Circular Economy and the Fate of Lithium Batteries: Second Life and Recycling

Chiara Ferrara, Riccardo Ruffo, Eliana Quartarone, and Piercarlo Mustarelli*

There is a growing demand of electrochemical energy storage, driven by automotive and stationary requirements. Lithium-ion batteries (LIBs) are expected to dominate the market from the current 0.5 TWh to about 2.5 TWh in 2030. This will lead to great difficulties in the procurement of critical raw materials and in the management of end-of-life systems. From a circular economy perspective, it is necessary to identify reuse and recycling strategies that can make the demand fully sustainable. However, second life and recycling are not mutually excluding, while the final fate of the battery, or at least of its noblest components, should be recycling instead of disposal. In this context, to allow new strategies such as direct recycling of cathode powders, an accurate redesign of the battery system, from the single cell to the modules, which allows ease of separation of the compartments, should be considered. The correct evaluation of the best strategies cannot be separated from an accurate and transparent life cycle assessment (LCA), which would take into account both economic and environmental aspects. Herein, the most advanced recycling methods are analyzed and the issues underlying the efficient reuse and recycling of battery packs from electric vehicles are critically discussed.

1. Introduction

The lithium-ion battery (LIB) was first introduced in the market by Sony in 1991 and A&T Battery in 1992[1] to power new portable electronics tools. The first generation of LIBs used a LiCoO₂-based cathode and a carbonaceous anode, meeting the main requirements for portable electronics, i.e., gravimetric volumetric and energy densities at around 100 Wh kg⁻¹ and 250 Wh dm⁻³, respectively, as well as safety.[2] In just 10 years, LIBs reached a market share of 95% with about 10 GWh of installed energy and a corresponding business of about 10$ billion per year.[3] Later, the introduction of new alternative chemistries (LiMn₂O₄, LiFePO₄, graphite)[4] and the optimization of the battery pack brought the energy density to over 200 Wh kg⁻¹ and 350 Wh dm⁻³. These improvements led to the implementation of LIBs in the automotive sector,[5] where their actual impact in the electrical vehicles (EV) market has been growing exponentially in the past years,[6] and it is driving the continuous and renewed interest toward rechargeable batteries. The future of LIBs, indeed, seems even more brilliant and analysts agree that the penetration of LIBs in the automotive market will lead to an estimated demand >500 GWh energy production in 2025 and >2000 GWh by 2030 (Figure 1a) at the compound annual growth rate (CAGR) of 30% in the next decade.[6] The success of LIBs is also favored by cost reduction, which in turn accelerates their penetration into the automotive industry. The LIB price for kWh, indeed, dropped steadily over the past few years, from around 1000$ in 2005 to 120$ nowadays, and it is expected to reach the threshold of 100$ kWh⁻¹ in 2025.[7] The pressure from car manufacturers promoted the emergence of scale economy, which is the main reason, together with technological improvement, for price reduction (Figure 1b), which has slowed in the past years, due to critical input to cell costs, such as the raw materials, that cannot be ignored anymore, in particular for the cathode material.

According to the EU roadmap, next-generation LIBs for EV (Gen. 3b, time horizon 2025) will be still based on intercalation chemistries with C/Si composites at the negative electrode, and high-energy low-Co content positive materials, like Ni-rich layered oxides of general formula LiNiₓMn₁₋ₓCo₂O₂ (x + y + z = 1, NMC).[8] Although Co-free cathode has also been designed (NMA, where Al is introduced instead Co or spinel structures based on Mn and Ni, like LiNi₀.₅Mn₁.₅O₄), the ubiquity of Ni or Co cations is widely accepted for positive materials due to their intrinsic electrochemical properties. The exact composition of the cathode depends on several factors, e.g., the nature of the final application, the trade-off among different properties,[9] and strategic company choices. However, the positive electrode...
is getting a predominant role in the determination of the final LIB price (Figure 1c).

With the attempt of several governments (EU, USA) to enlarge the LIB production basin, the concern has turned to the availability of raw materials. Lithium primary sources are coming mainly from Australia and South America, with the largest Ni ore reserves in Australia, Brazil, Russia, and Congo being the main producer of graphite for LIBs. It is clear that the control of raw materials is as strategic as the implementation of cell production capacity. However, while the latter can be incentivized by government, the control of unevenly distributed raw materials passes through complex economic agreements with often unstable governments and uncontrolled exploitation clashes with the emerging policies of due diligence and responsible sourcing. As an example, with the cell chemistry moving forward less Co content, the European demand for raw material will grow, at least, by one order of magnitude between 2020 and 2030 (Figure 2a). The global situation is becoming even more bleak, because, at the expected CAGR, a lithium supply deficit will occur from the end of the 2020s, and it could become the main obstacle to the future EV commercialization (Figure 2b).

One of the main strategies to remedy the uneven distribution of raw materials and the future supply issues, therefore, is the development of effective and economical recycling strategies, which is one of the biggest challenges of the coming years (the so-called urban mining approach). There are many obstacles in the way of the nascent LIB recycling industry, among which the most important are 1) low collection rate and poorly regulated collection strategies; 2) heterogeneity in input feedstock due the presence of different LIB chemistries; 3) lack of knowledge of LIB chemistry by producers due to IP protection; 4) low selectivity of mechanical treatments; 5) poor global efficiency (e.g., up to now plastic and organic components are completely lost) and high cost of chemical processes; and 6) competition with battery life longevity push, which is one of the main priorities of current technological developments. Despite of these problems, the recycling industry is expected to grow about four times in 2025, and governments as well as companies are investing to pursue capacity build-up (Figure 2c). There is also growing evidence that recycling cannot be separated by the reuse concept, which, in turn, will imply a strong effort toward materials/cell/battery pack redesign.

