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RECENT RESEARCH RELATED TO Li-ION BATTERY RECYCLING PROCESSES – A REVIEW

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ABSTRACT. Recycling Li-ion batteries (LIBs) is a globally discussed issue in relation with waste management and environmental protection. This review reports the recent research and industrial progresses in LIBs recycling. After a brief introduction concerning the importance of LIBs recycling, and their structure and applications, the main aspects dedicated to LIBs recycling (pre-treatment, pyrometallurgical and hydrometallurgical processes) are presented. Greater attention has been paid to the hydrometallurgical processes due to their complexity but also because they can lead to the production of valuable products without a high ecological impact (compared to pyrometallurgical ones). Given that today, worldwide, only about 5% of spent LIBs are recycled, the data presented in our review can inspire the design and implementation of competitive technologies that fit into a sustainable circular economy that preserves the primary resources and ensures the environmental protection.

Keywords: *Li-ion batteries recycling, pre-treatment, pyrometallurgy, hydrometallurgy, environmental protection.*

INTRODUCTION

Spent LIBs recycling challenge

In the coming decades, the global production of LIBs will increase significantly due to the growing demand from the electric vehicles (EV) and portable electronics (PE) [1, 2]. At the same time, there is a growing concern about the supply of raw materials, especially rare metals such as Co. In this context, LIBs recycling provides crucial solutions for: (i) securing the supply

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of raw materials, (ii) compensating of price fluctuations, (iii) reducing pollution and (iv) preserving of mineral resources [3, 4]. According to literature, China is the largest market for LIBs recycling and, by 2040, more than 50% of spent LIBs (equivalent to 4.3 million tonnes) will be recycled in China. Although, at the beginning of 2020, most of the available LIBs for recycling came from consumer electronics, starting from 2025, the EV sector will significantly dominate the LIBs recycling market [5].

LIBs structure and advantages

LIBs are mainly composed of housing, anode, cathode, separator, electrolyte, and other components [6, 7]. The cathode is made of metal oxides (including Li), the anode is a porous structure of carbon (graphite) that contains Li atoms, and the electrolyte solution consists in a solvent (organic or aqueous) containing a Li salt and additives. LIBs are manufactured in different geometries according to their beneficiaries' requests.

Considering $LiCoO_2$ and graphite as typical electrode materials, as presented in Figure 1, the charging/discharging of LIBs consists in the Li oxidizing/reducing processes, simultaneously with the transport of Li^+ ions between the two electrodes. The corresponding electrochemical reactions are [5, 8, 9]:

Cathode: $LiCoO_2 \leftrightarrow Li_{1-x}\,CoO_2 + xLi^+ + xe^-$
Anode: $6C + xLi^+ + xe^- \leftrightarrow Li_xC_6$
Total reaction: $LiCoO_2 + 6C \leftrightarrow Li_{1-x}\,CoO_2 + Li_xC_6$

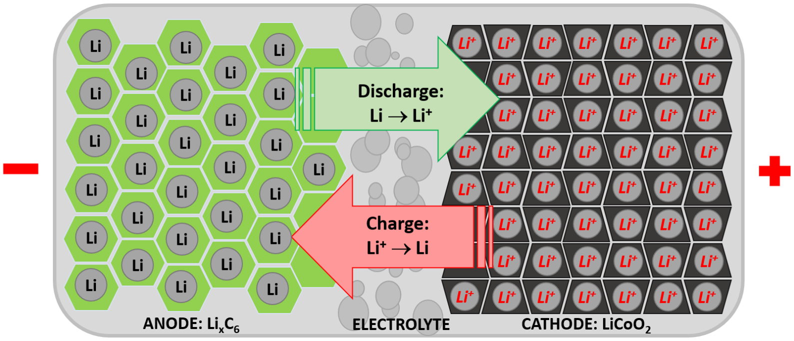

Figure 1. Schematic presentation of the LIBs charging - discharging processes

Among other aspects, the efficient energy storage is the key factor for the future of decarbonisation [9]. Due to the included components, operation mode, advantages, and their role in a sustainable future, LIBs play an important role in the energy storage, being currently the most competitive type of battery [5, 10, 11].

The efficiency and low cost of LIBs favour the development and the number increase of EVs and hybrids. The high gravimetric and volumetric energy density of LIBs and their mass production has brought the price of electric vehicles closer to the classical ones. In terms of operating costs, the price of electricity for the operation of EVs is lower than the cost of fuel for internal combustion engines [11, 12].

Types of LIBs and their recycling assessment

Due to its complexity, the cathode material represents an essential element of LIBs, and, depending on its composition, several types were developed. The most known 6 models of LIBs are presented in Table 1, along with some specific parameters.

