

Research article

Lithium-ion batteries towards circular economy: A literature review of opportunities and issues of recycling treatments

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ABSTRACT

Nowadays, Lithium-ion batteries are widely used in advanced technological devices and Electric and Hybrid Vehicles, due to their high energy density for weight, reduced memory effect and significant number of supported charging/discharging cycles. As a consequence, the production and the use of Lithium-ion batteries will continuously increase in the near future, focusing the global attention on their End-of-Life management. Unfortunately, wasted Lithium-ion batteries treatments are still under development, far from the optimization of recycling processes and technologies, and currently recycling represents the only alternative for the social, economic and environmental sustainability of this market, able to minimize toxicity of End-of-Life products, to create a monetary gain and to lead to the independence from foreign resources or critical materials. This paper analyses the current alternatives for the recycling of Lithium-ion batteries, specifically focusing on available procedures for batteries securing and discharging, mechanical pre-treatments and materials recovery processes (i.e. pyro- and hydrometallurgical), and it highlights the pros and cons of treatments in terms of energy consumption, recovery efficiency and safety issues. Target metals (e.g. Cobalt, Nickel and Lithium) are listed and prioritized, and the economic advantage deriving by the material recovery is outlined. An in-depth literature review was conducted, analysing the existing industrial processes, to show the on-going technological solutions proposed by research projects and industrial developments, comparing best results and open issues and criticalities.

1. Introduction

A battery is a portable electro-chemical device able to convert the stored chemical energy into electrical energy with high efficiency and no gaseous emissions (Scrosati and Sun, 2011). Based on this concept, Lithium-ion batteries (LIBs) were preliminarily developed by Armand in the late 1970s, but the first commercialized cells appeared in 1991 by Sony, after countless researches on electrode materials, safety issues, economically sustainable processes and performances optimization (Blomgren, 2017). The typical composition of LIBs, net of the variability due to different manufacturers, is reported in Table 1 and consists of two

electrodes wound by lamination to a polymeric separator and impregnated by a suitable electrolyte, allowing ionic conductivity of Li ions (Li^+) (Xu et al., 2008).

During the use of LIBs, the Li^+ generated through reversible reactions at the negative electrode, move towards the cathode, where they combine to form metal oxides (Fig. 1). Vice versa, during the charging mechanism, an external power supply provides electrons that combine with Li^+ to form metallic Lithium (Li), stored between anodic graphite layers through intercalation mechanisms (Chen et al., 2018). Due to the intrinsic properties of materials, LIBs operate between 1.5 and 4.2V: a lower voltage degrades the Copper (Cu) foil, while a higher one forms reactive Li dendrites increasing the potential safety hazards of the

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Nomenclature

Al	Aluminium
Co	Cobalt
Cu	Copper
ESS	Energy Storage System
EV	Electric Vehicle
Fe	Iron
HF	Hydrogen Fluoride
Li	Lithium
Li ⁺	Lithium ions
LIB	Lithium-ion Battery
Mn	Manganese
Ni	Nichel

product (Tarascon and Armand, 2001). Besides the active material of electrodes, fundamental LIB components are the highly dielectric solvent allowing the transfer of Li⁺, the polymeric separator preserving electrodes from direct contact and Cu and Aluminium (Al) current collector foils, on which active powder is adhered through an organic binder (Hanisch et al., 2015).

Thanks to its low atomic weight and high energy density (120 Wh/kg) ensuring product lightness, low self-discharge rate, good longevity (500–1000 cycles), absence of heavy metals (Lead or Cadmium) and a wide operating thermal range (−20/60 °C), LIB applications are significantly increased in the last years (Al-Thyabat et al., 2013). The use of LIBs in portable electronic devices, such as mobile phones, laptop, cameras, toys, e-cigarettes and electric and garden tools, has doubled from 2014 to 2019, of which 37.2% are Lithium Cobalt Oxide (LCO), 29% Lithium Nickel Manganese Cobalt Oxide (NMC) and 5.2% Lithium Iron Phosphate (LFP) (Boyden et al., 2016). LIBs market also moves from small-scale applications to large-capacity sectors, such as Electric Vehicles (EVs) and Energy Storage Systems (ESSs), to reduce greenhouse gas emission and oil dependency or to solve intermittency of alternative green energy sources (Kim et al., 2012B). In EV applications, for

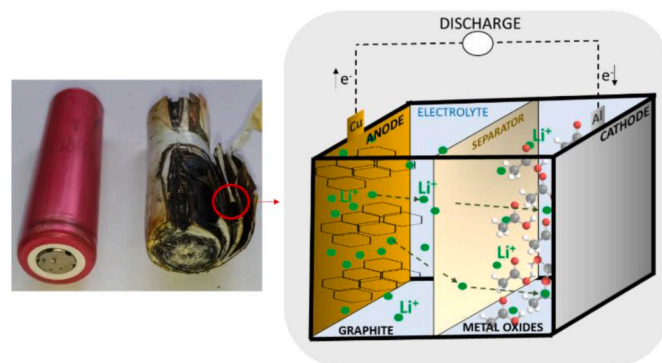


Fig. 1. Inner structure of a cylindrical LIB (left) and electro-chemical operating mechanism of a LIB cell (right).

example, LIBs sales will increase from 5 million of 2015 to 7 million of 2020, till reaching 180 million in 2045 (Gao et al., 2018). Although the production is mainly localized in Asian countries (40% of the production is in Japan, followed by South Korea and China), the major consumption is in USA (28.4%) and EU (27.2%), where the battery sector represents the fastest growing waste stream due to the increasing electrification in automotive sector (Georgi-Maschler et al., 2012). According to LIBs short estimated life span (3–8 years), it was predicted that more than 25 billion units and 500 thousand tons of LIBs will become a waste in 2020, even if a strong collection system and a well-established recycling process are still missing (Yu et al., 2018). In the case of portable batteries, only the 30–50% of population, in fact, properly dispose of LIBs, being unaware of the potential harmfulness of post-use products. The presence of metallic Li due to an incorrect cycling of the battery, highly reactive with moisture, and the inner presence of a flammable electrolyte could cause explosive reactions and the emission of harmful gases (such as Hydrogen Fluoride, HF) in case of mechanical damages, overheating or degradation phenomena, exposing people to serious injuries (Sonoc et al., 2015). The main numbers of LIBs market and waste production are summarized in Table 2.

