_4 , $\text{Na}_2\text{S}_2\text{O}_6$ and remaining Li is cooled to 5°C and stirred for 2 hours to crystallize $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$ and $\text{Na}_2\text{S}_2\text{O}_6 \cdot 2\text{H}_2\text{O}$. Solids are then filtered out and heated at 120°C to decompose the $\text{Na}_2\text{S}_2\text{O}_6 \cdot 2\text{H}_2\text{O}$ to Na_2SO_4 and SO_2 , which can be recycled to the leaching step. The remaining solution can be recycled to the leaching step to minimize water consumption and maximize lithium recovery, or alternatively can undergo a nanofiltration to produce clean water, that can be used for washing steps. The concentrate from nanofiltration is recycled back to the crystallizer to maximize the Na_2SO_4 recovery.

Concerning black mass treatment, the process starts with a floatation step to separate the graphite and conductive carbon from the cathode active material. So, the black mass is mixed with water achieving a solid concentration in the range 5-30 wt%. Then kerosene is added and the slurry is stirred. 100 g of frother for each ton of solution are added to create stable bubbles, air is fed to create an air-bubble froth. Graphite and carbon are separated in the froth, whereas the cathode active material remains in the slurry. More than 90 wt% of the graphite is separated through this step. The slurry is then leached with H_2SO_4 and SO_2 as explained before. An impurities removal step is added to eliminate Al, Cu and Fe impurities. Thus the pH is raised to 5 using 50 wt% NaOH, causing the precipitation of $\text{Al}(\text{OH})_3$, $\text{Fe}(\text{OH})_3$ and $\text{Cu}(\text{OH})_2$. The lithium recovery step and the crystallization of Na_2SO_4 follow as previously explained. A simplified flowchart of RecycLiCo process for NMC-111 cathodic scraps from battery manufacturers can be found in ⁴⁷

S2.8 Lithion Recycling Inc

Lithion Recycling Inc. is a Canadian company, which has developed a solution to recover over 95 wt% of a lithium-ion battery, including not only high value elements like cobalt and nickel but also lithium, graphite, and electrolyte^{25,48,49}. Lithion battery recycling process is based on a combination of mechanical treatments, hydrometallurgy, and electrometallurgy-based extraction steps. Lithion is supported by strong partners such as Seneca experts-conseil, Call2Recycle, Hydro-Québec's Center of Excellence in Transportation Electrification and Energy Storage (CEETES) and Centre d'étude des procédés chimiques du Québec (CÉPROCQ). The Sustainable Development Technology Canada (SDTC) has rewarded Lithion with a 3.8 M\$ contribution, and the partners also invested in Lithion project reaching an overall investment of 12 M\$. First Lithion has developed its process steps with laboratory continuous operations, then in 2020 a pilot plant with a capacity of 200 t y⁻¹ started operating in Montreal, Canada. The company is planning to open a commercial plant with a capacity of 2000 t y⁻¹ in 2023 in Quebec, Canada.

The process starts with a wet crushing using a light organic solvent, in particular one already included in the battery electrolyte such as DMC. The organic solvent is fed at 40°C and it also works as heat sink thus reducing hazards concerning exothermic reactions and local short circuits and as extraction agent for the electrolyte conductive salt LiPF₆. The crushing step can be also done in inert atmosphere using a N₂ or CO₂ flow. The shredded fragments with particle size of about 5 to 10 mm undergo a further solvent extraction to ensure an optimal washing of the LiPF₆. The extraction is carried out with the same organic solvents at 50°C for 1 hour in a jacketed stirred tank. Then fragments are separated from the liquid by filtration and the liquid phase is fed to an evaporator operating at about 90°C and atmospheric pressure, which is the boiling point of pure DMC. In this step the lighter organic compounds evaporate and are following condensed, whereas the heavier solvents and the electrolyte salt remain in the slurry. The condensed light organics are in part re-circulated to the wet crusher and in part are sent to a purification step which is done with three

atmospheric distillation columns in series. The first column operates at 90°C and separates a head product of pure DMC and a bottom one including EMC, DEC and EC. The second distillation is operated at 107°C to obtain battery grade EMC in the column overhead and a mixture of DEC and EC in the bottom. The operating temperature in the third column is 126°C and the distillation allows to recover a head product of high purity DEC and bottom product of EC. The heavier organics slurry is burned off at 500°C to eliminate organo-fluorophosphate compounds and fluorides. The off-gas contains toxic HF and P₂O₅, which are removed respectively by using a dry scrubber with CaO and a wet scrubber with re-circulating alkaline solution of NaOH. Battery fragments are dried at 270°C to evaporate the remaining light organics, that are sent to evaporator outlet.