Table 1. Types of LIBs depending on the cathode material [5, 13]

Cathode material (Battery type)	**Cost**	**Thermal stability**	**Life cycles**	**Applications**	**Safety**
$LiCoO_2$ (LCO)	High	Low 150°C	Short (500÷1000)	Mobile Phones, Cameras, Laptops	Poor
$LiMn_2O_4$ (LMO)	Low	High 250°C	Short (300÷700)	EV, power tools, medical instruments	Moderate
$LiNiMnCoO_2$ (NMC)	High	Moderate 210°C	Moderate (1000÷2000)	Power tools, e-bikes, EV, home energy storage	Moderate
$LiNiCoAlO_2$ (LCA)	Low	Moderate 150°C	Moderate (2000)	Medical devices, industrial applications, EV	Poor
$LiFePO_4$ (LFP)	High	High 270°C	Long (3000÷7000)	Starter Batteries, high current applications	Excellent
$Li_4Ti_5O_{12}$ (LTO)	Very Low	Excellent	Long	Electric powertrains, solar streetlights	Excellent

From the types of LIBs included in Table 1, LCOs, NMCs and LFPs are the most used, and, therefore, they will present the largest percentage of recycled spent LIBs. Related to these 3 types, Table 2 presents the mass percentage of the different components of LIBs, and Table 3 details the elementary mass composition of cathode materials [14].

Table 2. Composition of different types of LIBs [14]

Battery type	LIBs composition [%]						
	Al cathode current collector	Cathode active Material	Cu anode current collector	Anode active material	Electrolyte	Housing	Sepa-rator
LCO	4	41	7	18	10	17	3
NMC	7	26	17	15	10	22	3
LFP	6	25	10	13	16	27	3

Table 3. Composition of active cathode material [14]

Battery type	Cathode composition [%]						
	O	Co	Li	Mn	Ni	Fe	P
LCO	33	60	7	-	-	-	-
NMC	21	39	5	17	18	-	-
LFP	41	-	4	-	-	35	20

Given the high ratio of valuable metals in LCO and NMC cathodes and the popularity of these models in many consumer applications, their recovery will become increasingly important. The recovered materials could be used to produce new batteries, reducing the manufacturing costs. At present, these materials represent more than half from the batteries cost [15].

The price of Co reached the highest level in 2018, due to its increased demand for EVs production. The growth appeared because the cars' manufacturers tried to secure the long-term supplies of Co to be used in LIBs [16, 17]. The concentrations of valuable metals in LIBs (Li, Co, Ni, Mn) can exceed their concentrations in the natural ores. If these metals can be widely recovered from spent LIBs in a more economical way than the extraction from natural ores, the price of PE, and EVs should decrease [15]. In Table 4, the estimated values of the recycled metals from LIBs are presented.

Table 4. Estimated values of the recycled metals from LIBs [18]

Battery type	Price/ton of spent LIBs [USD]							
	Co	Li	Mn	Ni	Fe	Cu	Al	Total
LCO	20100	473	-	-	-	472	72	21117
NMC	13065	338	153	2698	-	1146	126	17526
LFP	-	270	-	-	43	674	108	1095

As it can be seen from Table 4, the recycling of LCO LIBs is the most advantageous due to the high Co value, followed by NMC batteries. At opposite, the LFP batteries recycling is economically unattractive due to the lack of valuable metals.

Assuming a LIBs collection rate of 65% and a Li recycling efficiency of 57%, the value of the recovered materials (Co, Ni, Al and Li) could reach $ 408 million in 2030, contributing to the preservation of these materials into EU economy and the creation of 2,618 jobs [19]. In terms of the LIBs recycling contribution to the supply chain security, it could reduce the globally demand for both Li and Co from ores [20-22]. Countries with the highest battery recycling rates include Belgium, Finland, France, the UK, the USA, Canada, and China. Most of these countries are based on European battery recycling initiatives [8]. Among the listed countries, the relevant common features of the recycling systems include: (i) collection and recycling systems with clear objectives; (ii) development of visually attractive containers for collecting of spent batteries; (iii) involvement of producers and traders in the collection process; (iv) efficient transport regulations for LIBs; (v) partial taxation of the producers and traders for the recycling process; (vi) application of hydrometallurgical treatments for metal refining; (vii) green tax. Incorporating these features is essential to the success of LIBs recycling systems.

LIBs RECYCLING PROCESSES

The waste management of LIBs includes a series of steps, starting with collection, sorting, and transport of the waste, followed by recycling using pre-treating, and pyrometallurgical (PMP) and/or hydrometallurgical (HMP) processes [23].

Pre-treating processes

The pre-treating processes (PP) involved in the LIBs waste recycling are based on the physical properties of the materials (density, magnetic susceptibility, electrical conductivity, etc.) and they include “Discharging-Shredding-Crushing-Sieving-Separation” [24]. The products resulting from PP are metallic and non-metallic fractions. The metallic fraction represents a mixture of various metal: Mn, Li, Co, Ni, Al, Cu, etc. [25, 26].

In addition to the previous invoked phases, PP may also include drying steps [27-29]. Most of the spent LIBs PP includes a first discharging step [30-33] by immersion in a solution of NaCl 5% and then drying [34]. The cells are discharged or short-circuited to reduce the electric-shock hazard, to recover the remaining energy, and allow safe disassembly and grinding [35-37].

After the complete discharge, LIBs are disassembled manually or mechanically. Depending on the type of the recycled LIBs, the obtained parts after disassembling can be divided into metal or plastic housings, cables, battery management units, anode, cathode, and organic compounds [38]. The housing and organic membranes can be treated in a centralized mode due to their high purity and value in the market [39]. The active anode and cathode materials represent a mixture of oxides ($LiCoO_2$, $LiNi_xCo_yMn_zO_2$, $LiMn_2O_4$, $LiNiO_2$ and $LiPePO_4$), a small amount of conductive agent, and a polyvinylidene fluoride binder (PVDF). Due to the complex structure and composition of those materials, the recovery of the cathode material has become the most studied in this field [14, 39-45].