Considering that in the mid and near future only Li-based batteries

Table 1

Typical chemical composition of Lithium-ion batteries. Data are obtained as average of %wt reported in Al-Thyabat et al., 2013, Bankole et al., 2013, Boyden et al., 2016, Chen et al., 2018, Diekmann et al., 2017, Dorella and Mansur, 2007, Georgi-Maschler et al., 2012, Gratz et al., 2014, Hanisch et al., 2015, Huang et al., 2016, Jha et al., 2013, Kim et al., 2012B, Lee and Rhee, 2002, Li et al., 2013, Meshram et al., 2014, Nirmale et al., 2017, Nitta et al., 2015, Rahman and Afroz, 2016, Tarascon and Armand, 2001, Xu et al., 2008, Wang et al., 2017 and Winslow et al., 2018.

Cell components	Chemical composition	%wt.	Additional information
External casing	Fe–Ni alloy	20–26	Steel case is typical of cylindrical cells.
	Al	10	Aluminium case is found in prismatic cells.
Cathode		25–30	
Aluminium	Al	5–8	
Binder	Usually PVDF	1–2	Alternatives: PTFE, butadiene-styrene rubber (SBR) or modified cellulose (e.g. CMC).
Metal oxide	Li	1.5–7	LCO gives better performances but is highly expensive.
	Co	5–20	It is replaced by NMC, LMO (where Mn gives structural stability) or C-coated LFP (LiFePO ₄) that is safer.
	Ni	5–10	
	NCA (LiNi _{0.8} Co _{0.15} Al _{0.05} O ₂)		
	Mn	5	
	LMO (LiMnO ₂)		
	NMC (LiNi _x Co _y Mn _z O ₂)		
Polymeric separator	Microporous PP or PE	4–10	
Electrolyte		10–15	EC is the most used organic solvent, combined with others to lower its high melting T.
Li salts	LiPF ₆ , LiAsF ₆ , LiClO ₄ , LiBF ₄		LiPF ₆ has high conductivity in any medium.
Organic solvents	DMC-EC, PC-DME, BL-THF		
Anode		15–25	
Copper	Cu	8–10	
Binder	Usually PVDF	1–2	Inert, thermo-resistant and current-resistant binder helping the adhesion.
Graphite		15–17	Low storage capacity of graphite (372 mAh/g). Alternatives: C-NT, Sn compounds, metallic NP.

Table 2
LIBs evolution according to market demand and End-of-Life management.

	LIBs evolution		
	Past years	2010–2020	Near Future
LIBs Market			
Applications	Mainly electronic devices: toys, lights, electric tools, mobile phones, laptops	35% electronic device 50% e-mobility	Increasing adoption for intermittent renewable power generation
Global LIBs market	500 million LIBs units produced in 2000	30 billions \$ market (2017)	100 billions \$ market (2025)
EV LIBs sales	–	>5 million	180 million (2045)
Constituent	LCO 37%, NCM 29%	NMC 31%, LFP 38%, LCO 14%	68% NMC, Li-S, Li-O
Materials	LFP 5%	Liquid electrolyte	Solid electrolyte
Price	1000–500 \$/kWh	150–300 \$/kWh	>100 \$/kWh
End-of-Life LIBs			
Waste production	200–500 tons/year (China 2006)	500 thousand tons (China 2020) 200 million tons (2017)	300 million ton (2015–2040 just from EVs)

could satisfy the automotive requirements and the great power demand of portable devices, the aim of the present paper is the analysis of current processes for the treatment of post-use LIBs, highlighting the potentiality of a circular approach and pointing out the relevance of LIBs recycling together with re-design, reuse and remanufacturing. A comprehensive view of the advancements in each process step allows to overcome the peculiar fragmentation of post-use treatments, where waste preparation is studied separately to the hydrometallurgical or pyrometallurgical phases. Furthermore, along with an in-depth literature review, actual industrial processes are critically analysed in order to point out their advantages and disadvantages through the use of comparison tables. Together with that, legislative and economic barriers related to the development of sustainable and innovative management waste solutions are reported.

2. Circular economy for LIBs

The Circular Economy (CE) is a regenerative approach designed to reduce waste, and aimed at guaranteeing the eco-sustainability of post-use products. A CE approach for LIBs represents the core topic analysed in more than 3000 researches conducted in the last 10 years, and focused on the investigation of its main steps (Yun et al., 2018).

• Design

The main features of the product are defined during the design phase. In the automotive sector, the crash safety, the centre of gravity and the space optimization represent the major drivers for design, along with the functionality of each specific component (Elwert et al., 2018). In a CE scenario, however, design should facilitate the second use and the final disposal of the product through a proper LIBs labelling (e.g. QR codes, RFID tags), a standardization of formats, structure and composing materials, a reversible assembly strategy and a clear classification of inner hazardous components (Gu et al., 2017). On this topic, the US Society of Automotive Engineers and the European EUROBAT create a series of working groups to discuss and develop solutions for LIBs sustainable re-design (Wang et al., 2017).

• Raw material mining

The main natural reserves of Li are in China and South Africa, where the mineral is extracted from igneous rocks through roasting and leaching processes (Meshram et al., 2014). The extraction from brine, in fact, is limited due to technological barriers: 2'000 tons of water are needed to obtain 1 ton of Lithium (Katwala, 2018). However, these sources are not endless, and it was predicted that in 2023–2025 the demand of Li will be greater than the mining supply, unless LIBs are not recycled with a 90% efficiency (Sonoc and Jeswiet, 2014). Even worse, Ni will require 170 fold the current extracting capacity, while Co (25%

of total request used for LIBs production) is obtained by deposits in the Democratic Republic of the Congo, sanctioned for human rights abuses (Nkulu et al., 2018).

• LIBs production and use

Continuous improvements of LIBs chemistries and performances are studied at lab and industrial scales to satisfy the increasing market demand (Scrosati and Sun, 2011). Along with LIBs portable applications, the e-mobility is more and more significant in the automotive sector, with more than 1 million of EVs sold in 2017 worldwide (International Energy Agency (IEA), 2018).

• Post-use collection

Nowadays, although the huge amounts of produced and sold portable LIBs, only 29.5% of population properly collect them, versus 59.6% that store LIBs at home and 15.9% who throw them in the trash bins (Wang et al., 2014). To overcome this issue, some regulations have been recently introduced. The European Battery Directive 2006/66/EC ensures a minimum batteries collection rate of 45% by 2016 and forces producers to be responsible of LIBs collection and treatment, as well as the Chinese Extended Producer Responsibility (ERP) plan of 2017 (Ruffino et al., 2011). In USA, instead, despite the absence of national regulations, different organizations (e.g. Call2Recycle, Battery Solutions and Big Green Box program) operates to properly collect and manage post-use LIBs (Winslow et al., 2018).