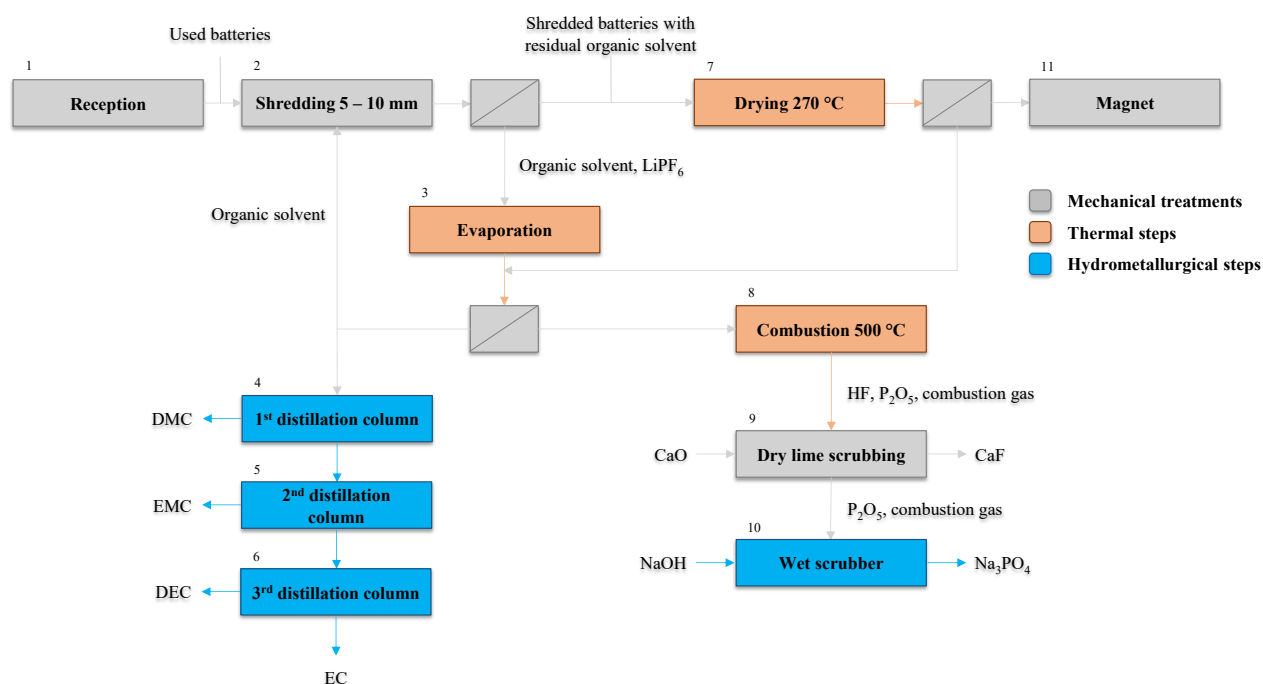


Figure S13. Lithion recycling process: shredding section ⁴⁹.

Then magnetic ferrous pieces are separated using a magnetic separation, whereas the non-magnetic fraction undergoes a further comminution step in a hammer mill to obtain a particle size of 0.1-2 mm. The crushed fragments are sieved at mesh size of 1 mm separating an oversized fraction of plastics and an undersized fraction containing active materials and current collector foils. The

oversized one is fed to a cutting mill to ensure the separation of active materials from current collector foils, and it is sieved another time. The coarser fraction, including mostly aluminum, copper and plastics, undergoes an eddy current separation to separate current collector foils. Then this metallic fraction is separated in lighter aluminum fraction and a heavier copper fraction using an air classifier. The sieved black mass, that is composed by graphite, cathode active material and Al, Cu and Fe impurities, is sent to the hydrometallurgical section.