The separation of cathodic and anodic materials from the Al or Cu foils represents the first step to recover the electrode materials. The bonding strength between graphite and Cu foil is relatively weak, making it easy to break. After the battery disassembling, some graphite can be separated from the Cu foil and recycled. The industrial recovery of waste graphite is necessary, but the existing methods for graphite recycling are not feasible for large-scale industrial processes due to excessively high costs [29]. The cathode material is more difficult to separate due to the strong adhesion onto the Al foil substrate and need to follow a separation process [46, 47].

The mechanical separation includes the shredding, drying, and sieving of the electrode materials to obtain a powder fraction (black mass), followed by hydraulic (flotation), magnetic or pneumatic separation.

The drying step aims to remove the electrolyte. At temperatures above 100 °C and low pressure, the organic electrolytes are vaporized. The organic solvents from the electrolyte present in LIBs are a mixture of carbonates such dimethyl-, ethyl-, methyl-, diethyl-, propylene-, and ethylene-carbonate. The vapours can be recovered by condensation or burned for energy recovery [48]. Elimination of the electrolyte leads to an improved separation process and minimizes the possibility of obtaining impure products. Sieving process allows to separate the rests of plastic, paper, Cu, Al from the black mass, leading to a fraction that contains only valuable materials like Li, Ni, Co and Mn, that can be recovered in a further process.

Pyrometallurgical process

The pyrometallurgical process (PMP), representing a branch of extractive metallurgy or metals recycling from spent LIBs, includes stages of incineration, melting in electric arc or plasma discharge furnaces, sintering, and gas phase reactions at high temperatures [49-51]. For LIBs recycling using PMP, the waste is heated up to 1000°C or more [52], to recover the Li salt and other materials from the electrodes [53, 54].

For LIBs recycling, the physical-chemical properties of the batteries are crucial for the choice of PMP conditions because, unlike the ore, LIBs contain various materials such as plastic, metals, oxide, carbon, salts, and organic solvents. Some of them are volatile, flammable, sensitive to water, toxic and unstable at high temperatures [55].

PMP for recycling of spent LIBs involves pyrolysis. The electrolyte solvents are first removed [56, 57] at temperatures above boiling point, of approx. +250 ° C. In spent LIBs is also present the carbon (graphite) which, in mixture with the active materials of the electrodes, can act in PMP as a reducing agent or is burned in the calcination stage. The binders used in LIBs for the preparation of electronic materials (for example, PVDF) can generate CO_2, PF_5, HF etc. [51, 58, 59] therefore for these toxic gases must be ensured their capture and treatment to avoid air contamination [59]. On the other hand, binders may be dissolved in an organic solvent [60].

Sometimes, the removal of organic materials and graphite from spent LIBs is done by roasting [61], optionally with the addition of inorganic salts [40, 62]. After that, most residual materials are Li metal oxides and some metal scraps from current collectors [63]. Separation of Li metal oxides is possible at high temperatures, having the disadvantage of high-energy consumption. Separation of Li_2O and metal oxides encounters difficulties in the extraction process [63]. Mn oxides can be reduced to MnO with C, CO and H_2, but it is difficult. Al can reduce $LiCoO_2$ to generate Co and $LiAlO_2$, but Li is difficult to separate from $LiAlO_2$ [51].

Another PMP for treating spent LIBs is the reductive melting (RT) [64] in furnace using reducing agents as carbon, CO, and natural gas. After RT, the heavier liquid metals accumulate in the lower part of the furnace and the Li oxide remains in the upper layer of the slag. Impurities present in the two molten phases can be removed by typical refining treatments such as metal-slag, metal-metal, and metal-gas phases [40, 65]. Due to their high economic values, the most recovered metals by PMP are Co, Li and Ni. For example, Li et al. [66] managed through PMP to recover 95.72% of Co and 98.93% of Co, and Guoxing et al. [67] recovered, by pyrolysis, a Co-Ni-Cu alloy with recovery degrees of 81.6%, 93.3% and 90.7% for Co, Ni and Cu, respectively, but also Li in slag in proportion of 82.4%.

Worldwide, the companies are applying mainly PMPs for recycling waste LIBs [51, 68]. The process developed by Accurec GmbH use high temperature furnaces to melt pre-briquetted cathodic materials, resulting a Co-Mn alloy and a Li containing slag [55]. The Sony/Sumitomo process includes the burning of flammable components and the magnetic separation of metals, which are then processed hydrometallurgically [55]. In the Umicore process, the flammable components are burned firstly, and the metals are melted to generate Ni, Co, Cu, and Fe alloys, which are subsequently separated hydrometallurgically [51].

Due to the high costs of recycling by PMP, the companies that use this technique do not recover all materials from LIBs [63-65]. The main disadvantages of PMP are high-energy consumption and the generation of highly toxic and volatile products. Due to high environmental hazard, the resulting gases must be carefully treated [62].

Hydrometallurgical process

The hydrometallurgical process (HMP) is based on the: (i) leaching of the grinded spent LIBs in acidic or alkaline aqueous solutions, (ii) possibly purification of leaching solutions, (iii) separation of valuable components by solvent extraction, chemical precipitation, electrodeposition, etc., and (iv) treatment/regeneration of the solutions to be recirculated in process [69, 70].