While the state of the art of the present recycling strategies (pyrometallurgical and hydrometallurgical) has been reported in several reviews, in this Perspective, we will focus on new promising technologies and their future developments.

In Section 2, we describe two promising strategies to improve the LIB life cycle: 1) recovering lithium and transition metal cations in the form of salts and precursors via green chemistry and 2) direct electrode recycling, which heals and reconditions the cathode (or anode) material for use in new batteries.

In Section 3, we discuss the relationships between recycling and SL of large EV battery packs, remembering that the “final destiny of a battery is recycling versus. disposal.” In Section 4,
we will consider the crucial role of life cycle assessment (LCA) in
guiding technological development of the battery industry.

Finally, in Section 5, we give an outlook toward the future,
including safety issues.

2. Emerging Strategies of LIB Recycling Technology

After a short résumé of the state-of-art approaches, we will dis-
cuss some of the most promising techniques, which, in the
future, could make the overall recycling process more sustainable
from technological and economic points of view.

2.1. The Recovery of Cathode Critical Raw Materials (CRMs)

As stated earlier, recent LIBs are constituted by NMC cathodes
and graphite as anode. Once spent, they are considered hazard-
ous wastes, due to the presence of toxic heavy metal oxides.
Today, spent LIBs usually contain 5–20 wt% cobalt, 5–10 wt%
nickel, 5–7 wt% lithium, 5–10 wt% other metals (Cu, Al, Fe,
etc.), ≈15 wt% organic compounds, and ≈7 wt% plastic.\(^{17}\)
The metallurgy processes to remove metals from wastes have
to fit the complexity of the batteries and electronic nature of
the waste. Especially, they must be able to separate metals from
unconventional components, such as polymers (see Figure 3).
The most common recovery methods for the extraction of
Co, Li, Ni metals are chemical, mainly pyrometallurgical
(Europe), and inorganic acids-based hydrometallurgical
(China, South Korea) processes\(^{18}\) depending on the volume
of the received waste. They are based on metal leaching by sol-
vent or supercritical fluid extraction or chemical precipitation,
with the use of strong inorganic acids, (HCl, H\(_2\)SO\(_4\), HNO\(_3\))
and reducing agents to improve metal dissolution. However,
these processes are highly energy consuming, use harsh condi-
tions, and produce toxic materials. In addition, they do not lead to
solid precipitates due to the strong acidity of the leaching
components\(^{17–20}\)

The aforementioned chemical processes represent just the
core part of the global recycling process, as shown in Figure 3.
Indeed, the preliminary steps of 1) collection and sorting, 2) bat-
tery discharge to work under safety conditions, avoiding risk of
fires and explosions, and 3) disassembly of the end-of-life (EoL)
LIBs with the removal of the plastic and metal cases and/or the
mechanical shredding are at the same time strictly needed and
poorly optimized operations. All these steps, which are globally
defined as “pretreatments,” are generally disregarded from the
scientific community and considered just as technical aspects
(see also Section 3). At the same time, the lack of research in
this field constitutes a weak point in the global optimization
of the recycle chain and more efforts are expected in the future
to fill this gap.

Finally, despite their efficiency, such technologies are not
pretty sustainable from both the cost and environmental points

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**Figure 2.** European raw material demand growth from 2020 to 2030 a) per cathode type and b) elements\(^{13}\) c) Lithium supply demand (metric ton)\(^{6}\) and
d) recycling of LIBs by geography\(^{15}\). Reproduced with permission\(^{13}\) 2020, AABC Europe. Reproduced with permission\(^{6}\) 2020, AABC Europe. Reproduced with permission\(^{15}\) 2020, AABC Europe.
of view. Other green chemical and biological methodologies are now under development, called “high-quality recycling,” which combine high recovery rate for the critical raw materials with lower environmental footprint with respect to the traditional hydrometallurgical approach (see Figure 4). In the following paragraphs some recent and innovative approaches will be discussed, including: 1) biohydrometallurgy and 2) the use of deep eutectic solvents (DES). Concerning the more assessed methods such as traditional pyrometallurgy, traditional hydrometallurgy, and the use of organic acids, we refer to some excellent reviews available in the literature.  

2.1.1. Biohydrometallurgy

A very recent alternative proposed for the extraction of noble elements/materials from wastes is biohydrometallurgy, which is more favorable in terms of energy consumption, environmental impact, investment costs, and recovery rate. This technique...
was successfully demonstrated in the extraction from ores and in the treatment of waste electrical and electronic equipment (WEEE) to recover critical metals from printed circuit boards or electrical products like rare-earth elements, e.g., Au, Cu, Al, Pb, Ni,[24] but also other highly toxic and environmentally hazardous elements, such as As, Cd, Cr(VI), Be.[25] It consists of a bioleaching step that uses acidophilic microorganisms, which are naturally able to dissolve metals from the waste. Three kinds of microorganisms can be typically used to convert metals in the soluble ionic form, namely, 1) autotrophic and 2) heterotrophic bacteria and 3) heterotrophic fungi. The first class of bacteria is sulfur, iron–sulfur, and iron-oxidizing bacteria, such as Acidithiobacillus thiooxidans, Acidithiobacillus ferrooxidans, and Leptospirillum ferrooxidans, respectively, which act through a contact chemolithotrophic and acidophilic mechanism. They attach to the surface of metal-including solid particles and fix CO2, triggering the oxidation of iron- and sulfur-based compounds to metal sulfides, which, being soluble, decrease the environment pH and consequently dissolve the metal in the waste. In contrast, heterotrophic leaching does not require any physical contact between waste substrate and microbes. It occurs thanks to microorganisms that are capable of growing onto organic sources, secreting complexing organic acids as metabolic byproducts (e.g., citric, acetic, gluconic), which allow the metal solubilization at average pH (typically 3–5) through ligands formation. Heterotrophic microorganisms often used in bioleaching are, for example, filamentous microfungi like Aspergillus, Penicillium species, and Bacillus bacteria.[26]