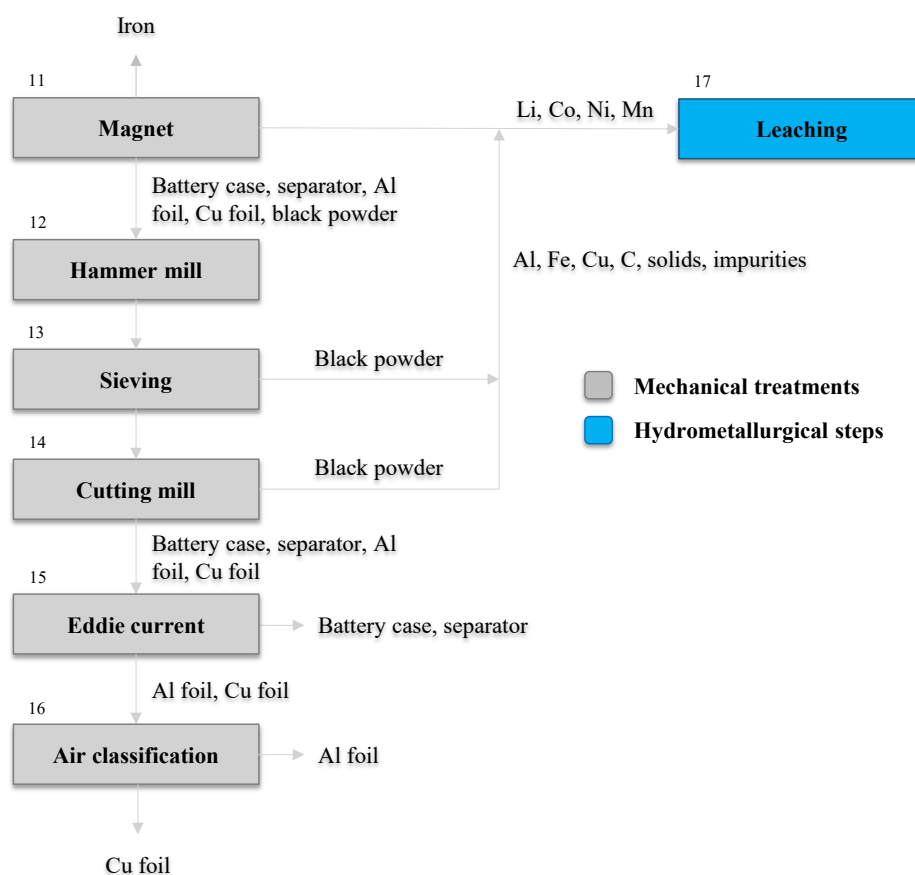


Figure S14. Lithion recycling process: mechanical separations section ⁴⁹.

The black mass is leached at 20°C with H₂SO₄ and Al, H₂O₂ or MnO₂ as reducing agent, achieving a solid concentration of 100 kg m⁻³. Metals are dissolved as sulphates, whereas the graphite does not react and is filtered out using a solid-liquid separation. The overall recovery efficiency of metals is 98.5 wt%. Graphite is then mixed with the same leaching solution to remove the remaining metals. Next there is another filtration, followed by a washing step with water. Finally, the graphite

cake is fed to a furnace operating at 600°C to dry the powder and operate the pyrolysis of plastics impurities. The leachate is mixed with Na₂S at pH 2 and 40-80°C to selectively precipitate CuS. An amount of 2-5 kg of Na₂S per kg of leachate is added and a Cu recovery efficiency of 99 wt% is achieved. Precipitated solid is filtered out and the solution is fed to another tank, where pH is adjusted to 3.5-5 by using NaOH to precipitate aluminum and iron hydroxide, that are separated with an overall efficiency of 97 wt%. Then the solution is mixed with an organic extraction solvent diluted in a petroleum-based compound, in particular a mixture of 10 vol.% Cyanex 272 and 90 vol.% naphta with a 1:1 organic-to-leachate ratio. The pH is adjusted to 5.4-6.2 to minimize the nickel extraction, thus obtaining a light organic phase, containing Co and Mn, and a heavy aqueous phase containing Ni. The recovery efficiency of Co in the organic phase is 99.9 wt% and the Ni/Co separation factor is 4000.

