# A sustainable raw chemical process for metal recovery from unsorted spent lithium-ion batteries.

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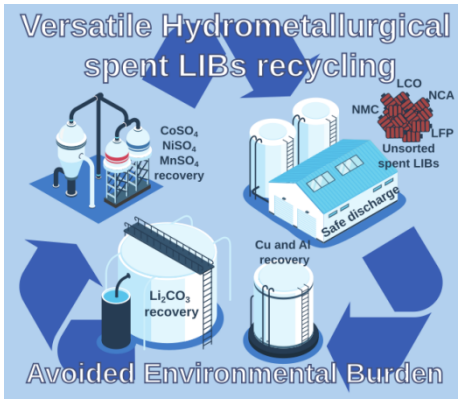
**KEYWORDS:** Sustainable process for End-of-Life Lithium-Ion Batteries; Recovery of Critical Raw Materials; Circular Economy; Hydrometallurgical treatment;

## GRAPHICAL ABSTRACT:




A varsatile hydrometallurgical lithium battery recycling process recovers lithium and other metals (Cu, Al, Co, Ni, Mn), with high efficiency and with environmental benefits, from real unsorted EoL-LIBs addressing the sorting issue.

## ABSTRACT

Waste lithium battery cathode chemistry panorama changes continuously following progressively the introduction on the market of novel material-based batteries. Anyway, it is safe to assume that lithium will not be replaced by sodium or potassium in the short term, indeed mobile and automotive applications demand is forecasted to grow. One of the most important topic in the recycling field of LIBs is the sorting by chemical composition. In this study a simple, versatile and sustainable hydrometallurgical process for the recycling of unsorted LIBs, such as LCO, NMC, NCA, and LFP, provided by a real disposal plant was presented. Lithium is recovered as carbonate salt with a yield of 85% and a purity of 95%, regardless of the cathode chemistry, followed by recovery of the other metals as secondary raw material in a form that can be further processed. Current collectors and graphite are recovered in pure form for recycling. Life Cycle Assessment (LCA) study shows that the balance between the environmental impact of the process and the credits from the recovery of secondary resources is overall beneficial.


## 1. Introduction

Since 1991, Lithium-Ion Batteries (LIBs) have powered portable electronic devices due to their high power, energy density, low recharging time, and cost. LIBs are now crucial for the energy transition to renewable sources, helping reduce greenhouse gas emissions.(Calvin et al., 2023) The growth of electric vehicles (EVs) relies on LIB performance, with the European Union planning to phase out internal combustion engine vehicles by 2035.(Friedman-Rudovsky, 2011; Greim et al., 2020) However, the increasing demand for LIBs poses production and disposal challenges, risking unsustainability without recycling strategies. Several Governments promotes a circular economy to minimize waste and resource exploitation, for example the

European Commission's Green Deal.(Speirs et al., 2014) Despite efforts to improve recycling processes, End-of-Life LIBs (EoL-LIBs) remain difficult to recycle due to their varied forms, the number of components, and different chemistries.(Graedel et al., 2011)

The recycling of EoL-LIBs typically involves pyrometallurgy, hydrometallurgy and direct recycling processes. Pyrometallurgical treatments include four steps: heating to 350 °C to evaporate electrolytes and to inertize the LIBs, mechanical milling to recover metal scraps, heating over 600 °C to form alloys and mixed oxides, and leaching the alloys with acids for refinement of Co, Ni and Cu. Despite optimizations, pyrometallurgy dissipates lithium, emits hazardous gases, and needs a lot of thermal energy.(Lan, 2025)

Hydrometallurgical recycling has become the predominant method in the industry, particularly in Asian countries. The process generally involves two main steps: the first is leaching with opportune reagents, such as inorganic acids, organic acids, alkali, and eventual reducing agents, are added to convert the metals oxides in the cathode active materials into metal ions in a solution. The second step involves separating and recovering valuable metals from the leaching solution through various chemical methods, including precipitation, solvent extraction, and electrolytic deposition. Hydrometallurgical treatments need a lot of chemicals and show high water consumption and waste water production, limiting their worldwide spread.(Jung, 2021) To address these drawbacks, solvometallurgical approaches are suggested due to the potential use of Ion Liquids and the safe and environmental friendly solvents like Deep Eutectic Solvents (DESs).(Tran et al., 2019; Yasa et al., 2023; Zhang et al., 2022)