The performances of biohydrometallurgy processes are affected by several operating parameters, which require proper optimization.[23e] Some of them are concerned with the microorganism metabolic activities, i.e., 1) temperature, which should be not too high to favor their accumulation and survival; 2) the nutrients, which may change depending on the types of microbes (e.g., C. violaceum for gold extraction) and whose concentration is critical for the metal biorecovery, especially in case of glycine and some salts as sodium chloride and magnesium sulfate; 3) the redox couples, such as elemental sulfur and particularly Fe ions, which may improve bacterial growth; and 4) pH of the environment and concentration of dissolved O2, which are critical for cell population increase and for aerobic microorganisms’ breathing. Other influencing factors are related to waste substrates, as the waste particle size and pulp density that have a key role in the metal chemistry and, consequently, in the metal recovery. The role of particle size was investigated, for both e-waste and batteries, in the dimensional range of 50–3400 μm during the bioleaching process of metals as Co, Zn, and Ni. It was observed that such a factor acts by modulating cell attachment, air diffusion, medium viscosity, and even medium toxicity with consequent effects on the leaching rate.[23e]

Recently, biohydrometallurgy was also tested to recover critical metals from spent LIBs, especially Li, Co, Ni, and Mn, obtaining very promising results.[17] The overall process follows three key steps: 1) pretreatment which separates the raw metals from other components as C, Al, Fe, or Cu; 2) metal extraction using organisms with microbial activity, such as heterotrophic bacteria and autotrophic fungi; and finally 3) product preparation to recover metals (as salts or oxides) or to directly prepare the cathode materials. The first lab-scale tests were conducted using Leptospirillum ferrophilum and other Fe- and S-oxidizing bacteria such as Acidithiobacillus ferrooxidans and thiooxidans onto LIB wastes containing different types of cathodes, e.g., LiFePO4, LiMn2O4 and NMC. Such microorganisms allowed Ni, Co, Mn, and Li leaching rates better than 45%, and higher extraction efficiency was obtained by optimizing culture growth conditions, e.g., lower pulp density, pH < 2, and eventual multiple sequential leaching steps.[26] Co and Li recovery efficiency as high as 99% and 80%, respectively, were obtained with copper-catalyzed processes, low pulp density (1%), and application of mixed culture of properly adapted acidophilic bacteria.[26,c]

Other studies evidenced that the fungal bioleaching is more effective than bacterial one, because they can grow even at high pH and can extract organic acids, as lactic, citric, oxalic, aspartic, or gluconic, which are able to induce chelating or complexation reactions with the metal ions.[23e-27] In this context, the most interesting species are indeed Penicillium simplicissimum and Aspergillus niger.[28] Bioleaching technology exploiting fungi was deeply explored in the past, especially for metal extraction from ores, and it was particularly efficient toward elements like Zn, Ni, Mn, and Fe. In the past years, such processes were also applied to the treatment of batteries waste. Strains of Aspergillus niger and tubigensis were tested for separation of metals in Li-ion batteries with recovery up to 100% Li and Cu, >70% for Mn, and >50% and 40% for Ni and Co, respectively,[29] and in some cases the high fungal activity led to extraction rates higher than the alternative chemical approaches.

Despite the great potential and several advantages of biohydrometallurgy, some technoeconomic challenges still exist, which have to be overcome for transfer to large-scale processes, e.g., 1) low leaching kinetics, which can be eventually improved by adding metal-ion catalysts such as Ag+,[30] and 2) metal toxicity toward Aspergillus (the only investigated fungal microorganisms in LIB recycling), which is a more critical issue. To better address the toxicity aspects, future research should check other strains in combination with the materials contained in LIB. Furthermore, new strategies to improve the fungal tolerance over long time could be to subculture fungi on the gradually increasing concentration of Li and/or other metals or to isolate fungal microorganisms, which generate genic resistance from proper metal-contaminated landfills. Another particularly innovative and not yet fully explored approach is, finally, the synergistic action of microbial communities made of properly selected bacteria and fungi.

2.1.2. Deep Eutectic Solvents (DES)

As stated earlier, state-of-the-art hydrometallurgy approaches are based on the use of mineral acids such as hydrochloric or nitric, which are strongly detrimental for the environment. For this reason, greener chemistry strategies have to be developed, as, for instance, the use of natural organic acids similar to those excreted by the fungal bioleaching or the use of DES as metal extraction agents. DES is an emerging class of green solvent mixtures, which are characterized by a huge depression in the melting point at the eutectic composition compared with those of pure single components. They are obtained by mixing hydrogen bond donors (HBDs) (e.g., urea, glycerol, carboxylic acids, acetamide, etc.) and hydrogen bond acceptors (HBAs) (typically
choline chloride, ChCl, or other quaternary ammonium chlorides, but also aminoacids such as glycine, alanine, etc.). DES is easy to prepare, highly versatile due to the wide availability of mixture components, homogeneous, generally nontoxic, biodegradable, and economically sustainable. The solubility of metal oxides in ChCl-based DES is similar to that in aqueous HCl solutions. For all these reasons, DES were investigated as a valid opportunity for novel solvometallurgical processes for metal leaching, extraction, and even electrochemical recovery. In particular, DES could be a breakthrough toward more sustainable strategies of recycling technology for spent batteries, especially Li-ion batteries and beyond.