• Re-use

Considering the residual capacity of end-of-life LIBs coming from EVs ($\approx 80\%$, 6700 cycles) and the upgrading offered by EVs manufacturers to increase the sales, LIBs reuse and remanufacturing for second life applications represent an interesting opportunity for new businesses. ESS for renewable sources, management of network loads or emergency generators could be suitable due to their required lower performances, as well as cleaning and agricultural machines, construction equipment, forklifts, e-bikes, etc. Different researches are being conducted to determine the remaining life and LIBs degradation phenomena and the first experimental systems were developed thanks to the collaboration between EVs manufacturers (e.g. BMW, GM and Nissan Motor) and energy management companies (e.g. Vattenfall, ABB and Sumitomo) (Natkunaratjah et al., 2015). Along with a fast and efficient diagnostic of LIBs status, another major challenge for reuse is a safe and non-destructive disassembly of battery pack, based on automated processes able to overcome product variability (Arora et al., 2018).

• Landfill and thermal recovery

When LIBs are irreversibly damaged, a possible destination is land-fill, where LIBs could contaminate soil and groundwater due to electrolyte and metals leaching (Gu et al., 2017). Furthermore, in contact with moisture, LIBs release toxic gases (i.e. HF) and ignite fire accidents, as in the case of incineration (Winslow et al., 2018).

• Recycling

LIBs recycling allows to reduce energy consumption and CO₂ emissions, save natural resources avoiding virgin materials mining and imports, minimize environmental toxicity, create an economic gain, decrease waste and manage safety issues (Bankole et al., 2013). It was estimated that metals recycling can save 13% of LIB cost per kWh, but nowadays less than 3% of LIBs are recycled in the world (Sonoc et al., 2015).

One of the major drivers for LIBs recycling is the economic value of metals contained in the cathodic active layer, representing 90% of the total value, as reported in Table 3 (Lain, 2001). According to the combined effect of recycling feasibility and final gain, only Co, Cu, steel, Ni and Al are currently recycled, while plastics are incinerated for energy recovery and Li, Mn and graphite are rarely considered (Dewulf et al., 2010). At industrial level, most of the recycling processes are specifically set for LCO (8900\$/ton) and NMC chemistries, extremely profitable due to the high Co content; on the contrary, they are not convenient for the lower-value LMO (860\$/ton) and LFP (not containing precious metals) (Winslow et al., 2018). Considering also the predominant trend to substitute Co to lower production costs, recycling processes should be developed to recover LIBs regardless their specific compositions and to balance treatment costs with the final effective revenue (Chen et al., 2015).

Along with the need to develop flexible processes, able to deal with the variability of input chemistries, presence of impurities, geometric variability and new market developments, other challenges of recycling are (i) the safe handling of LIBs components due to electrolyte or Li atoms exposure, (ii) the scalability, (iii) standardization and (iv) simplification of treatment steps (Diekmann et al., 2017).

A usual recycling process is shown in Fig. 2. After a preliminary phase for waste preparation (i.e. sorting, discharging and dismantling), LIBs are pre-treated to segregate the active valuable materials through thermal (e.g. organic components evaporation), mechanical (e.g. crushing and shredding), physical (e.g. floatation), chemical (e.g. binder or current collector foils dissolution) or mechano-chemical processes (Ra and Han, 2006). Thanks to these treatments, the metals-enriched separated fraction is subsequently subjected to acid solutions leaching or high temperature pyrolysis and smelting. Hydrometallurgy strongly depends to solid/liquid ratio, reductant species, leaching times and temperatures and it allows metals recovery with high purity, good efficiency and low energy requirements (Li et al., 2018). At the opposite, pyro-metallurgical processes are characterized by high capital costs, significant emission of hazardous gas, loss of Li content and intensive energy consumption, but are able to treat indifferently all LIBs

chemistries with a relatively simple procedure (Jha et al., 2013).

The bio-hydrometallurgical process is a minor alternative, mainly studied at lab scale, for the treatment of wasted LIBs: electrode materials are dissolved by metabolites excreted by microorganisms or fungi (e.g. *Acidithiobacillus ferrooxidans*, *Aspergillus Niger*, *Penicillium Semplicissimum* and *Alicyclobacillus*), enhancing dissolution obtained with pure acids (Xin et al., 2016). Although this technique has been successfully applied in other sectors to extract metals from low-grade ores (e.g. from acid mine drainage, spent refinery wastewaters and e-waste), the implementation at large scale is still challenging and researches specifically targeted to LIBs are still under investigation (Calvert et al., 2019). The main issues are related to the required pulp density during the process, the low concentration of Fe and S in LIBs waste allowing the activation of the reaction and slow leaching times (Boxall et al., 2018). However, this technique is characterized by low costs and mild operative requirements (Horeh et al., 2016).

Differently to these processes aimed at individually recovering valuable metals, regeneration method recycles and directly resynthesizes cathodic powder after re-lithiation through co-precipitation or sol-gel technologies. D1EPHA in kerosene is generally used to leach and precipitate NiCoMn hydroxide, while, dispersing cathodic materials in a solvent containing complexing agents (e.g. citric acid), a sol-gel is formed and extracts the desired metal product via thermal treatment (Yang et al., 2017b). Similar studies have been conducted for graphite anodes, demonstrating the possibility to regenerate them with properties comparable with ones of pristine graphite (Sabisch et al., 2018). Avoiding metals separation and purification steps thanks to the minimal morphological changes induced by recycled materials, the recovery efficiency and operating times are optimized, but the supply of LIBs with the same chemistry strongly influences the scalability and reproducibility of the process, extremely sensitive to contamination (Dunn et al., 2012).

Lots of processes mix two or more of these technologies to increase recovery efficiency, spending time and efforts to properly optimize operating parameters. The final step is common to all and leached metals are deposited into their metallic form through solvent extraction, precipitation (e.g. through pH-controlled NaOH solution) or electrolysis (Xu et al., 2008). In the next paragraphs the most studied and used technologies are deeply investigated and reported in order to identify technical and economic advantages and barriers.

3. Waste preparation and pre-treatment processes

The preliminary treatment phase is addressed to the battery deactivation, with the aim to lower the risks associated to the LIBs handling and manipulation, due to the presence of residual stored energy. The discharge, in particular, reduces the amount of metallic Lithium, minimizing the risk of explosions, and it generally consists of LIBs immersion in NaCl- or Na₂SO₄-saturated solutions (Wang et al., 2017). The operating times are strongly dependent on the specific solution conductivity, operating temperatures and LIBs state of charge, but the use of aqueous salt baths could also generate toxic HF gases due to the leakage of electrolyte in water, particularly for high-voltage battery pack, and contaminate inner materials with undesired ions (Al-Thyabat et al., 2013). Other proposed alternatives are covering LIBs with stainless steel chips to stimulate a controlled short circuit or connecting them to resistors to collect and reuse the residual energy (Lv et al., 2018). This latter allows to recover 7 MJ from 1 ton of battery (195 Wh/kg) discharged from 3 to 0V, and which could be used to heat the leaching vessel during hydrometallurgical step (Sonoc et al., 2015). However, the modest gain usually is not sufficient to cover investments costs and, if not recovered, the liberated energy should be properly dissipated through cooling systems (Harper et al., 2019). In addition, dismantling aims to decrease the volume of the product to be treated, particularly for EV LIBs. Different research studies investigate human-robot cooperation to significantly reduce times and costs, proposing solutions to face

Table 3
Economic value of LIBs components.