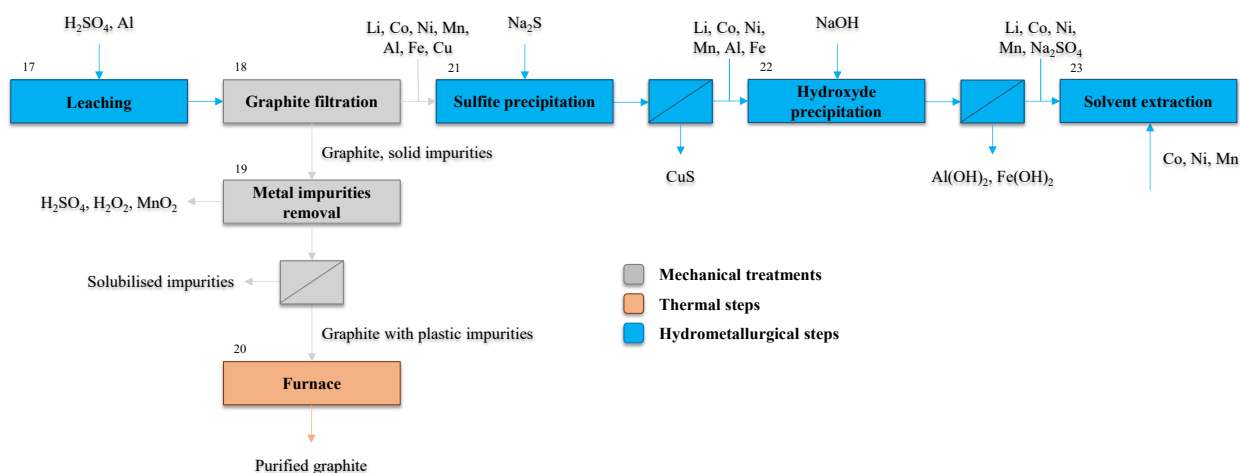
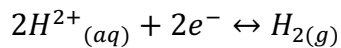
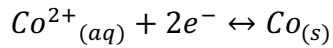


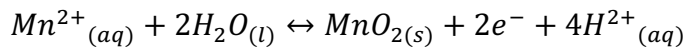
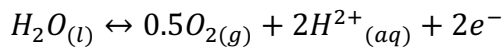
Figure S15. Lithion recycling process: hydrometallurgical section (part 1)⁴⁹.

Co and Mn are stripped from the organic phase using a H₂SO₄ solution with a pH between 1 and 2. The cleaned organic phase is re-circulated to the solvent extraction step, whereas the aqueous solution containing Co and Mn is sent to an electrochemical separation step. Even with accurate pH adjustment it is impossible to precipitate Co or Mn hydroxide with high enough selectivity, but these metals have different standard reduction potentials (-0.28 V for Co and -1.18 V for Mn) that make it possible to carry out a separation through electrowinning. The cobalt plates the cathode as elementary

metallic Co, whereas Mn deposits on the anode as MnO₂. The electrowinning process takes place at a voltage between 2.7 and 5 V and current density between 200 A m⁻² using a metallic Co cathode, a DSA (mixed metal oxide) anode and an electrolyte at pH 3.5 and at 50°C, according to the following cathodic reactions:



and anodic reactions:



After the electrowinning step, the sulfuric acid solution with poor metal concentration is recirculated to the stripping step. The Ni aqueous solution is adjusted to pH 10.8 using NaOH solution, then precipitated Ni(OH)₂ is filtered out, washed and dried obtaining a product with 99.3 wt% purity. The remaining solution is rich in Na₂SO₄ produced by neutralization reaction of H₂SO₄ and NaOH, thus the pH is adjusted to 8 and Na₂SO₄ is crystallized by cooling the solution between 0 and 10°C obtaining Na₂SO₄·10H₂O, that is filtered out. Recovered hydrated Na₂SO₄ has a purity of 99.9 wt%, thus it is dissolved in solution and fed to an electrolysis cell, that produces H₂SO₄ at the anode and NaOH at the cathode. The electrolysis is carried out at voltage of 10 V and current density of 1000 A m⁻² using a solution of 18 wt% Na₂SO₄ at 25°C. The mother liquor from crystallization step is rich in lithium, thus it is heated to 90°C and is added with Na₂CO₃ to precipitate Li₂CO₃. Then the precipitate is filtered out, washed and dried.