Leaching process of the metals from spent LIBs

Leaching aims the transfer of metals, oxides and salts that are part of the anode and cathode materials of LIBs into a liquid phase. The leaching step of spent LIBs is done after the removing of some battery components. This approach helps to minimize the volume of aqueous leaching solutions and the number of the subsequent purification and separation steps.

The rate and degree of the metals recycling from LIBs depend on the leaching efficiency and rate [70], which are influenced by the used leaching reagent, concentration of chemicals, solid/liquid ratio (S/L), temperature, duration, ultrasound support, mechanical agitation, and other factors [24]. In this context, in Table 5, we present summarized literature data concerning the specific leaching parameters for different types of LIBs. As it can be seen, the researchers use widely mineral acids (H_2SO_4, HCl, HNO_3 and H_3PO_4 [71 -74]), but also organic acids, as citric acid ($C_6H_8O_7$), oxalic acid ($C_2H_2O_4$), lactic acid ($C_3H_6O_3$), malic acid ($C_4H_6O_5$), tartaric acid ($C_4H_6O_6$) and aspartic acid ($C_4H_7NO_4$) [75-79]. To accelerate the leaching process of metal compounds, various reducing agents have been used, as H_2O_2 [80], Na_2SO_3 [81], $NaHSO_3$ [82], and carbohydrates, including D-glucose [83] and ascorbic acid ($C_6H_8O_6$) [84]. The reducing agents are added to convert Co^{3+} to Co^{2+} and Mn^{4+} to Mn^{2+}, which are easier to be dissolved in the solutions [85].

The acidic leaching has been shown to be highly efficient [69, 77, 78] even more efficient than the alkaline one [86].

Table 5. Leaching specific parameters for different types of spent LIBs

Battery type	Leaching agents	S/L ratio [g/L]	Temp. [°C]	Time [min]	Leached metals / Leaching rate [%]	Ref.
Mixed	0.5 N $C_6H_8O_6$	10	65	90	Co: 52.19; Li: 59.23; Mn: 57.98	[76]
	1.5 M H_2SO_4 30% H_2O_2 5 g $C_5H_8O_4$	10	90	120	Co: 87.85; Li: 99.91; Ni: 91.46	[77]
	2.0 M H_2SO_4 4.0% H_2O_2	10	70	180	Al: 97.8; Cu: 64.7; Co: 99.6; Li: 98.8; Ni: 99.6; Mn: 97.4	[80]
	0.5 M H_2SO_4 0.1 M Na_2SO_3	20	120	360	Ni: 93.11; Co: 92.84; Mn: 90.18	[81]
	1 M H_2SO_4 0.075 M $NaHSO_3$	50	95	240	Li: 91.6; Co: 96.4; Ni: 87.9; Mn: 83.69	[82]
	1M H_2SO_4	20	95	240	Li: 96.7; Co: 91.6; Ni: 96.4; Mn: 87.9	[87]
	1M H_2SO_4	50	95	240	Li: 93.4; Co: 66.2; Ni: 96.3; Mn: 5.02	[88]
	15% H_2O_2 0.5 M $C_6H_8O_7$	20	90	60	Li: 99.1; Co: 99.8; Ni: 98.7; Mn: 95.2	[89]
	28% NH_3	10	80	120	Co: 100; Cu: 100; Ni: 100	[90]
	1 M H_2SO_4 0.5 M HNO_3	25	75	60	Li: 93.2; Co: 90.5; Ni: 82.8; Mn: 77.7	[91]
	2M H_2SO_4	240	20-23	500	Co: >96; Ni: >96; Mn: >96; Li: >96	[92]
	2 M H_2SO_4 6 % H_2O_2	10	75	90	Co: 100	[93]
	98% H_2SO_4 78% $Fe_2(SO_4)_3$	240	90	60	Ni: 97.09; Co: 97.65; Mn: 96.88; Li: 98.32; P: 0.21	[94]
NMC	0.5 M $C_6H_8O_6$ 1M HNO_3	20	85	10	Li: ~100; Mn: ~100; Co: ~100; Ni: ~100	[41]
	4 M NH3 1 M $(NH_4)_2CO_3$ 0.3 M Na_2SO_3	50	80	300	Li: 79.1; Co: 86.4; Ni: 85.3	[95]
LCO	2M HCl	50	60–80	90	Li: ~100; Co: ~100	[72]
	1.5 M H_3PO_4 0.02 M $C_6H_{12}O_6$	50	80	120	Li: 100; Co: 98	[74]
	1 M $C_6H_8O_7$	20	80	60	Co: 90	[75]