The suitability of ChCl-based DES as leaching media for the cathode powders of spent Ni-metal hydride (MH) and Li-ion batteries with the specific topic to recover the most CRMs, e.g., Co and Ni, was recently demonstrated by Tran et al. They reported the feasibility of DES based on ChCl and ethylene glycol (ChCl:EG) in molar ratio 1:2 to leach Li and Co (III) from LiCoO2 cathode with yield higher than 90%, by setting high temperatures (>200 °C) and long treatment times (>72 h). Similar leaching rates were obtained for the same material using different DES, such as ChCl–citric acid, ChCl–urea, and eventually Al and Cu as reducing agents for Co (III). The leaching process requires higher temperatures (>160 °C) than those used with conventional mineral acids like HCl and HNO3, which need less energy to dissolve the metal oxides. In contrast, DES act without any evolution of toxic and dangerous gases, such as, for instance, Cl2 in case of hydrochloric acid. Shorter dissolution times of only 15 min, lower amounts of solvent, and temperatures lower than 90 °C were successfully explored by replacing the HBA component in the ChCl-based DES. In particular, eutectic mixtures composed of ChCl and p-toluene sulfonic acid, and related aqueous dilutions, allowed to avoid any other reducing agents and obtain Co recovery efficiency above 94% with consequent potential economic benefits.

Indeed, recycling of cathodes including multiple transition metal ions (e.g., NMC) is more complex, as far as the separate extraction of metals is concerned, due to their similar chemical properties. In this case, the selectivity of DES is a relevant factor. The solvent extraction fractionation of a Li–Mn–Ni–Co matrix was recently discussed. Such a process was based on the use of N,N,N',N'-tetra-n-octyldiglycolamide (TODGA) diluted in an ionic liquid (1-butyl-3-methylimidazolium bis(trifluoromethanesulfonylimidide) and was used to first extract Mn with a rate of 99% in one step with a separation factor in the range of 40/60 for Mn/Co. Subsequently, more than 90% of Co was recovered by means of alkyl phosphonium chloride and finally Ni and Li via a DES based on lidocaine and oxalic acid.

Due to the large amount of possible HBD/HBA combinations to give DES potentially suitable as lixiviants for the metals of LIBs, a prescreening strategy is mandatory to select the best mixture for recycling spent batteries. Recently, DES reducing power was proposed as a selectivity criterion for the metal recovery. Leaching and consequent metal oxide dissolution by DES takes place through metal complexation via HBD. Here, solution pH and HBD coordination strength play a very important role. Typically, a lower pH of DES with poorly complexing HBDs favors high solubility, which is expected to depend on the lattice energy and Gibbs free energy of formation of complexes. Higher lattice energy results in lower solubility. However, the complexing action seems to have a greater effect on oxide solubility. Indeed, stronger HBDs as ChCl better dissolve metal oxides by complexion reactions that occur onto the surface, followed by ligand exchange for Cl− in the solvent, which is generally the most important ligand. An exception is ChCl:oxalic acid, where the precipitation of metal complexes occurs through the strong interaction of the metal with the oxalate anion. Oxalic acid is highly selective toward Ni, Co, and Mn, so it can be particularly effective in the extraction processes involving the LIB cathode materials.

2.2. Direct Electrode Recycling

An alternative (or additional) strategy to chemical recovery of elements is the direct recycling (see Figure 3) of selected battery components, consisting of the removal of cathode or anode material from the electrode for reuse in a remanufactured Li-ion device. This approach implies the reconditioning of the active components, which underwent to degradation during the cell use, causing performance failure. Generally, in the case of cathodes, the performance decay is ascribed to the progressive degradation of the material structure and composition, due to irreversible metal-ion migration into the electrolyte (especially Ni and Li), loss of crystallinity, and irreversible evolution of the microstructure. The carbon-based anode is also affected by irreversible Li intercalation and exfoliation.

These compartments can, in principle, be extracted from the battery and reincorporated into a new electrode with minimal modification to restore both crystal structure and morphology. This revitalization typically requires the correction of the lithium stoichiometry to compensate losses due to material degradation.

The cathode powder is generally reconditioned through conventional solid-state reactions by adding external Li sources (e.g., carbonate or sulfate). More recently, a new approach of direct recycling, namely, cathode healing, was introduced to produce low-cost and battery-grade recycled electrode materials. The proposed strategy is based on soft and nondestructive chemical treatments of the recovered active materials with concentrated Li-ion solutions, through hydrothermal process and subsequent heating steps. The work demonstrated the use of healing to regenerate crystal structure, capacity, and lifetime of used NMC523 and LiFePO4 with limited cost (2.50 $ Kg−1used-cathodes). Despite less interest being devoted to anode materials due to their lower recovery values, the successful reuse of spent graphite anodes was also proved with functional performances comparable with those of pristine material. These results suggest that direct recycling could be more effective in case of lower-value cathodes such as LiMn2O4 and LiFePO4, whose manufacturing costs are the major contributions to the overall production ones.