Recovering cathode and anode active materials directly is promising because it takes a shortcut to obtain materials with a near-perfect crystal structure, but shows drawbacks such as high energy consumption, high-cost equipment, reagents consumption, and difficult to full recover the

crystalline structure and chemical composition to restore the electrochemical performances.(Lan, 2025)

Furthermore, both hydrometallurgical and direct recycling processes need an inertization step to avoid short-circuits and fires due to the residual charges in batteries. Aqueous electrolyte discharge, thermal venting, and manual discharge are the common methods used in the state-of-the-art (SoA).(Gao et al., 2024)

While most of the experimental articles were focused only on one step of the EoL-LIBs treatment, the present work reports the results of a lab-scale full hydrometallurgical process, based on versatile and raw chemical treatments, which covers each part of the EoL-LIBs recycling: from the safe deactivation to the recover of valuable metals. Furthermore, the process was able to treat any kind of battery's shape and chemistry provided by a real disposal plant, without any sorting step. Furthermore, a deep Life Cycle Assessment (LCA) and a preliminary economic assessment are evaluated to highlight the sustainability of the process. The article reports part of the research work covered by the patent WO2019/150403A1 (Vizza et al., 2019).

## 2. Materials and methods

All chemicals are bought from Sigma Aldrich or Merck in high purity form to avoid metals contamination and used without any further purification.

The used EoL-LIBs were ICR186x0 and pouch-type cells coming from a real disposal plant, thanks to the collaboration with Haiki Cobat spa in this research work. The batteries were not sorted by chemistry (LCO, NMC, NCA, LFP, etc) but only for the fact that were LIBs. Then, the samples were a mean "photography" of the actual EoL-LIBs stored in a real disposal plant. Furthermore, we manually removed the battery management system (BMS) to expose the electrodes if needed, such as for the pouch-type equipped in several 2010's smartphones.

Because of our intention to treat unsorted EoL-LIBs, the reagents were chosen to be versatile for each kind of battery's chemistry.

A typical experiment was performed by selecting mixed ICR186x0 and pouch-type cells accounting for about 600 g. All reagents, products, and yields were referred to the actual amount of batteries or materials.

ICP-OES was conducted on the samples to evaluate the metal content following the "EPA 6020 A 2007" method.

The proposed process can be divided into 6 steps, as reported in Figure 1: safe deactivation (inertization), active material separation and current collectors recovery, black mass leaching, lithium separation by precipitation, and other metals recovery.

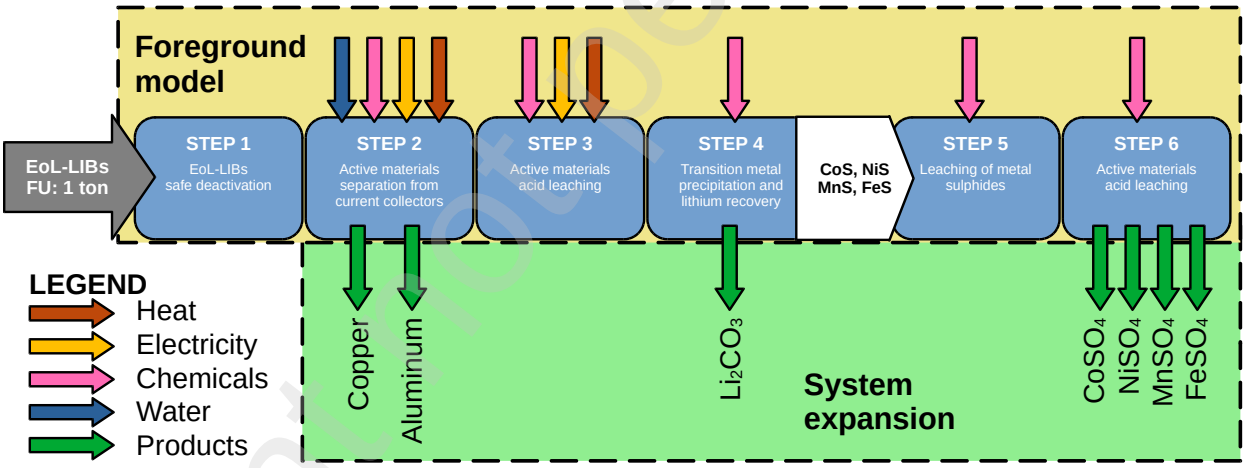



**Figure 1.** Scheme of the proposed process with the foreground and expansion systems used in the LCA. The arrows indicate the material and energy flows.