Battery type	Leaching agents	S/L ratio [g/L]	Temp. [°C]	Time [min]	Leached metals / Leaching rate [%]	Ref.
	2 M H_2SO_4 4 M HCl 10% H_2O_2 10% $C_5H_8O_4$	20	80	300	Co: 100; Li: 95.1; Ni: 100	[78]
	1.5 M $C_4H_4O_4$ 0.5% H_2O_2	20	70	20	Ni: 98.27; Co : 98.06; Mn : 98.54; Li: 95.74	[79]
	1M $C_6H_8O_7$ 0.2 M $C_6H_8O_6$	20	80	360	Co: 80; Li: 100	[84]
	1 M $C_4H_4O_4$ 0.3 M $SnCl_2$	20	60	40	Li: 98.67; Co: 97.5	[96]
	1.5 M H_3Cit 6% H_2O_2	15	90	90	Co: 99.5; Li: 97	[97]
	2 M H_3Cit, H_2O_2	50	70	80	Co: 98; Li: 99	[98]
	0.9% H_2O_2	20	90	35	Li: 90; Co: 80	[99]
	1.5 M $C_6H_8O_7$ 2% H_2O_2	20	20	95	Al: 93; Co: 90; Li: 96; Mn: 94; Ni: 94	[100]
	99.8% Na_2CO_3	60	80	60	Li: 92.82	[101]
	2M H_2SO_4 10% H_2O_2	33	70	120	Co: 98.5; Li: 99.8; Ni: 98.6	[102]
LFP	0.3 M H_2SO_4 2 M H_2O_2	100	60	120	Li: 96.85	[103]
LCA	4 M HCl	5	90	1080	Li: 100; Ni: 100; Co: 100; Al: 100	[104]

Data from Table 5 indicates that the mineral acids (2M H_2SO_4 and 4M HCl) are the most efficient for solubilizing metals from LIBs, and the presence of hydrogen peroxide increases their efficiency [77-78, 95, 97,104]. However, toxic gases such as Cl_2, SO_3 and NO_x are released in the process of extracting metal ions with mineral acids, determining serious threats to environment [79]. Generally, the leaching temperature is between +60°C and +95°C, and the leaching rate of the targeted metals is close to 100%.

Compared to inorganic acids, the organic ones are considered green leaching agents, which could effectively recover the precious metals, while avoiding secondary contamination [75-76, 98, 105]. Compared to acid leaching, the dissolution of the cathodic materials in alkaline solutions have been rarely reported. However, the alkaline leaching with an ammonia-based system is relatively selective for specific elements, such as Ni, Co, and Li, due to the formation of stable complexes [29, 105].

From an economic point of view, HCl proved to be the most effective due to its relatively strong reductive capacity, assuring the complete dissolution of the cathodic material without the addition of additives [105].

Recovery of metals from the leaching solutions of spent LIBs

After the metals leaching from the spent LIBs, they can be separated from the solution in form of pure metals or chemical compounds by solvent extraction (SE), chemical precipitation (CPP), electrodialysis (EDy), and electrodeposition (EDP). In this context, Table 6 summarizes the efficient HMP for the recovering of the metals/compounds from the leaching solutions of grinded spent LIBs.

SE is based on the different solubility of metal compounds in two immiscible solvents, usually water and an organic solvent dopped with a selective extractant. At the equilibrium of the extraction system, each liquid phase contains different concentrations of metal ions, assuring they separation. By mixing the charged organic phase with a stripping solution, such as H_2SO_4, the extractant can be regenerated and reused [105,106]. The SE efficiency is influenced by the extraction yield and the phase separation capacity. This method has been widely used in various HMP and has proven to be an effective method for separating valuable metal ions or removing impurities from the aqueous leaching solution. The commercially available selective extractants such as Cyanex 272, D2EHPA or PC88A have been widely used to separate the metal ions from the leaching solutions of spent LIBs [93]. Extraction with PC88A and Cyanex 272 is very efficient in separating Co against other metals from the leaching solution [106, 107], and D2EHPA has been studied for the separation of Mn [106, 108].

The most used technique for the metals recovering from LIBs leaching solutions is CPP, based on the different solubility of metal compounds relative to pH. Lithium hydroxide and oxalate are much more soluble than similar compounds of other metals. Thus, Fe^{3+}, Al^{3+} and Cu^{2s+} ions precipitate at a relatively lower pH [109, 110]. For the Li precipitation, the two commonly used chemical agents are Na_2CO_3 [27, 111-113] and Na_3PO_4 [98, 114, 115], resulting high purity Li salts. As examples, Natarajan et al. [116] obtained 99.7% purity Li_2CO_3 and Chen et al. [117] precipitated Li_3PO_4 with a purity of 93%. For the precipitation of transition metals, $H_2C_2O_4$ is widely used [99, 117], and Mn can be also precipitated as hydroxides [94].

EDP has the advantage of obtaining high purity products due to the small amount of impurities included in deposits, but has a relatively high electricity consumption. For example, Quintero-Almanza et al. [93] obtained Co with a purity of 99%. In electrodeposition, pH is a very important factor that can strongly influence the energy efficiency. The pH variations can be minimized by addition of H_3BO_3 in solutions, determining higher current efficiencies [93, 118, 119]. Co oxide obtained by electrodeposition can be used as material for supercapacitors [105].