With respect to traditional recycling, direct recycling has the advantage that the demanding and time-consuming pretreatments, metal–ion separation, purification, and isolation can be avoided (but for the separator and electrolyte). While highly selective direct recycling approach is extremely appealing in view of an effective closed-loop circular economy scheme, at the same time,
it is not ready implementable in the today scenario, where EoL LIBs represent the majority of waste products. Indeed, current batteries are not designed to be opened, and partial disassembly and subsequent reassembly are not possible without compromising their functional state. Moreover, this regeneration approach is by far less considered from the research point of view with respect to recycling and SL methods, and only few, even if relevant, research lines are today active in this sense.\cite{44} This approach requires a complete redesign of the LIBs and the development of new materials (such as binders) that allow for the selective extraction of components and their reparation and sets the goal for long-term actions in the development of the batteries of the future. Furthermore, the potential success of direct recycling is strictly related to other important obstacles, such as the battery state of charge (SOC), which must not be too low for an efficient process, and the low flexibility in case of the multielement/multioxide composition of the cathode.

3. Recycling Versus SL

Recycling and SL are often considered as alternative routes for the EoL LIBs, even if the final competition is indeed recycling versus disposal and, at best, SL is an intermediate stage which can influence pack/module/cell depreciation or even reduce the capital cost. Both sustainable SL and recycling exploitation are today at their initial stage in terms of sustainable technologies, regulation, and planning. The LIBs today on the market are not designed for ease and cost-effective implementation either for SL or for recycling (both traditional and direct). Thus, the identification of the best strategy for the destination of today EoL LIBs is not trivial and depends on many factors such as: 1) cell chemistry, 2) size, 3) state of health (SoH) and SoC, 4) local regulations for EoL LIBs disposal and recovery, 5) transport and logistics, and 6) cost of electricity. The sustainability and the economic convenience of each process must be carefully evaluated through LCA (see Section 4). Due to the complexity of each topic, here, we propose only a general discussion to highlight the most relevant aspects affecting the SL and recycling issues. For a policy/market analysis on the reuse of EV batteries for integrating renewable energy into stationary grid, see the study by Gur et al.\cite{45}

3.1. Recycling Issues

Recycling of EoL LIBs is already implemented at the industrial level by many companies, mainly located in the LIB producers-based countries, as discussed in the Introduction (i.e., China [GEM] and South Korea [SungEel], followed by USA [Retrieval Technologies], Europe [Umicore], and Australia [Envirostream]).\cite{46} Although the recycling rates are often claimed to be low and insufficient, actually the figures for LIBs are as good as demonstrated by the spread of specialized recycling companies all over the world. What still has to be improved is chiefly 1) the economic/environmental sustainability, as discussed before and 2) the collection rate, which is strongly dependent on local regulations and policies and challenges of technological issues.

As stated, up to now, recycling was chiefly based on pyrometallurgical (Europe) and inorganic acids-based hydrometallurgical (China, South Korea) processes,\cite{47} depending on the volume of the received waste. At the same time, the technological processes to replace these high-efficiency environmental impact technologies are already available, as demonstrated by the wide literature on high-quality recycling, combining high recovery rate for the CRMs with low environmental footprint. Besides the biobiohydrometallurgical and DES-based processes highlighted in Section 2, other approaches characterized by low environmental impact are under development, including mild mechanochemistry,\cite{48,49} ultrasound,\cite{50} and microwave-assisted\cite{51} mild acid leaching, bioelectrometallurgy,\cite{52} coextraction, and coprecipitation.\cite{53}

Indeed, these new processes can ensure a lower environmental footprint with respect to the traditional hydrometallurgical approach, maintaining high CRM recovery yields.\cite{18,19,21}

Thus, today, the core activity of recycling does not represent a technological challenge. Again, the major problem related to high-efficiency recycling is the economic sustainability of these new green processes, which is closely related to the volume economy. The transition from the traditional hydrometallurgical processes to greener technologies can be promoted with adequate incentive policies and regulations.

As stated earlier, what still has to be improved to increase the quality and the efficiency of the overall recycling process are the former steps: collection rate and “pretreatments,” namely, all the steps needed to move from the as-collected EoL LIB to the black mass ready to go to recycling. Generally, these steps included the complete discharge of the EoL LIBs, the removal of the plastic and metal cases, and the separation of different compartments.\cite{54} An alternative approach is the grinding of the whole device without any preliminary separation and dismantling. Only few studies have been dedicated to this topic that has the potential to highly impact the global efficiency and sustainability of the recycling process. Today, the greatest obstacle in the development and implementation of efficient pretreatments is represented by the wide variety of chemistries and designs of LIBs, especially for portable devices, and the wide range of sizes of batteries and battery packs, related to the different market segments (from portable devices to automotive). Moreover, it must be recalled that there is a growing tendency to embody a nonremovable battery in the device, further complicating the proper collection of both the batteries and the electronic parts. Some of these aspects should be addressed with proper regulations, requiring a clear identification of the battery chemistry, the possibility for the end user to separate the battery from the devices, without the need of risky or specialized operations, and the establishment of dedicated collecting stations and recycling routes.