Cell components	Value (US\$/ton)		
	2001	2017	2019
Cathode			
Al	1'250	2'000	1'800
Li	7'500	9'000	10'000
Co	38'000	55'000	35'500
Ni	8'600	10'000	13'200
Mn	1'100	2'000	2'000
Anode			
Copper	1'800	5'500	5'800
Graphite	550	1'000	800

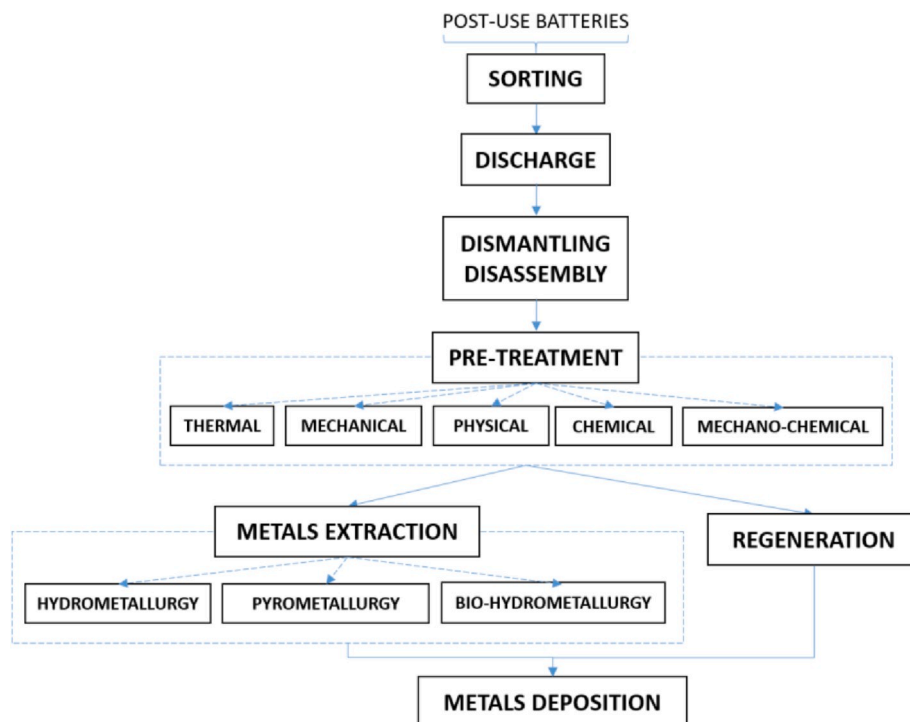


Fig. 2. Typical LIBs recycling process.

different product sizes, joining connections, disassembly tasks, etc. (Wegener et al., 2015).

Once secured, LIBs are ready for recycling processes. The main objectives of pre-treatment processes (Table 4) are the enrichment of metallic fraction, the reduction of scrap volumes and energy consumption, the improvement of recovery rate and the management of safety issues, obtained exploiting different chemical and physical properties of LIBs components (Shin et al., 2005). Pre-treatments are necessary before hydrometallurgical processes due to the high selectivity of leaching agents, hindered by the presence of impurities (e.g. Al and Cu coming from the current collector foils).

3.1. Thermal pre-treatments

High temperature treatments principally act on LIBs organic components. Among them, Polyvinylidene Fluoride (PVDF) binder is responsible of the active powder adhesion on current collector foils and, although the adherence decreases during the ageing of batteries, it could represent the major challenge for materials separation. The optimal temperatures range to decompose it, was found at 500–600 °C, as confirmed by SEM analysis (Chen et al., 2018), leading to the spitting of Carbon chains into shorter units and allowing the easy material detachment from Al and Cu layers (Yang et al., 2016). On the other hand, a temperature of 550 °C induces also metals phase transformation (from LiCoO_2 to Co_3O_4 and from $\text{Li}_4\text{Mn}_5\text{O}_{12}$ to LiMnO_4) increasing the subsequent leaching efficiency, while higher temperatures (>600 °C) affect Al foil integrity, making it fragile or partially melting it to create a surface barrier on Li–Co particles (Sun and Qiu, 2011).

Another relevant operating parameter of thermal pre-treatments is the pressure. The use of vacuum chambers protects metals from the oxidation but requires a higher energy consumption, contrary to the enclosed vacuum environment, that starts the process at high vacuum degree avoiding continuous pressure monitoring (Xiao et al., 2017). Otherwise, oxygen-free roasting in N_2 atmosphere significantly improves wet magnetic separation: the slightly-soluble roasted LiCO_3 dissolves in water, metallic Co is attracted by the magnetic stirrer (with 75% efficiency) and graphite precipitates on the bottom of the vessel (Li

et al., 2016).

High temperature thermal treatments improve Li recovery efficiency up to 90% thanks to carbon removal, but requires air-filtering systems and gas scrubbers due to significant toxic gaseous emissions (e.g. dioxins, HF, CO, CO_2 , etc.) (Paulino et al., 2008). As alternative, cooling treatments were used by TOXCO, exploiting cryogenic temperatures (−200 °C) to make Li inert (Al-Thyabat et al., 2013). The cooling requires 198 MJ and is generally combined with wet grinding (565 kJ) to isolate the active cathodic material (Sonoc et al., 2015).

3.2. Mechanical and physical pre-treatments

The combination of mechanical and physical pre-treatments is the most used technique at industrial level and allows to remove the outer case, to segregate valuable materials and to reduce scrap volumes. The impact stresses generated during grinding transform the kinetic energy in breakage energy, detaching the active powder from the substrate, breaking the agglomerates and creating two or more fractions (Hanisch et al., 2015). The increased surface area will affect the subsequent hydrometallurgical process, promoting metals dissolution during acid leaching (Al-Thyabat et al., 2013).