Table 6. HMP for the separation and recovery of products from the cathodic materials of spent LIBs

Battery type	Recovery method	Additives	Recovery products/ Recovery rate [%]	Ref.
NMC	CPP	Na_2CO_3, EDTA	Li_2CO_3: 99; Co_3O_4: 98	[27]
		NaOH	Li_3PO_4: 88; CoC_2O_4: 98.7	[74]
		$H_2C_2O_4$	CoC_2O_4: 80; Li_3PO_4: 90	[99]
		$H_2C_2O_4$; H_3PO_4	$CoC_2O_4 \cdot 2H_2O$: 99; Li_3PO_4: 93	[117]
		$C_2H_2O_4$	CoC_2O_4: 98	[123]
		$C_4H_6O_6$	$C_4H_4O_6Co$: 98	[124]
		Na_2CO_3	Li_2CO_3: 99.93	[125]
		Na_3PO_4	$Al(OH)_3$: 90~95	[126]
Mixture	CPP	Na_2CO_3	Li_2CO_3: 99.4	[112]
		Na_2CO_3 $(NH_4)_2S$	Li_2CO_3: 99.7; Co_xS_y: 99; $MnCO_3$: 98.7	[116]
		$H_2SO_4 + H_2O_2$	Co: 100; Ni: 100; Mn: 100	[127]
LCO	EDy / CPP	Na_2CO_3	Li_2CO_3: 88.3	[113]
Mixture	SE / CPP	D2EHPA, Cyanex 272, DMG, Na_2CO_3	$Co(OH)_2$: ~99.5; $Ni(OH)_2$: ~99.5; $Mn(OH)_2$: 90; Li2CO3: ~99.5	[108]
NMC	SE / CPP	D2EPHA, Cyanex 272, NaOH	$Co(OH)_2$: 95	[128]
LCO	SE / CPP	$Na_2S_2O_5$ Na_2CO_3	Co: 60-70; Ni: 60-70; Mn: 60-70; Li_2CO_3: 60; Al: 81; Cu: 91; Fe: 98	[129]
		Acorga M5640, Cyanex 272, Na_2CO_3, $(NH_4)_2C_2O_4$	Li_2CO_3: 80; $CuSO_4$: 98; $CoC_2O_4 \cdot 2H_2O$: 90	[130]
Mixture	SE	D2EHPA TBP	Fe: 98.5; Co: 99.8; Mn: 99.8; Al: 99.7; Cu: 97.8; Ni: 98.6	[116]
		D2EHPA	Mn: 84; Co: 8.6; Ni: 6.2	[119]
LCO	SE	D2EHPA	Mn : 100; Co: 94; Ni: 90	[45]
NMC	SE	D2EHPA, $MnSO_4$	Mn: 84	[104]
NMC	SE	D2EHPA PC88A	Li_2CO_3: 84.7; $NiSO_4$: 99; $CoSO_4$: 99; $MnSO_4$: 99	[107]
LFP	CPP	Na_3PO_4	Li_3PO_4: 95.56	[100]
		NaOH, Na_3PO_4	LiOH: 99.9; $FePO_4$: 99.97	[114]
		Na_2CO_3	Li_2CO_3: 99	[130]
Mixture	SE / EDP	Cyanex 272	Co: 97-99	[93]
Mixture	EDy	EDTA	Ni: 99.3; Co: 87.3; Li: 99	[110]

Recently, the regeneration technique for the recycling of the cathodic materials has become a new direction that refers to the resynthesis of the materials directly from the leaching solutions. This method, although sophisticated, has proven to be energy efficient as it is integrated into the synthesis of the cathodic materials [105]. The sol-gel and co-precipitation methods are two main ways of regenerating the cathodic materials. Before regeneration, the content of each metal ion must be accurately analysed and then adjusted to the desired value by the addition of certain chemical compounds [120, 121, 122].

The leaching solution of organic acids is usually removed by the sol-gel method because the existing organic products can serve as chelates. After the addition of acetic salts or nitrates to regulate the ratio of metal ions, sols are obtained by the increasing of the solution pH and the gel by drying. The cathodic materials are synthesized by calcination of the gel [131].

In cases of co-precipitation, sulphates are used to adjust the ratio of metal ions when the spent cathodic material is leached into H_2SO_4 [132].

By adding NaOH or NH_3 to increase the pH of the leaching solutions, the transition metal hydroxides (precursors) could be co-precipitated. Then, by calcination of the stoichiometric mixture of Li_2CO_3 and precursors, resynthesized cathodic materials are obtained [133].

INDUSTRIAL RECYCLING TECHNOLOGIES FOR SPENT LIBs

The industrial recycling technologies (IRTs) of the spent LIBs focus mainly on the recovering of valuable or critical metals such as Co, Cu, Li, Mn, and Ni [8]. Most often, the IRTs are based on the above-mentioned processes (PMP and HMP), combined with mechanical (MP) and thermal (TP) processing. For the recovery of valuable materials, these technologies are used successively [41], as follows: (i) Direct PMP, with optional HMP pre-sorting and refining; (ii) Thermal pre-treatment by pyrolysis, with MP processing in several stages and PMP or HMP refining; (iii) Treatment at low temperatures, by electrical and/or cryogenic depollution, with multi-stage processing and HMP refining.

From the quantitative point of view, only 5% of spent LIBs have been reported as recycled by 2019. However, several institutions, as Circular Energy Storage [134], claim that this value is inaccurate and that the overall recycling rate is around 50%. The recycling potential in Asia and Europe is far ahead of the other continents. According to recent data [135], China and Europe's recycling capacity accounts for about 80% of the spent LIBs world's recycling. The manufacture of LIBs is largely concentrated in China, South

Korea, and Japan. Consequently, their recycling is also highly developed in Asia due to the large amounts of spent LIBs generated in this region [136, 137]. American and European recycling industries show a wide variety of technologies, although the available volume of spent LIBs is too small for a profitable operation [85, 138].

In this context, representative examples of LIBs recycling companies around the world and the used processes are presented in Table 7.