3.2. SL Issues

While recycling received relevant attention from the battery community, as attested by the huge number of publications per year in this field, reuse and SL are still less considered from the scientific point of view. Indeed, the first initiatives toward SL of LIBs were proposed by some electric car producers, such as Nissan (partnered with Eaton for xStorage),\cite{55} BMW,\cite{56} Renault,\cite{57} and General Motors.\cite{58} Similar initiatives are under
way in China, based on the agreement between the administration and the major LIB producer companies, to exploit EoL LIB for power stations.\[90\] Proof of concepts can be found also in Europe, e.g., the Johann Cruyff Arena.\[68\] Such initiatives mark the pace for new paradigms of energy exploitation, in particular for renewable electricity management, and also promote the active engagement of end-users and citizenship in the issue of reuse and recycling.

Up to now, the majority of works in the SL field were based on simulations and not on experimental data, which are still lacking on a large scale.\[61–63\] The environmental sustainability of SL has been assessed, based on different models and markets. Therefore, SL should have a beneficial effect on both the footprint of the EV and that of the SL application, e.g., renewable energy storage.\[64\] However, the economic and technical viability still has to be fully addressed, and different results have been reported based on the considered models and boundary conditions.\[61,62,65,66\]

SL of EV LIBs consists of reusing a pack (or a part of it) that no longer satisfies the stringent requirements of the automotive sector, for applications in less demanding fields (e.g., smart grids, power stations in combination with photovoltaic systems, etc.). Indeed, SL extends the life of the battery and therefore reduces the depreciation costs. In turn, the cost is not a fixed parameter but also positively depends on the production volumes. In contrast, the increase in production leads to a greater demand for CRM, such as Co, Cu, Li, and Ni, as outlined in the Introduction\[67\] with a consequent increase in costs, which can be compensated to some extent by effective recycling policies. This must be considered when the economic viability of SL is analyzed.

Several fields of SL applications have been evaluated, ranging from residential battery energy storage systems\[68,69\] microgrids,\[70,71\] large-scale grids\[66,72\] and energy-storage systems for remote rural areas.\[70\] Indeed, final application also contributes to determining the economic sustainability of SL, but again it is strongly dependent on the local cost of electric energy, local policies and incentives for the exploitation of renewable energies, etc.

Some general guidelines to evaluate the advantages of SL are the following.

The minimization of the balance of system. The collection and repurposing of the EoL LIBs must be carefully assessed to minimize the cost of SL implementation. This includes several steps: the creation of an efficient network of collecting points, transportation, securing of the EoL LIBs, dismantling, full mechanical and electrochemical characterization, and partial remanufacturing. EV packs are made of modules built up by many cells, and a battery monitoring system (BMS) is embodied for the monitoring of the pack and the single cells’ performances. The degradation of the cells in the pack is not uniform, due to temperature gradients, connectivity scheme, and exploitation, and the different cells undergo different aging. Accordingly, the first step for repurposing is the dismantling of the pack/modules with the identification of the degraded cells and their substitution/elimination for recycling. The remaining cells/modules must be fully characterized in terms of SoH and SoC and then destined to specific SL applications. These operations, remanufacturing and testing, are both time and cost demanding. The development of new pack schemes with BMS systems able to monitor single-cell SoH/SoC and suited for easy removal/substitution of single units can favor the SL exploitation. Such guidelines were recently drawn in the proposal for the new EU battery directive that is expected to update the EU Battery Directive 2006/66 by July 2023.\[73\]

The identification of the target market. The identification of the best and most profitable SL market is a crucial issue. Some relevant aspects such as the availability of related structures and ancillary services and the electricity cost must be accounted in this analysis. Again, environmentally friendly policies, such as the Europe Green Deal, constitute the best framework for the exploitation of SL LIBs in combination with renewable energy sources.\[74\]

The full knowledge of the battery performances during the first life (FL) and at the EoL, and reliable predictions of SL lifetime and performances, based on the application. Monitoring the battery aging during the FL is essential to determine the degradation level and the best SL application. The full SoH/SoC characterization, with the definition of EoL, and the introduction of protocols and standards, will represent the core scientific and technological issue related to the concept of SL. The first time this was introduced, for Ni-MH batteries in EVs and hybrid vehicles, the end of FL was defined when two criteria were met: battery capacity was 80% of the initial nominal value and battery power was 80% of the pristine battery power capability.\[75\] Nevertheless, it must be considered that Ni-MH batteries are significantly different from LIBs in terms of chemistry, energy, and power density; thus, the first step for the optimization of FL and SL applications should be the definition of new dedicated EoL parameters for this technology, in case of also diversifying the parameters on the basis of the specific chemistries involved.

Besides the scientific/technical assessment, optimal exploitation of the SL requires considering the active business model. In fact, it is possible to make more use of the FL rather than SL depending on the battery ownership model (BOM) and the definition of the extended producer responsibility, e.g., if the battery belongs to the EV owner, to the EV manufacturer, or even to a third party. The exploitation of FL can vary significantly among these three models, leading to EoL LIBs with different SoH and SoC. Up to now, due to the novelty of EV LIB technology, experimental data on degradation rate during the FL are scarce and partial; nevertheless, this constitutes a crucial point for the proper economic viability of SL approach; thus, more efforts in the definition of protocols for analysis and monitoring of LIBs’ aging are needed.\[76,77\]

The need for standardization of parameters defining the life-span of FL and SL, as well as of analytical techniques able to characterize SoH/SoC, is clearly highlighted in consortia and platforms manifestos (e.g., EUROBAT,\[78\] Battery 2030+\[79\]) and technical literature, clearly stating that the priority in this field comes directly from the scientific community, cars and battery producers, and policymakers.\[80–85\]