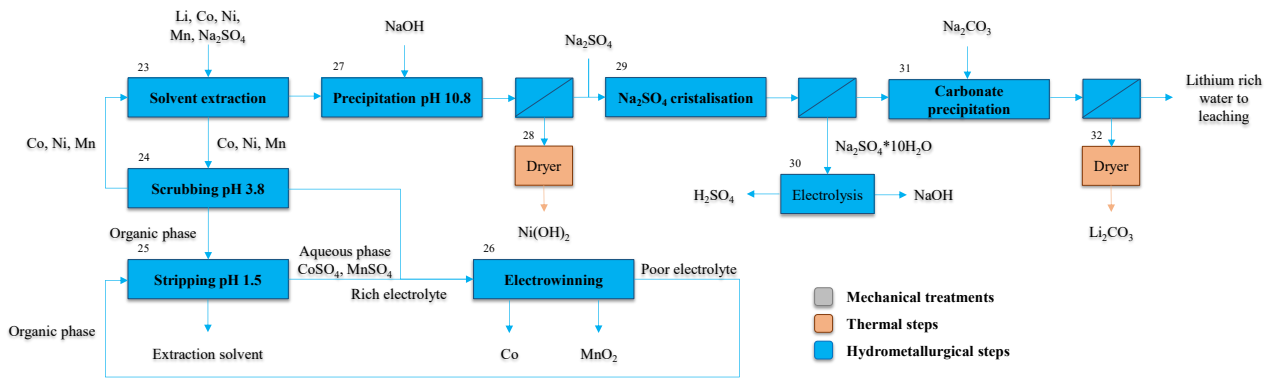


Figure S16. Lithion recycling process: hydrometallurgical section (part 2)⁴⁹.

S3. Description of recycling processes in Asia

S3.1 Sony-Sumitomo

The Sony-Sumitomo process was developed in 1996 by a cooperation between Sony Electronics and Sumitomo Metal Mining Company^{11,13,21,50–53}. It has a capacity of 150 t y⁻¹ of spent LIBs and it is composed by two main steps: battery incineration carried out at Sony plants at about 1000°C to open the batteries, followed by the cobalt extraction and recovery step, which is performed at Sumitomo plants. The incineration opens the batteries and eliminates the flammables such as electrolyte and separators. The organic solvents, lithium, and fluoride in the batteries are lost as fly ash, which is removed from the flue gas by a scrubbing system. An estimate of the minimum energy required to heat 1 ton of automotive Li-ion polymer batteries is 992 MJ. The pyrometallurgical step leads to the recovery of a Co, Ni and Fe alloy and a slag containing other components including lithium, which is not recovered. The main product of the Sumitomo–Sony process is CoO with battery-grade purity, thus the alloy is sent to Sumitomo plants to carry out hydrometallurgical processes. Optionally a thermal pre-treatment step is carried out to reduce the hazard of large format Li-ion batteries. This step is a calcination at medium-high temperature, which burns plastics and electrolyte and decomposes the conductive salt into HF, thus the off-gas must be carefully treated. Large battery packs after the thermal deactivation are disassembled to recover steel or aluminum

housing. Recently, Sumitomo announced its first practical method to recover copper by pyrometallurgy and nickel by hydrometallurgy.

S3.2 Brunp

Brunp Recycling Technology Co. Ltd. is a battery recycling company founded in 2005 in Fushan, China^{9,54-56}. Brunp facilities are located in Guangdong and Hunan provinces. In 2015 Brunp became a subsidiary of CATL. Brunp recycles mostly LIB waste materials from production scrap, but also spent LIBs from portable devices and EVs. Brunp produces over 10,000 t y⁻¹ of nickel cobalt manganese hydroxide.