Table 7. Worldwide representative examples of spent LIBs recycling companies

Country	Company	Processes	Capacity [t/year]	Ref.
ASIA				
Japan	DOWA Eco-Systems Co. Ltd.	TP + PMP + HMP	>1000	[139, 135]
	KYOEI Steel	PMP	>1000	[139]
	Nippon Recycle Center Corp	PMP	<1000	[140]
	Sony Corp. & Sumitomo Metals and Mining Co	PMP	150	[135]
	4R Energy Corp	PMP + HMP	n.d.	[68]
	Mitsubishi	PMP	n.d.	[135]
Indonesia	DOWA Eco-Systems Co. Ltd	TP + PMP + HMP	n.d.	[139]
China	DOWA Eco-Systems Co. Ltd.	TP + PMP + HMP	n.d.	[139]
	Anhua Taisen Recycling Technology Co. Ltd.	MP + HMP	> 5000	[140]
	Fuoshan Bangpu Ni/Co High-Tech Co	n.d.	n.d.	[44]
	GHTECH	n.d.	n.d.	[68]
	Highpower International Inc	MP + PMP + HMP	> 1000	[139]
	Huayou Co New Material Co Ltd	MP + HMP	> 1000	[139]
	JX Nippon Mining and Metals Co.	TP + MP + HMP	<1000	[139]
	Shenzhan BAK Battery Co	Disassembly	n.d.	[68]
	Shenzhen Green Eco Manufacturer Hi-Tech. Co., Ltd.	MP + HMP	<5000	[42]
	Shenzhen Tele Battery Recycling Co	HMP	3000	[68, 139]
	Soundon New Energy Tech. Co. Ltd.	n.d.	n.d.	[68]
	TES-AMM	MP (inert gas)	<1000	[139]

Country	Company	Processes	Capacity [t/year]	Ref.
Singapore	DOWA Eco-Systems Co. Ltd	TP + PMP + HMP	n.d.	[45]
	JX Nippon Mining & Metals Co	TP + MP + HMP	<1000	[139]
Hong Kong	GHTECH	n.d.	n.d.	[68]
	Highpower International Inc	MP + PMP + HMP	> 1000	[139]
Thailand	DOWA Eco-Systems Co. Ltd	TP + PMP + HMP	n.d.	[139]
South Korea	SungEel Hitech Ltd	MP + (T +) HMP	>1000	[139]
	JX Nippon Mining and Metals Co.	TP + MP + HMP	<1000	[139]
	SK Innovation Co	n.d.	n.d.	[68]
EUROPE				
Spain	Pilagest	MP + HMP	<1000	[68]
Belgium	REVATECH	n.d.	n.d.	[68]
	Umicore	PMP + HMP	>1000	[139]
France	AFE Group (Valdi)/ ERAMET	PMP	n.d.	[68]
	Euro Dieuze Industrie/ SARP	HMP	200	[135]
	Recupyl S.A.S	HMP	110	[135]
	S.N.A.M. AFE Group (Valdi)/ ERAMET	MP + (TP) + HMP	n.d.	[68]
Germany	Accurec Recycling GmbH	TP + MP + PMP + HMP	>1000	[139]
	AERC Recycling Solutions	PMP	n.d.	[68]
	Brunp Recycling Technology Co	TP + MP + HMP	3000	[139]
	Chemetall	n.d.	n.d.	[68]
	DK Recycling und Roheisen GmbH	PMP	n.d.	[45]
	Düsenfeld GmbH	MP + PMP	n.d.	[68]
	NIHÜTTE AUE GMBH	TP + PMP + HMP	>1000	[139]
	GRS Batterien	PMP		[68]
	PROMESA GmbH & Co. KG	MP (aqueous shred) + unknown	<1000	[139]
	JX Nippon Mining and Metals Co	TP + MP + HMP	<1000	[139]
	Primobius	PMP + HMP	n.d.	[141]
	REDUX GmbH	TP + MP + unknown	<1000	[139]
UK	AEA Technology Batteries	HMP	n.d.	[135]
	Cawleys	n.d.	n.d.	[68]
	Ecobat Technologies Ltd	PMP + HMP	n.d.	[68]
	Metal-Tech Ltd.	n.d.	n.d.	[68]

Country	**Company**	**Processes**	**Capacity [t/year]**	**Ref.**
Sweden	SAFT. AB	PMP	n.d.	[68]
	Erasteel Kloster AB Vikmanshyttan	n.d.	n.d.	[68]
Finland	AkkuSer	MP	4000	[135]
Switzerland	Batrec Industrie AG	MP +HMP + TP + PMP	200	[135, 68]
Norway	Xstrata/ Glencore	PMP + HMP	7000	[135]
NORTH AMERICA				
Canada	Earthtech	Disassembly	n.d.	[135]
	Lithion Recycling	HMP	n.d.	[68]
	Li-Cycle	MP + HMP	5000	[65]
USA	American Mn	HMP	n.d.	[68]
	Battery Resourcers LLC	n.d.	n.d.	[68]
	Battery Safety Solutions	Collection + Discharge + Disassembly	n.d.	[68]
	Euro Dieuze Industrie/ SARP	Disassembly+ PMP + HMP	>5000	[68, 139]
	Inmetco	PMP	6000	[135]
	OnTo Technology Oregon US	Direct recycling	n.d.	[135]
	Salesco Systems	PMP	n.d.	[68]
	Toxco/ Retriev Tech.	MP (aqueous shred) + HMP	n.d.	[139]
	DOWA Eco-Systems Co. Ltd	TP + PMP + HMP	n.d.	[139]
	JX Nippon Mining and Metals Co.	TP + MP + HMP	<1 000	[139]
AUSTRALIA				
West Perth	Neometals Ltd	MP +HMP	Lab Scale	[135]

(n.d. = no data)

As it can be seen from Table 7, most companies around the world mainly use PMPs to recycling the spent LIBs. Overall, recycling efficiency decreases if it is operated at high temperatures due to the decomposition of organic components [141]. On the other hand, HMP can recover a greater number of materials.