Generally, a first crushing step is performed to remove external steel case by magnetic separation with 1.7% cathode loss, followed by a fine grinding that segregates current collector foils and organic materials from the active leachable powder (Gratz et al., 2014). Cu and Al layers, in fact, are malleable and tend to curl rather than be crushed, remaining in larger sieves (>300 μm) (Zhou et al., 2010). As reported in Fig. 3, coarser fractions are then subjected to Eddy Current technologies to separate Al and Cu and to densimetric table to remove plastics, with a final yield of 85% (Pagnanelli et al., 2016). The described mechanical pre-treatment could be performed under brine solution to decrease emissions and Li reactivity or in dry and controlled atmosphere avoiding electrolyte hydrolysis or violent short circuit in presence of oxygen (Lain, 2001). If any thermal evaporation or binder dissolution are preliminarily performed, a mass decrease of $\approx 8\%$ is recorded during crushing, due to gaseous emissions from electrolyte decomposition (Diekmann et al., 2017).

Table 4
Optimal operating conditions of pre-treatments.

Process	Operating parameters	Ref.	Pros	Cons
<i>Discharge</i>				
Salts saturated solution	10%wt NaCl 36 h; 5%wt NaCl 24h	Chen et al., 2018; Wang et al., 2018		Ionic contamination
<i>Thermal pre-treatment</i>				
Calcination	In a muffle furnace 300 °C; 450 °C 15 min; 500 °C 2 h; 500 °C 30 min with an heating rate of 5 °C/min; 500–580 °C; 700 °C 1h	Diekmann et al., 2017; Wang et al., 2018; Xu et al., 2008; Paulino et al., 2008;	Cell opening and deactivation; Binder and organic compounds removal; Easiness; Economically sustainable.	Cu corrosion; Toxic gaseous emission; High energy consumption.
Oxygen-free roasting	N ₂ atmosphere in a tube furnace 1000 °C 30 min; 600 °C 15 min	Yang et al., 2017a; Rahman and Afroz, 2016		
Enclosed-Vacuum Environment	800 °C	Li et al., 2016; Yang et al., 2016		
Vacuum Pyrolysis	Pressure 1 kPa 600 °C 30 min with an heating rate of 10 °C/min	Xiao et al., 2017 Zhang et al., 2013; Sun and Qiu, 2011		
<i>Mechanical pre-treatment</i>				
Grinding	4 mm crushing; hammer mill 6 mm 2000 RPM Initially 20 mm, then fine crushing 10 mm	Jha et al., 2013; Gratz et al., 2014; Shin et al., 2005	Valuable metals segregation; Scrap volume reduction; Reduces costs and high throughput; Flexibility; Increased hydrometallurgical selectivity.	Not complete separation; Impurities; Gaseous emissions. Cathodic powder input.
<i>Physical pre-treatment</i>				
Sieving	Sieves at 300 µm; 300 µm 3 min; 250 µm Sieves at 850 m, 200 m and 106 m Vibrating sieve 10 min 2 mm, 1 mm and 500 µm Air jet separation 1 min with sieve 50 µm	Al-Thyabat et al., 2013; Zhou et al., 2010; Gratz et al., 2014; Pagnanelli et al., 2016; Hanisch et al., 2015		LiCoO ₂ -graphite contamination.
Ultrasonic washing	12 mm aperture 15 min With NMP 70°C 90 min 240W	Huang et al., 2018; Zheng et al., 2018	Low energy consumption No external impurities.	
Floatation	Combined with grinding 5 min	Yu et al., 2018; Wang et al., 2018		
<i>Chemical pre-treatment</i>				
Electrolyte dissolution	Supercritical CO ₂ ; anhydrous solvent with boiling T < 80 °C	Al-Thyabat et al., 2013; Huang et al., 2018		Wastewater production;
Binder dissolution	NMP 40 °C 14 min; NMP 100 °C 1 h DMF 60 °C 2h Citrus Fruit Juice 90 °C; <i>Citrus Pseudolimon Tanaka</i> 90 °C 20 min DMAC 30 °C 30 min with solid(g)/liquid(mL) 1:20	Bankole et al., 2013; Al-Thyabat et al., 2013; Zhou et al., 2010; Lv et al., 2018; Pant and Dolker, 2017; Huang et al., 2018		
<i>Mechano-chemical pre-treatment</i>	EDTA chelate agent in a grinding mill at 600 RPM 4 h with ball/powder 80:1 EDTA-2Na 2 h with a cathode/agent ratio 3:1 PVC + Fe 12 h with cathode/agent/Fe ratio 1:1:2	Wang et al., 2016; Yang et al., 2017a; Wang et al., 2017	Room temperature; Low energy consumption; Simple procedure; Economically sustainable; Environmentally-friendly.	Long reaction times; Noise generation.

In recent years, as alternative to conventional crushing, High Voltage Fragmentation (HVF) has been scaled from the mining industries to the recycling of complex and high added value products (Bluhm et al., 2000). In this process, the product is put in a grounded metallic vessel and is covered by a dielectric liquid (typically deionized water) in order to feel a pulsing fast-rising voltage up to 40–200 kV, generated through a couple of electrodes. When reached the dielectric voltage breakdown, a plasma channel is created and the product is crushed by the resulting shockwaves (McCluskey et al., 1994). For LIBs recycling, specifically, the Electro-Hydraulic Fragmentation (EHF) was tested, where the spark channel is generated in the liquid medium outside the product, showing promising results in the almost complete separation of the black mass from the Al current collector foil in the lower dimensional fraction (Horn et al., 2018). Nevertheless, the high costs related to the current equipment make HVF process less convenient than a traditional crushing (Leißner et al., 2018).

An additional physical pre-treatment is ultrasonic washing, where the cavitation phenomena (i.e. rapid change of pressure in a liquid able to create impact waves) is combined with the rinsing effect of the stirring, significantly improving the separation of active metals powder to Al foil. If coupled with the use of solvents acting on PVDF binder, this technique could reach 99% efficiency, with lower energy consumption with respect to multiple crushing steps or thermal treatments (Huang et al., 2018).

Floatation, instead, exploits physical surface wettability of LIBs

components to separate LiCoO₂ powder (polar and hydrophilic) from graphite (non-polar and hydrophobic). This technique is characterized by a high potential industrial applicability and a high efficiency, but requires PVDF removal that, being used as binder in both electrodes, minimize the different wettability properties of particles. PVDF could be evaporated by roasting, that generates harmful gases and could damage graphite, or could be dissolved through chemical solutions, introducing impurities. An alternative solution is the combination of floatation and grinding, exploiting the generated horizontal shear forces to expose powder active surfaces (Wang et al., 2018).

3.3. Chemical and mechano-chemical pre-treatments

Chemical pre-treatments use organic solvent and supercritical fluids (e.g. CO₂) to extract the electrolyte or dissolve the binder. Minor applications are aimed at selectively recover cathodic current collector foil through alkali solutions (2M NaOH), exploiting the amphoteric properties of Al. This technique reaches 97%wt. Al extraction efficiency and preserves other metals but introduces hardly-leachable ions, thus producing harmful alkaline wastewaters (Zhang et al., 2013).