The LIBs recycling process developed by Brunp is based on the co-precipitation method and mainly includes over-discharging, thermal pretreatment, mechanical treatments, and hydrometallurgy. The company hydrometallurgical treatment is based in the facility in the Hunan province. The cathode scraps including Al foil, binder, conductive carbon black and cathode active material are firstly pulverized by a vertical high-speed rotating disintegrator and fragments are sieved at mesh size of 177 μm . Then the powder undergoes a thermal treatment to decompose the PVDF binder at 600°C for 4 hours. After that Al is recovered using a selective leaching step with 0.2 mol L⁻¹ NaOH solution. The leaching occurs at 65°C and the solution is let to react for 40 min allowing the recovering of nearly 100 wt% of Al. The unleached solid containing active material is filtered out, whereas the leachate is washed several times to neutralize it and the precipitated Al is dried at 150°C for 12 hours. The active material is leached at 60°C using a solution composed by 1.1 times the stoichiometric of 4 mol L⁻¹ H₂SO₄ and 1.6 times the theoretical consumption of 50 wt% H₂O₂. The unleached carbon black additive is filtered out, whereas the solution undergoes solvent extraction to remove impurities. First iron is removed by adjusting the pH between 1 and 3 with NaOH solution and precipitating iron hydroxide. Next copper is extracted using 25% N902 solvent or a mixture of M5640 and LIX series solvents. Copper extracted in the organic phase is recovered as CuSO₄ using

H₂SO₄ as stripping agent. Then, using 20% P₂O₄, Al, Cu, Zn, Ca impurities are removed from the aqueous phase obtaining a purified solution of Co, or Ni, Co and Mn depending on the feed LIB chemistry. When the feed is NMC chemistry, the total concentration of Ni, Mn and Co is adjusted to 2 mol L⁻¹, then 2 mol L⁻¹ NaOH solution and 1 mol L⁻¹ Na₂CO₃ are added. The solution is kept at pH 8 and stirred for 5 hours, then NMC carbonate is filtered out. The solution is sent to lithium recovery. The NMC carbonate is calcined at 550°C in air atmosphere for 5 hours to produce NMC oxide, then an amount of Li carbonate or hydroxide equal to 1.05 times the stoichiometry is added and finally the mixed solid precursors are sintered at 900°C for 10 hours to obtain NMC cathodic material. Concerning LCO chemistry the purified solution is adjusted to a cobalt concentration of 1 mol L⁻¹, 1 mol L⁻¹ Na₂CO₃ is added and it is stirred for 5 hours keeping the pH at 8. The precipitated cobalt carbonate is filtered out and the remaining solution is sent to a lithium recovery step. The cobalt carbonate is calcinated at 500°C for 4 hours in air atmosphere, then a quantity of Li source equal to 1.05 times the theoretical consumption is mixed with the obtained cobalt oxide and the solid mixture is sintered at 850°C for 10 hours in air atmosphere to produce LCO cathodic material. In some cases also LMO and LFP cathodic scraps can be recycled.

S3.3 Green Eco-Manufacturer Hi-tech (GEM)

Green Eco-Manufacturer Hi-tech Co. Ltd is a recycling company founded in 2001 in Shenzhen, China ^{9,57}. GEM core business is new energy materials, in particular battery active materials, and “Urban Mines”, including electronic wastes, battery wastes, discarded cars, Ni-Co-W metal scraps and others. Since 2006 the company has started recycling batteries. GEM recovers mainly Co and Ni via a hydrometallurgical process as battery-grade Co and Ni sulphate or carbonate salts or as ultrafine elementary Co and Ni powders. Recovered materials are directly integrated by GEM in the production of cathode active materials, in particular NMC and NCA types, that the company produces with a capacity of 5000 t y⁻¹. GEM treats spent LIBs together with Ni-Co

containing wastes. The spent LIBs case is broken and then batteries are disassembled to separate anode and cathode. The cathode material is then calcined or soaked in appropriate organic solvents to remove the binder and to easily separate the active material from the aluminum current collector foil. Then the cathode is comminuted with Ni-Co containing scraps and sent to the hydrometallurgical section. The first step is leaching under acid conditions using H_2O_2 or $\text{Na}_2\text{S}_2\text{O}_3$ as reducing agent. This is followed by an impurities removal step. Then a Ni rich solution and a Co rich solution are obtained via selective solvent extraction. Finally, these metals are recovered through precipitation as salts for cathode active material production or as ultra-fine metal powder used in superalloy industry.

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