A harmonized context of norms, regulations, and environmental policies to incentivize the collection and reuse of LIBs. EU is acknowledging the necessity of a dedicated regulation framework for the LIB category. The regulation currently in force is the Battery Directive EU 2006/66, for which collection and recycling practices are defined for Pb-, Cd-, and Ni-based energy-storage devices.\[86\] The evaluation report on the results
of such a directive, that appeared in 2019, clearly indicates the necessity to explicitly account for the LIBs as a separate category of devices,\cite{87} and efforts in this sense are addressed in the new text, which was recently proposed for approval at the EU Parliament.\cite{73} This new proposal fits in the more general context of the European Green Deal, promoting the ambitious zero greenhouse gases in 2050. Similar policies are being adopted also in the US with the aim to decrease the dependency on CRMs, promoting reuse and recycle through improved collection, sorting, storage, and transport of spent LIBs.\cite{88,89}

4. Life Cycle Assessment

The major driving force pushing toward emerging, more sustainable, and scalable recycling technologies, as those discussed in the previous sections, is the urgent need to balance the process environmental and economic viability. Two critical aspects must be considered. On one hand, the use of the current recycling technologies to treat the ever more-growing volumes of batteries from the electric vehicles will not necessarily lead to a reduction of greenhouse-gas emissions compared with primary manufacturing. On the other hand, such recycling processes have been tailored to recover chiefly cobalt, which is the most important CRM. However, this element is doomed to be replaced in the next-generation cathodes, making these methods less convenient because of the expected lower economic value of incoming materials, e.g., manganese, aluminum, nickel, etc. In this context, the study of the entire cradle-to-gate story of batteries by means of life cycle analysis (LCA) is fundamental to concretely address the economic, social, and environmental impacts of the whole battery value chain, from production to recycling/disposal.\cite{90-92}

LCA investigations are not yet abundant in literature. Furthermore, considering fast research development in the field, the information currently available has to be continuously updated and harmonized. Very recently, the Argonne National Laboratory released the Greenhouse Gases, Regulated Emissions and Energy Use in Transportation (GREET) software,\cite{93} whose database is frequently updated with primary inputs from materials producers and battery manufacturers. This model was able to highlight the most critical contributors to the energy and environmental impacts of new-generation batteries, such as those including NMC cathodes. In particular, LCA results showed that, in typical NMC-based batteries, the most significant contributors are the active material powder, aluminum from the current collector, and the cell production process with the highest percentages of total energy use (>36%), greenhouse gases (GHG) emissions (>39%), SOx and NOx emission (more than 63 and 47%, respectively), and water consumption (>31%). Graphite, copper, and Li salt (generally LiPF6) also affect the environmental impact, even if to a lesser extent.\cite{94}

The main key factors affecting LCA results are 1) the production route of cathode-active material, due to the complex synthetic methods involving several energy-intensive and consuming steps, such as precursor coprecipitation and subsequent calcination; 2) the production of the cell, which consists of several energy-intensive processes, from the slurry preparation to electrode production by heating for solvent removal and to cell assembly and conditioning in dry room; 3) LIB full supply chain, in terms of large-scale production logistics and geolocalization of material sourcing, chiefly because of the electricity cost for element extraction, manufacturing, and transportation; 4) knowledge gaps, deriving from nonexistent or poor data on battery materials other than NMC, which often require strong data approximation; and, finally; 5) the need for comparison, which requires not only the harmonization of the results reported in different LCA studies, but also rationalizing the main differences among the various investigations, to make data interpretation clearer.\cite{94}

Ciez and Whitacre\cite{95} used life cycle assessment and process-based cost models to investigate GHG emissions, energy inputs, and overall cost of the recycling processes, making a performance comparison among pyrometallurgy, hydrometallurgy, and direct recycling, in case of batteries based on NMC, LiFePO4, and lithium nickel cobalt aluminum (NCA) oxide. What emerged is that recycling via both pyro- and hydrometallurgy resulted in a net increase in GHG emission for all the three examined chemistries, whereas direct recycling had great potential to reduce CO2 emissions (from the chemical treatment and, at a lower extent, from transportation of the starting materials and collection of spent batteries), being also economically competitive. In addition, both hydrometallurgy and direct recycling required a lower energy consumption than pyrometallurgy. In the latter technology, the yield of cathode recover was strongly correlated with chemistry and cell format. It was demonstrated that the environmental benefits of direct recycling in terms of avoiding CO2 emissions are higher for NMC than NCA, and cylindrical cells scored better than pouch ones.

The economic viability of recycling technologies is also linked to the market demand for metals that grows continuously. In this regard, the cost assessment for cathode reconditioning affects the constant price volatility of both Li and Co. In case of Li, the cost analysis takes into account the reconditioning process during cathode reconditioning and consequently the price and the required amount of Li2CO3.\cite{95} The case of cobalt is a little more complicated because of its social as well as economic implications. In fact, cobalt recovery assumes strategic significance not only for aspects related to the critical supply chain, but also for unethical labor practices, e.g., exploitation of children and health issues in the population surrounding the mining sites.\cite{96}

While the data collection is going to increase rapidly and the key factors in the LCA and technoeconomic analysis are well defined, the recycling programs on the Li-ion batteries, all things considered, are still in an embryonic stage, contrary to the lead-acid devices, whose recycling is fully consolidated, simple, and profitable.\cite{96} This uncertainty is caused by poor regulations, which, by the way, change from one nation to another and still have to be implemented with strong policies, such as, for example, 1) enabling the collection, now limited to small packs used in consumer electronics and also to the large batteries coming from the automotive sector, and 2) incentivizing collection by means of refundable deposits with battery purchase.