Another important aspect of companies is the maximum focus on the supply chain with spent LIBs and on the key stakeholders that are important for the supply of spent LIBs. The feed profile of spent LIBs must be central into the process development. The inherently heterogeneous nature of spent LIBs must be closely considered as part of any recycling process [142, 143].

Generally, the recycling companies recover Co, Cu, Li and Mn due to their high value, but, also, Fe, Ni and Al are often recovered. Even if Fe is the lowest value component from the battery, its extraction process is simplistic, as it can be magnetically separated if MPs are used in the first stage of the recycling process.

The MP steps allow the recovery of the solid parts, such as materials from the housing, modules and cells, electrical conductors, and steel screws. Zigzag sieving allows the separation of materials according to their density. A vibrating sieve can be also used to separate the electrode black mass from the separator and the current collector foils. Finally, the composition of the obtained fractions is determined after the manual or automatic screening and sorting. The fragments from the modules can be dried at temperatures between 100-140 °C [144] to remove the electrolyte, and the evaporated solvents can be recovered by condensation.

To support the efficient recycling of LIBs, state-of-the-art technologies are recommended to improve the flexibility and recovery rate of all materials from the spent devices. These technologies must consider a combination of safe and ecological MP and HMP with low costs, able to recycle all types and models of spent LIBs. As example, a combined process for the spent LIBs recycling was successfully implemented by the Li-Cycle ® Canadian company [142], based on a safe MP size reduction and dedicated HMP, allowing a recovery rate of the recycled materials up to 100%.

Concerning the economic aspects of the IRTs, American Manganese Company Inc. [145] presents a business plan developed to recover valuable metals from spent LIBs cathodes, using combined MP and HMP. The study shows that, at a 3 tons/day processing capacity of NMC type spent LIBs, with a recovery rate of 95%, and working 350 days/year, annual revenues of \$14.3 million can be obtained at production costs of \$7.9 million. We can conclude that the processing of Li batteries can generate generous profits, of \$6.4 million in this case. Implementation of the combined technology processes could also: (i) to remove Li-ion batteries waste from landfills; (ii) to diminish the manufacturing costs by the materials recycling from spent LIBs and reusing them in the manufacturing process of batteries; (iii) to significantly reduce the CO_2 emissions.

In the future, the recycling of spent LIBs is expected to be one of the main sources of Li supply. Unlike oil, where the price fluctuations only lead to the increased car operating costs, the potential Li price fluctuations will have an impact on the total purchase price of the car. Therefore, the recycling will be one of the means of mitigating the amplitude of the possible fluctuations due to the geo-political or other barriers.

CONCLUSIONS AND PERSPECTIVES

Worldwide, the lithium-ion battery will be for at least medium term, the most important and widely used electrochemical source of energy. However, the exponential increase in the production of LIBs also generates a huge amount of scrap. Their recycling is necessary for at least two reasons: (i) to avoid pollution and (ii) to reintroduce valuable and deficient materials (on the world market) from these batteries into the economic circuit [10].

The research on the spent LIBs recycling has been mainly focused on the recovery of metals from the cathodic material, especially Co, which is deficient on the world market. As the volume of spent LIBs increase, the interest in the complete valorisation of all materials also increase. Thus, more recent research is focused on the recovery of other metals such as Mn, Ni and, of course, Li.

A promising direction is the recent research aimed to reintroduce (direct recycling routes) in batteries the recovered cathodic material after a minimal processing [4, 146,147].

There are also concerns to develop processes for all-components recycling from the spent LIBs, based mainly on the: (i) discharging of spent LIBs, (ii) dismantling and classification of components, (iii) separation of electrode materials, and (iv) refining and value-adding technologies for the cathodic materials and graphite [148]. Future recycling strategies will need to be redefined so they can adapt to the large diversity of the spent LIBs and capitalize as many of their recoverable components.

The achieving of a reasonable economic profitability for the spent LIBs recycling is also conditioned by the responsibility of manufacturers to develop and implement innovative processes in the factory design to make the batteries more capable to fit into the circular economy [149].

For the future, to facilitate and make more efficient the recycling of the spent LIBs, it is essential their selectively collection (SC), followed by the robotization of the physical processes (RPP) of dismantling and sorting to minimize the need for labor. The SC and RPP processes can be significantly facilitated by intelligent and standardized labeling, which will allow the facile identification of spent LIBs from type, composition, lifetime, etc. Further, the spent LIBs pretreatments at low temperature and pressure, in inert atmosphere, will diminish the pollution with toxic products, as HF, PF_5, etc.

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