Different substances have been tested to dissolve PVDF binder: it has been proved that strong acids and basis, strong oxidants, halogens and hydrocarbons do not react with it; on the contrary, organic solvents such as N-methylpyrrolidone (NMP) or N, N-dimethylformamide (DMF) show a good solubility (200 g/kg) at moderate temperature (≈100 °C)

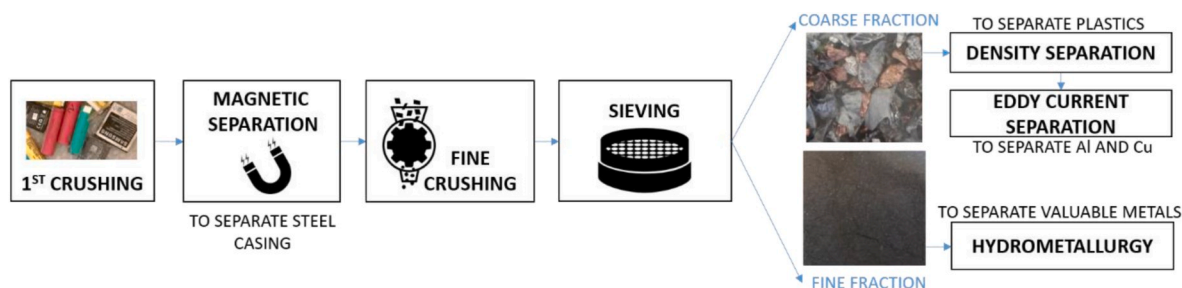


Fig. 3. Combined mechanical and physical pre-treatments to segregate cathodic active powder.

(Contestabile et al., 2001). Between them, DMF is preferable due to its higher efficiency and reduced costs, as well as for a lower potential toxicity compared to the NMP (Zhou et al., 2010). However, more recent studies also move to cheaper and greener alternatives, like the citrus fruit juice or the Hill Lemon Galgal solution, able to dissolve PVDF binder in about 20 min (Pant and Dolker, 2017).

The main disadvantage of these pre-treatments is the strong dependence to the chemical composition; in fact, the cited organic solvents selectively act on PVDF binder, resulting ineffective on Polytetrafluoroethylene (PTFE). The latter has non-polar properties and was introduced with LFP batteries thanks to its greater cohesive force and increased rate capability (Zheng et al., 2018). N-dimethylacetamide (DMAC) was used at 30 °C for 30 min to remove it with a good extraction efficiency (Huang et al., 2018).

Finally, mechano-chemical pre-treatments take advantage of the combined effect of grinding and metals chelate agents leaching, such as Ethylenediaminetetraacetic (EDTA) and PVC. The particle size reduction and the increase of surface area cause the bond breakage and the polymorphic transformation of cathodic material, increasing reaction activity for the subsequent acid leaching (Wang et al., 2016). The energy is transferred to LIBs components by the shearing, impact and squeezing action of a ball milling, supported by the presence of chelate agents able to destroy the original crystal structure. However, although the easiness and economical sustainability of this process is attractive, long reaction times make it really far from a possible industrial implementation (Wang et al., 2017).

4. Hydrometallurgical processes

After pre-treatments, the active cathodic powder is leached through hydrometallurgical techniques to separate and purify LIBs valuable metals. The main advantages of hydrometallurgical processes are (i) the reduced energy consumption thanks to lower temperatures, (ii) the recovery of Li in the carbonate form, (iii) the leaching of metals to be reused for LIBs new cathodes and (iv) a good efficiency on different battery chemistries (Gaines et al., 2018).

Traditionally, strong inorganic acids, like hydrochloric (HCl) (Takacova et al., 2016; Zhang et al., 1998), sulphuric (H₂SO₄) (Chen et al., 2011; Dorella and Mansur, 2007; Ferreira et al., 2009; Kang et al., 2010; Nan, 2005), nitric (HNO₃) (Lee and Rhee, 2002) and phosphoric acid (H₃PO₄) (Chen et al., 2017; Pinna et al., 2017), are used as leaching agents due to their ability to dissolve metals. The main operating parameters of these processes are temperature, acid and reducing agent concentration, reaction time and solid/liquid ratio. Although all of them were tested in laboratory, only HCl was investigated at pilot scale. During the experiments, the highest recovery efficiency for Co and Mn (≈99%) was obtained with a 1.75M solution of HCl, a temperature of 50 °C, a leaching time of 2 h and a pulp density of 20%w/v, indicating the industrial scalability of this LIBs recycling process (Barik et al., 2017).

At the opposite, new researches focus on the use of organic acids produced by microorganism, due to their reduced health issues and increased eco-friendly features. Organic acids (such as acetic, ascorbic or malic acids) do not represent a contamination for the environment,

are biodegradables and can be easily recycled (Jadhav and Hocheng, 2012). In Table 5 leaching efficiencies found in literature are reported and the best solution for wasted LIBs results as citric acid > malic acid > acetic acid dissolution. The optimal conditions for citric acid leaching, in particular, are two dissolution steps at 90 °C with H₂O₂ reducing agent (Golmohammadzadeh et al., 2018). The addition of a reducing agent allows to achieve higher leaching rates, especially in the case of organic acids.

The most used are Hydrogen Peroxide (H₂O₂) (Li et al., 2009) and Sodium Bisulphite (Na₂S₂O₅) (Meshram et al., 2015), enhancing recovery efficiency from ≈50% to ≈99% for Li and from ≈20% to ≈90% for Co, but they are not sufficiently environmental-friendly (Li et al., 2013).

As green alternatives, glucose (C₆H₁₂O₆), sucrose, lactose and ascorbic acid (Beolchini et al., 2001; Peng et al., 2018; Vegliò et al., 2000) were tested, as well as different wasted products, such as tea waste, powders of *Phytolacca Americana* branch, cornstalk, sawdust, molasses and corncob (Cheng et al., 2009; Hariprasad et al., 2007; Su et al., 2008; Tang et al., 2014; Tian et al., 2010). Also some more environmental friendly inorganic reducing agents are available, such as pyrite (Kanungo and Jena, 1988).

The main challenge of acid leaching is the selectivity: during the process also other metals and ions are dissolved, making harder the separation of the target materials. Some authors use Ammonia-ammonium Sulphate ((NH₄)₂SO₄) as leaching agent and Sodium Sulphite (Na₂SO₃) as reducing agent in order to selectively leach valuable metals (i.e. Li, Co and Ni) (Zheng et al., 2017).