### 2.1 EoL-LiBs safe deactivation

The EoL-LIB discharging was conducted immersing the batteries in brass shreds for several hours. The fire risk increases with the increasing of internal temperature of the batteries, then the temperature behavior was monitored. The discharging was conducted immersing the batteries in

brass shreds for several hours. All other technical details were reported in the Supporting information part.

### 2.2 Active materials separation from current collectors

After the complete discharge, the batteries (ca. 600 g) were manually disassembled, removed the shells, unrolled, and cut to pieces about 10×10 mm (ca. 480 g). Active material separation from current collectors study was performed by immersing 5 g of those pieces (Al foil with cathode active material, Cu foil with graphite, and polypropilene separator sheets) in 25 ml of four different solutions: a) 0.5M $H_2SO_4$, b) 3M HCOOH, c) 1M HCOOH, and d) ultrapure water. Each test was repeated four times. All samples were gently magnetically stirred for 1 hour and maintained at room temperature (ca. 25 °C). After, the leftover foils and separator were filtered with a 3mm grid in order to obtain the dispersion of the powders phase, mainly composed by graphite and lithium-metals oxides also called black mass, and the foils phase, mainly composed by current collectors and separators. The leftover foils and filtered powders phases were dissolved in *aqua regia* (solid:liquid ratio 1:15 g:ml) for ICP-OES analysis in order to determine the efficiency of the removal and foil contamination in the powders.

### 2.3 Active materials acidic leaching

The powder materials (ca. 260 g), obtained using a 3M HCOOH solution as described in the previous section, were dissolved with a proper leaching solution in order to recovery each metals. Cathodes based on $LiMO_2$ (where M can be either Co, Ni, Mn) usually need both an acid and a reductant in the formulation to ensure the complete dissolution of the materials, due to the presence of Co(III) species (Antolini, 2004; Dahéron et al., 2009; Zuo et al., 2023). Formic and hydrochloric acids were chosen due to the reducing activity of the former, the strong acidity and

chloride complexing properties of the latter. Different acid concentrations were explored. A given quantity of black mass (usually 5-7 g) was weighed and added into a round-bottomed flask in a heating jacket and equipped with a bubble reflux column. A quantity of leaching solution (20-30 ml) was added accordingly to the weight of the black mass (1:4 g:ml) and the solution was refluxed for 1h under magnetic stirring. Then the solid residue (ca. 1.5 g) was filtered off and washed while the liquid part was analyzed by ICP-OES to evaluate the metal ions content. The solid residue, made essentially by graphite, was then dried weighed and leached with ca. 22 ml of *aqua regia* (1:3 $HNO_3$:HCl, solid-to-liquid ratio 1:15 g:ml) at reflux and analyzed by ICP-OES to evaluate the metals impurities.

### 2.4 Transition metal precipitation and lithium recovery

The recovery of lithium from the leaching solution was performed first by precipitation of the transition metals with sodium sulfide ($Na_2S \cdot 9H_2O$, Sigma Aldrich, ≥ 98%), then by precipitation of lithium carbonate with potassium carbonate (Sigma Aldrich, ≥99%).

To an amount of the leaching solution (40 ml) was added a sodium hydroxide solution (1M) to obtain a pH around 6, then a solution of sodium sulfide (0,1M) was added in gradually and decanted till the further addition of sulfide did not induce any more precipitation. After 1h of digestion under stirring at room temperature, the precipitate was separated by filtration obtaining a colorless solution. The transition metal sulfides mixture was washed, dried, and then weighed to evaluate the efficiency. The solution containing lithium was then concentrated (to about one fourth) and sodium chloride salt was precipitated out of the solution, then 5 grams of $K_2CO_3$ were added under stirring keeping the solution at 90°C, indeed lithium carbonate gets less soluble at higher temperatures (Cheng et al., 2013; Stober, 1986). After 20 min digestion, a white powdery precipitate of $Li_2CO_3$ was formed, which was filtered and washed with small portions

of boiling water, then dried, weighed, and analyzed by ICP-OES to determine $Na^+$ and $K^+$ contamination. The ICP-OES analysis showed a yield of 85% with a purity of 95%.