5. A Look at the Future

Production of LIBs is increasing every year, together with the acceptance of EVs. This will lead to large volumes of retired
LIBs and shortages of CRM resources. A multilevel and multipurpose rational reuse of batteries is the first issue to be solved for reducing energy consumption and improving efficiency of energy utilization. However, it should be remembered again that the alternative is recycling versus disposal, the former being the desirable fate of every battery. While extended recycling is undoubtedly well perceived by the public opinion, it can be made economically acceptable by combining proper technical developments, regulations, and incentives. Therefore, a higher efficiency in the recycling of Li-ion batteries is necessary to decrease the impact of the fast-developing battery industry on our environment. The commonly used pyrometallurgical processes cannot provide sustainable recycling yet, even if some of them are already integrating hydrometallurgical steps to obtain high-purity metal salts. It should be also noted that mechanical pretreatments and separation processes, which are among the most crucial steps of recycling, are not well optimized to date.

At present, Li-ion batteries are designed, built, and managed without having recycling in mind. With the inevitable increase in EV production over the coming years, it is vital that properly scalable recycling schemes will be implemented and considered already during the design stage of the batteries of the future. This will require a change of the 3 R paradigm from “Reduce, Reuse, Recycle” to “Redesign, Reuse, Recycle,”[34] which will impact environment, sustainability, and safety of the new-generation batteries. A summary of the today scenario and future perspectives is shown in Figure 5, according to the modified Lansink ladder based on the new 3 R scheme.

5.1. From the Technical Point of View

Recycling-oriented design will allow more sophisticated approaches to battery recovery through automated disassembly, including better sorting technologies, methods for separating electrode materials, and increased process flexibility, allowing for direct recycling approach. Indeed, this process will have to go hand in hand with standardization of batteries. Among the technological areas of intervention, one is the development of module-less packs with cells that can be easily separated and processed. Automation of this process, particularly using robotics and artificial intelligence, is an area of significant interest. Another area is the use of adhesives and binders, which can simplify both macroscopic disassembly (cell separation and opening) and microscopic electrode delamination (separating active material from current collectors).[97] The reduction/elimination of adhesives is obviously desirable.

5.2. From the Environmental Point of View

While pyrometallurgical and hydrometallurgical processes do not significantly reduce life cycle greenhouse gas emissions, new high-quality recycling procedures and, even more, direct

![Figure 5](https://www.advancedsciencenews.com/)

*Figure 5. Lansink ladder for the EoL LIB industry. On the right side, in gray, the today scenario and in blue, the advisable perspective for the improved future economy and optimized value chain.*
compartment (e.g., cathode) recycling have the potential to reduce emissions and be economically competitive.\(^\text{[95]}\)

### 5.3. From the Safety Point of View

For both SL and recycling, it is essential to know the state of the battery/pack, as well as of each individual cell. Besides SoH and SoC, the state of safety (SoS) is becoming important to decide on the continued use or disposal. For example, in a crashed EV, when it is known that some cells of the battery are still safe, they can be reused or adopted for SL application. To this aim, it is mandatory to identify suitable and standardized safety parameters and to develop new nondestructive selective testing methods for cell diagnosis. The SoH, SoC, SoS, as well as full information on the chemistry of the battery, should define the so-called “battery passport,” identifying each individual cell, allowing for the battery traceability along the whole value chain, and constituting the first essential element for optimized SL and recycling routes.\(^\text{[73,80]}\)

An important point is how to enable the safe disassembly of the battery/pack into the individual cells, if the battery cannot be reused as a whole. For the process of disassembling—in a recycling company as well as in an emergency situation, e.g., after an accident—besides fast testing/diagnosis methods, proper information on product design and safety for workers is needed. This will imply the creation of proper databases containing relevant safety data on battery applications.

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### Conflict of Interest

The authors declare no conflict of interest.

### Keywords

batteries, electric vehicles, recycling, second life

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Chiara Ferrara obtained her Ph.D. in chemistry from the University of Pavia (Italy) and the École Normale Supérieure de Lyon (France) in 2014. She is currently an associate researcher at the University of Milano Bicocca, Department of Materials Science. Her research interests cover the structural investigation of functional materials for high-performing energy-storage devices, including rechargeable batteries, supercapacitors, and fuel cells.

Riccardo Ruffo is an associate professor at the University of Milano Bicocca. He received his Master’s in chemistry (1997), Master’s in materials science (2001), and Ph.D. in material science (2004). In 2008 and 2014, he was visiting scholar at Stanford University. His research interests cover several fields of applied electrochemistry such as active materials for energy storage (rechargeable batteries and supercaps) and production (fuel cells) and for electrochromic applications (electroactive polymers).

Eliana Quartarone is an associate professor of electrochemistry and physical chemistry at the Department of Chemistry of the University of Pavia. Her research field is focused on the development of advanced materials (synthesis and characterization) for energetics and electrochemistry and specifically Li batteries, PEMFCs, and energy harvesting.

Piercarlo Mustarelli is a full professor of physical chemistry and electrochemistry at the Department of Materials Science of the University of Milano-Bicocca. He is also the Chair of the INSTM Italian National Reference Centre for Electrochemical Energy Storage. His research interests span from materials for batteries and fuel cells, to biomaterials, to solid-state NMR characterization.