Another significant issue of hydrometallurgy is the strongly dependence to pre-treatments, able to separate the cathodic powder to be leached. Obtained results could vary enormously, depending on the technologies used in the previous steps. In many research studies, LIBs pre-treatment has been performed manually, indicating that dissolution rates could change at pilot or industrial scales if combined with an automated preparation phase. Pagnanelli et al., 2014, works on this topic and evaluates the leaching of LIBs electrodes powder, obtained through grinding and sieving processes in a pilot plant, with H₂SO₄+H₂O₂. In the paper, they develop a flexible purification section to ensure the application of the process at industrial scale and deeply study the dissolution adaptability according to LIBs metals concentration in input.

5. Pyrometallurgical processes

Pyrometallurgical processes are based on high temperatures and consist of three phases:

- **Pyrolysis:** the thermal degradation of organic LIBs components;
- **Metals reduction:** the production of metal alloys using ≈1500 °C and proper reductive agents;
- **Gas incineration:** the pyrolysis and quenching of gases at ≈1000 °C to avoid dioxins release.

The initial pyrolysis of electrolyte and plastic could be used to supply

Table 5

Recovery rates of Li and Co from spent LIBs.

	Leaching agents	Ref.	Metal recovery rates	
			Li(%)	Co(%)
Inorganic	H ₂ SO ₄ /HNO ₃ /HCl	Joulié et al., 2014	>80	100
	H ₂ SO ₄ + NaHSO ₃	Meshram et al., 2015	96.7	91.6
	H ₂ SO ₄ + H ₂ O ₂	He et al., 2017	>99.7	>99.7
	NH ₃ + (NH ₄) ₂ SO ₃ + (NH ₄) ₂ CO ₃	Ku et al., 2016	–	80
Organic	Oxalate	Zeng et al., 2015	98	97
	Ascorbic acid	Li et al., 2012	98	95
	Acetic acid	Golmohammadzadeh et al., 2017	75	30
	Lactic acid + H ₂ O ₂	Li et al., 2017a	98	99
	Iminodiacetic acid + H ₂ O ₂	Nayaka et al., 2016a	99	91
	Maleic acid		100	97
	DL-malic acid + H ₂ O ₂	Golmohammadzadeh et al., 2017	91	84
	Citric acid + H ₂ O ₂		92	84
	Citric acid + H ₂ O ₂	Mishra et al., 2008	99	98
	Citric acid + H ₂ O ₂	Li et al., 2010	100	90
	Citric acid + H ₂ O ₂	Santana et al., 2017	100	100
	Citric acid + TW	Chen et al., 2015	98	96
	Succinic acid	Li et al., 2015	100	96
	Tartaric acid + H ₂ O ₂	He et al., 2017	99.1	98.6
	Phosphoric acid + glucose	Nayaka et al., 2016b	100	98
Inorganic + organic				

energy for metals recovery, when in the shaft furnace is obtained the valuable alloy containing Cu, Co, Ni and Fe (Gaines, 2014).

Although 100% recyclable, Li is generally not recovered due to the economical unfeasibility of slag leaching and, along with Al, is currently used as aggregate in concrete (Rahman and Afroz, 2016). Pyrometallurgical processes, in fact, strongly depend on LIBs chemistries, particularly on Co content and price and the treatment of LMO, LFP and NMC has generally not a positive impact with respect to mining, both economically and environmentally (Winslow et al., 2018).

Along with material loss and hazardous gas generation, another significant disadvantage of high temperature treatments is the relevant energy consumption. The industrial Umicore process, for example, consumes 5000 MJ to smelt 1 ton of LIBs and to clean up generated gases, while in Sony-Sumimoto process 992 MJ/ton are required for organic material incineration (Sonoc et al., 2015). Pros and cons of pyrometallurgy are compared with hydrometallurgy in Table 6.

To face all these issues and to better exploit pyrometallurgy features, improvements for lab scale processes were always investigated. Some researches focused on slag leaching through sulphuric acid hydrometallurgy to make it economically feasible and to increase the low efficiency ($\approx 40\%$); while others developed new processes, combining roasting (650 °C 30 min), carbonated water leaching and sulphuric acid leaching to obtain Li₂CO₃, CoSO₄, MnSO₄ and NiSO₄ (Hu et al., 2017).

Table 6

Comparison between pyro- and hydrometallurgical LIBs recycling processes.

Process	Pros	Cons
<i>Pyrometallurgy</i>	Easiness of procedure; No necessity of passivation steps; Optimal technology readiness; Generation of exothermic reaction reducing energy consumption.	High energy consumption; Hazardous gaseous emissions; Material loss (Li in the slag); Need of Co LIBs chemistries (pre-sorting); High capital costs.
<i>Hydrometallurgy</i>	High recovery efficiency; High quality outputs; Good technology readiness; Moderated energy consumption; No gaseous emissions; Recovery of all LIBs cathodic metals; Mild reaction conditions.	Wastewater productions; Incomplete binder/electrolyte recycling; Complexity of procedure; Need of pre-treatments; Selectivity of reagents.

6. Industrial processes and patents

The first commercial production line for LIBs recycling was produced by Toxco in 1994 and since then an increasing interest in the topic arise, adapting pre-existent processes to this new waste. Pyrometallurgy currently is the predominant process due to its flexibility: the use of high temperatures to recover metals is not born specifically for LIBs battery, but can be used to efficiently separate Co, Cu and Ni. However, these processes are not able to extract Li and Al, representing a barrier to the full recycling of LIBs components (Lv et al., 2018).

As already mentioned, the recycling of LIBs is economically interesting due to the presence of valuable metals that make the process sustainable. However, many technologies are still at lab or small-scale pilot plants and currently just few companies are able to recycle LIBs with a satisfying revenue. Recupyl, for example, developed and patented a process mixing both mechanical and chemical treatments to recover Li₂CO₃, Co(OH)₂ and CoO, but the lack of incomings and the low profitability of the process hinder its real industrialization.

Along with industrial development reported in Table 7, many patents were published in the last ten years regarding LIBs recycling. Umicore, for example, patented a classical pyrometallurgical process recovering Co-alloy (WO, 2011/035915 A1), but also a hydrometallurgical process focused on retrieving Li from the electrolyte (EP 2 410 603 A1). Similarly, Duesenfeld in Germany patented the recovery of LIBs electrolyte (US, 2018/0301769), highlighting the industrial efforts to recycle Li despite its lower profitability. Finally, the Avestor Limited Partnership patented process combines pyro- and hydrometallurgy to obtain Li₂CO₃ high grade (US 7192654 B2).