### 2.5 Metals sulfides leaching

A typical test was done using ca. 10 g of precipitate and 15 ml of hydrogen peroxide and sulfuric acid solution (25%wt. hydrogen peroxide and 25 %wt. sulfuric acid) obtaining the oxidation of sulfides to sulfates and the full dissolution of the precipitate, resulting in a transparent solution. The treatment mimes the oxygen-sulphuric acid process, which is commonly used in the mining industry.(Moats and Davenport, 2014)

### 2.6 Metals sulfates separation

Extraction in organic solvents mediated by an appropriate chelating agent is a common methods used in the mining industry for metal refining. A 0.3M solution of bis(2,4,4-trimethylpentyl)phosphonic acid (commercially known as Cyanex 272) in organic solvent (kerosene) (Moats and Davenport, 2014) was used to proof the feasibility of the method extracting the cobalt sulphate.(Kang et al., 2010; Swain et al., 2007)

In a typical test, the pH of ca. 15 ml of the previous solution was adjusted to 4.3 using acetic tampon (1 M acetic acid) and 20%wt. NaOH solution. After, the solution was decanted in a separation funnel containing ca. 15 ml 0.3 M Cyanex272 in kerosene. Then, the funnel was shacked at room temperature and the phases were separated after 5 minutes. Cobalt sulphate was recovered washing the organic phase with 5 ml 2 M solphuric acid aqueous solution.(Kang et al., 2010; Swain et al., 2007)

### 2.7 LCA of the process

LCA is a methodology to calculate the potential environmental impacts of products, processes, and services. According to ISO 14040 and ISO 14044 (“ISO 14040:2006 - Environmental

178 management — Life cycle assessment — Principles and framework," n.d.; "ISO 14044:2006
179 Environmental management — Life cycle assessment — Requirements and guidelines," n.d.) 4
180 steps shall be implemented to conduct an LCA analysis properly: i) Goal and Scope definition,
181 ii) Life Cycle Inventory (LCI), iii) Life Cycle Impact Assessment (LCIA), and iv) Interpretation.
182 The application of the LCA methodology to the case study under evaluation is described as
183 follows.

184 The goal of the analysis is twofold: firstly, we want to assess whether the use of the recycling
185 process proposed here to recover secondary materials (i.e. aluminum, copper, $Li_2CO_3$, $CoSO_4$,
186 $FeSO_4$, $MnSO_4$, $NiSO_4$) is environmentally convenient compared to the production of the same
187 materials from primary sources. Moreover, the LCA analysis performed in this paper aims at
188 evaluating the main environmental hot-spots of the proposed recycling process. These insights
189 will be useful for the future scale-up from the actual lab-scale level. The function of the process
190 is the treatment of waste LIBs; therefore, the functional unit of the study is defined as 1 kg of
191 waste batteries subject to the hydrometallurgical treatment. Moreover, the potential
192 environmental benefits deriving from the recovery of secondary materials during recycling will
193 be accounted for using the system expansion approach.

194 The process could be divided into six steps: 1) EoL-LIBs safe deactivation, 2) Active materials
195 separation from current collectors, 3) Active materials acid leaching, 4) Transition metals
196 precipitation and recovery of Li, 5) Transition metal sulfides leaching, 6) Separation of transition
197 metals trough organic solvent extraction.

198 A graphical representation of the system boundaries and of all the inputs and outputs of the
199 process is available in Figure 1. Copper and aluminum are recovered from the current collectors
200 by step 2, while lithium is recovered as a carbonate by step 4. However, several sulfides

containing cobalt, iron, manganese, and nickel are also produced as by-products in Step 4. These sulfides can be transformed to valuable sulfates which can be employed to manufacture new LIBs. In order to exploit the possible implementation of a closed-loop recycling in the LIBs sector, the transformation process from sulfides to sulfates (Steps 5 and 6) have been included in the system boundaries. Coherently with the scope of the analysis and the functional unit definition, the system boundaries include all the steps of the recycling process presented here, whereas battery manufacturing and use phases are excluded.

All the steps demand chemicals, water, heat and electricity as inputs expect for the first, which resulted almost neutral. The output flows are the secondary materials, namely aluminum and copper from Step 2, lithium carbonate from Step 4, and cobalt, iron, manganese and nickel sulfates from Step 6. The LCI entails a collection and quantification of all the raw materials and energy consumption flows exchanged between the product system and the environment. More specifically, a foreground LCI measures the direct materials and energy consumption occurring during the recycling process (as represented in Figure 1), that have been directly measured in the laboratory. The background LCI associates indirect resources consumption and emissions to all flows involved in the foreground model; the background data rely on the Ecoinvent 3.6 database ("Ecoinvent 3.6 database," n.d.). An extensive version of the LCI is available in Table S1 in the Supporting Information.

The LCIA step allows to evaluate the potential environmental impact indicators from the LCI analysis; this operation has been done using the LCIA method Environmental Footprint 3.0 (EF 3.0) recommended by the European Commission ("Environmental Footprint 3.0 (EF 3.0)," n.d., p. 3).

In this paper, the LCA results will be expressed first as midpoint indicators that evaluate the effect of the recycling process on the environment based on sixteen impact categories. Then, midpoint results undergo a normalization and weighting process to assess a single score environmental impact that summarizes the contribution of all impact categories.("Environmental Footprint reference packages," n.d.)

## 3. Results and discussions

### 3.1 EoL-LIBs safe deactivation

Spent LiBs safe deactivated through the controlled discharge by covering the cells with conductive materials (brass shreds with 2-6 mm dimensions).(Vizza et al., 2019) This technique ensures a medium resistance contact between the cell terminals and a thermal conductivity to prevent excessive heating. In addition, it can be scaled up and automatized, with different discharge phase mobilization techniques that can be possibly employed to lower the cell duty cycle to a safe level. It is important highlight that the experiments, reported in the Supporting Information, were designed to minimize the heat dispersion to evaluate the generated heat during the batteries' discharging, while a real system should maximize the heat dispersion to avoid cell overheating and prevent unwanted solvent vapor emission or thermal runaway. Then, the experiments were conducted in the worst scenario.

The experimental temperature profiles reported in Figure 2 showed that the physical discharging is an effective method to address the batteries discharging issue (final potential 0.1-0.5V) due by its safe energy dissipation and low temperature increment. Furthermore, the process did not require directly energy and the brass shreds are full reusable without any material consumption resulting in a very low energy and materials impacts. Other common methods require a lot of energy or chemicals: the freezing approach uses liquid nitrogen and it requires

energy to be produced; the heating approach requires the use of fossil fuels and produce $CO_2$, and the chemical discharging use salts dissolved in water and makes slugs as byproduct.(Gao et al., 2024; Jung, 2021; Neumann et al., 2022; Yu et al., 2021) The physical discharging can improve the sustainability of the process thanks to its very low impact on resources, easy implementation, and easy scale up. Furthermore, shreds coming from the mechanical processing of brass for valves and connectors production represents an example of valuable use of waste.