An interesting closed-loop circular economy approach is represented by the Japanese Sony-Sumimoto process. It is specifically devoted to recover Co(OH)₂ from Sony's spent LIBs used in electronic devices, such as laptop computers, camcorders, digital cameras and mobile phones, then directly reused for the fabrication of new batteries. The process involves the calcination of spent cells and utilizes the cogeneration resulting from burning electrolytes (Bernardes et al., 2004). Similarly, the Chinese Green Eco-Manufacture (20'000 ton/year) and Bangpo Ni/Co (3'600 ton/year) obtain a regenerated cathodic materials through hydrometallurgical re-synthesis of spent LIBs (Lv et al., 2018).

Nowadays, industrial processes are aimed at recycling portable LIBs, because batteries coming from end-of-life EVs are still few. However, in the near future, a huge amount of wasted automotive battery packs will be collected and sent to recycling facilities, introducing new issues in terms of chemistries, sizes and safety issues. According to EV

dimensions, LIB packs will require an efficient and safe dismantling phase before pre-treatments, both to reduce their size and to separate electronics components (e.g. Battery Management System, BMS), demanding new efforts to transform current manual procedures in fast automated systems (Yun et al., 2018).

Practical barriers to the industrial exploitation of innovative recycling processes is represented by the production of new generation LIBs and the precarious legislative framework. The trend to reduce Cobalt in spent LIBs, for example, will subtract to the recyclers their main current profit source. It will be therefore necessary to implement innovative recycling processes able to valorize all the set of materials available in next gen LIBs, to effectively implement a profitable circular economy value chain for these products (Bini et al., 2015). On the other side, although several EU norms and legislations are aimed at the management of electric and electronic waste or end-of-life vehicles containing LIBs, in most of cases a direct connection with the Directive 2006/66/EC regulating spent batteries is missing. This represents a limitation for the industrial treatment due to lack of data sharing, the uncertainty on

Extended Producer Responsibility (EPR) and the unrealistic targets for collection and recycling. LIBs, in fact, are actually classified as industrial batteries and their take-back, collection and recycling are regulated like products whose safety issues, market availability and logistics framework are totally different. However, the Directive is currently under revision and in its new form it will surely consider the exponential growth of LIBs market and their peculiar features during recycling processes.

7. Conclusion

Currently, rechargeable LIBs are the prominent technology to store energy in portable devices, EVs and energy systems, so much that their applications are exponentially grown in the last ten years. Along with manufacturing challenges, the increasing amount of wasted LIBs is becoming an urgent issue to face in order to protect the environment from pollution, to save the natural resources from an unrestrainable mining and to avoid safety hazards for humans.

Table 7

Industrial processes for recovering metals from LIBs. In the table are listed industrial processes as described in the following papers: Al-Thyabat et al., 2013, Bernardes et al., 2004, Blomgren (2017), Dewulf et al., 2010, Diekmann et al., 2017, Georgi-Maschler et al., 2012, Harper et al., 2019, Huang et al., 2018, Lain (2001), Lv et al., 2018, Meshram et al., 2014, Winslow et al., 2018 and Zhang et al., 2013.

Company	Process	Patent
<i>Accurec GmbH</i> Germany	Pyrometallurgy Hydrometallurgy Capacity: 4000 ton/year	–
<i>AEA Technology</i> UK	Hydrometallurgy Electrolysis Recovery: LiOH, CoO	–
<i>Akkuser Ltd</i> Finland	Pre-treatment Capacity: 4000 ton/year Recovery: metals powder	US 8 979 006 B2
<i>Batrec Industrie AG</i> Switzerland	Hydrometallurgy Capacity: 200 ton/year	–
<i>Duesenfeld</i> Germany	Pre-treatment Hydrometallurgy	US 2019/0260101
<i>Glencore plc (Xstrata)</i> Canada, Norway	Pyrometallurgy Hydrometallurgy Capacity: 7000 ton/year	A1
<i>Inmetco</i> USA	Pyrometallurgy Capacity: 6000 ton/year Recovery: Co-alloy	–
<i>Lithorec</i> Germany	Hydrometallurgy Recovery: CoO, Li salts	–
<i>OnTo Technology</i> USA	Pre-treatment Recovery: metals powder	–
<i>Recupyl</i> VALIBAT PROCESS France	Hydrometallurgy Capacity: 110 ton/year Recovery: Co(OH) ₂ , Li ₂ CO ₃	US 2017/0196725 A1
<i>Retriev Technology</i> (Toxco) USA/Canada	Hydrometallurgy Capacity: 4500 ton/year Recovery: CoO, Li ₂ CO ₃	US 5888 463d and US 8 616 475
<i>SNAM</i> France	Pre-treatment Capacity: 300 ton/year	–
<i>Sony</i> SUMIMOTO PROCESS Japan	Pyrometallurgy Hydrometallurgy Capacity: 150 ton/year Recovery: CoO	–
<i>Umicore</i> VAL'EAS PROCESS Belgium	Pyrometallurgy Hydrometallurgy Capacity: 7000 ton/year Recovery: LiCoO ₂ , Ni(OH) ₂ Efficiency: 70%	WO 2011/035915 A1

In an optimized circular economy model, wasted LIBs management starts with product design, developing systems easy to be reused and recycled and minimizing the amount of materials to be landfilled or incinerated. Furthermore, the residual features of end-of-life LIBs would be tested, in order to promote the reuse or to suggest remanufacturing solutions for new secondary applications. Recycling processes should be used as final option, developing treatments with the highest recovery efficiency and the lowest environmental impact, allowing primary raw material saving, economic gains, energy consumption reduction, waste minimization and safe management of harmful components.

Technically and economically feasible recycling processes are currently investigated at lab scale to develop the optimized sequence of steps. Generally, a preliminary deactivation of LIBs cells is performed, followed by pre-treatments able to segregate the fraction containing valuable metals. The cathodic powder represents the main focus of pyro- and hydrometallurgical processes, due to the presence of Co, Ni, Mn, Fe and Li oxides.

At industrial scale, pyrometallurgy is the most used technique to recover metals due to its simple process. However, the use of high temperatures produces lots of harmful emissions, requires high energy consumption and is not able to extract Li, that is generally slagged, pushing the research towards greener and more efficient solutions. Contrarily, the hydrometallurgy is complex and strongly dependent on the cathode chemistry, leading to unsustainable industrial treatments. All the technical solutions investigated in this paper underline the strong fragmentation of current processes and the economic and environmental barriers to be faced in the near future, when the return amounts of LIBs will become significant.

Author contributions

Conceptualization, E.M. and N.P.; methodology, E.M.; paper and literature review, E.M., O.R. and J.M.P.; writing—original draft preparation, E.M., O.R., J.M.P.; writing—review and editing, E.M., N.P., L.G. and M.C.; supervision, M.C.

Declaration of competing interest

The authors declare no competing interests.

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.

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