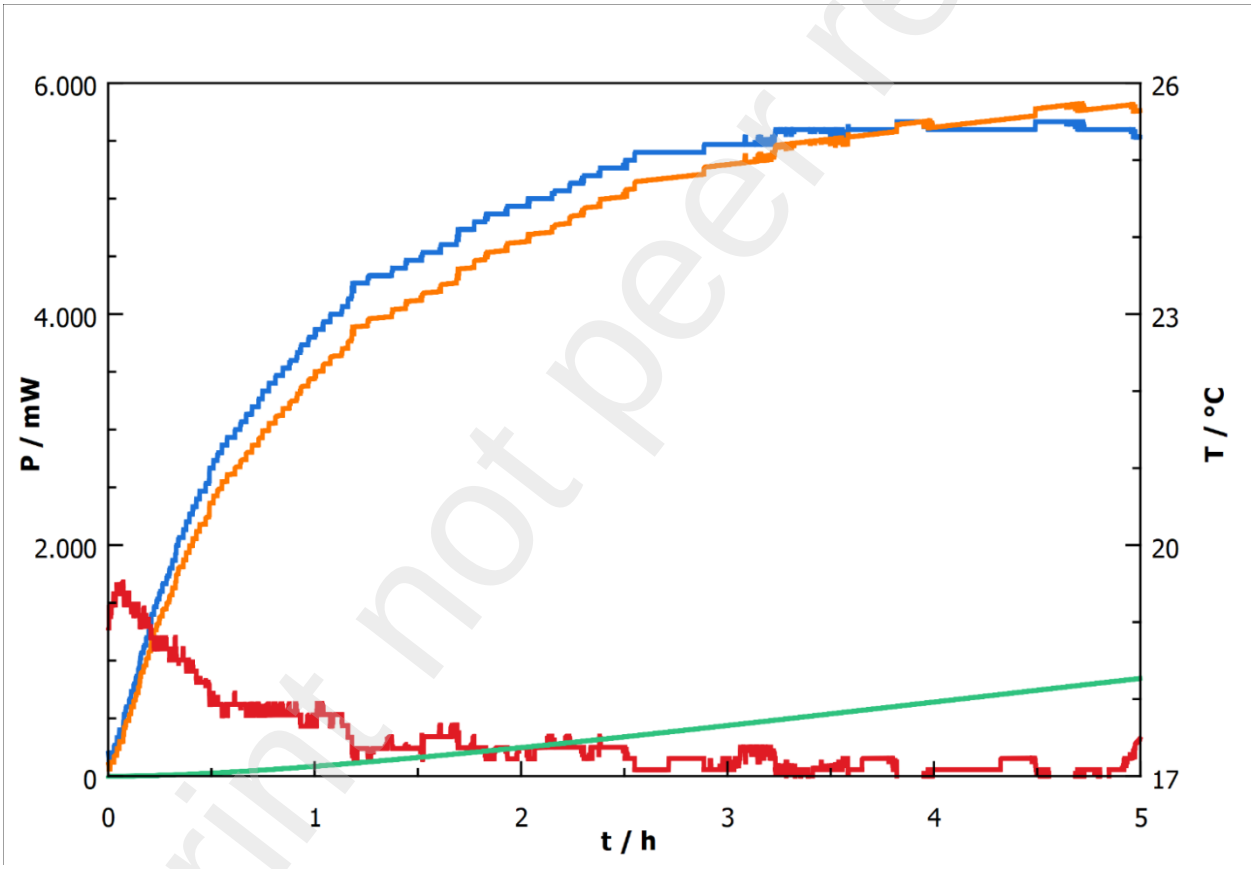



**Figure 2.** Temperature profile during discharge (blue = measured, orange = estimated accounting heat loss) and power (red = generated, green = lost to ambient).

### 3.2 Active materials separation from current collectors

The results of the separation experiments are presented in the radar plots in Figure 3, the farther from the origin the higher the content of the metal in the given phase. For clarity, we named "powder" for the black mass phase, which containing metal oxides (Li, Co, Ni, etc.) and graphite, while we called "foils" the phase made by copper and aluminum current collectors and polymeric separator. As we can see, all experiments can remove more than 85-90% of the elements present in the cathodic materials from the foils. The treatments with 0.5 M $H_2SO_4$ and 3 M HCOOH gave similar removal efficiency, with almost total removal of Co and Li from the foils for the latter. Aluminum contaminates more the powder phase in the case of 3 M HCOOH while copper is almost untouched in both cases. The treatment with 1 M HCOOH preserved 90% of the aluminum foil, demonstrating a lower removal efficiency of 90% for Co and Li (see Table S2).

The superficial partial acid dissolution of the metal substrate causes the collapse of the adhesion properties of the binder and the removal of the active materials, overcoming the drawbacks (time-consuming filtration, low-pressure distillation, and high viscosity increment) of using a specific solvent, such as 1-metil-2-pirrolidone (NMP) and N, N-dimethyl formamide (DMF),(Sarkar et al., 2021; Zhang et al., 2022) to dissolve the binder.(Kaya, 2022) Instead, the acid method allows the almost complete recovery of foils, that can be processed by existing scrap Al and Cu recovery pathways with a production of a dispersion of powders in an acid solution that can be recovered by filtration or used directly in the leaching phase, as will be discussed later.

For comparison, a treatment with ultrapure water was conducted but it did not give any significant metals removal. Then, we tried adding an ultrasonic treatment with a probe inserted in the aqueous phase also to investigate the mechanical treatment. In this case, it was observed a

high efficiency removal but more than 80% of Al and 20% of Cu foils were disrupted and delivered with the powders (see Table S3 for more detailed).

In the end, the treatment with 3M formic acid showed a performance very close to sulfuric acid, representing a sustainability improvement than the state-of-the-art. Furthermore, formic acid could be also used in the leaching step due by its reducing activity toward Co(III) species, as reported in the next section. This represents a tempt to decrease the requires of chemicals: one reagent for two purposes.

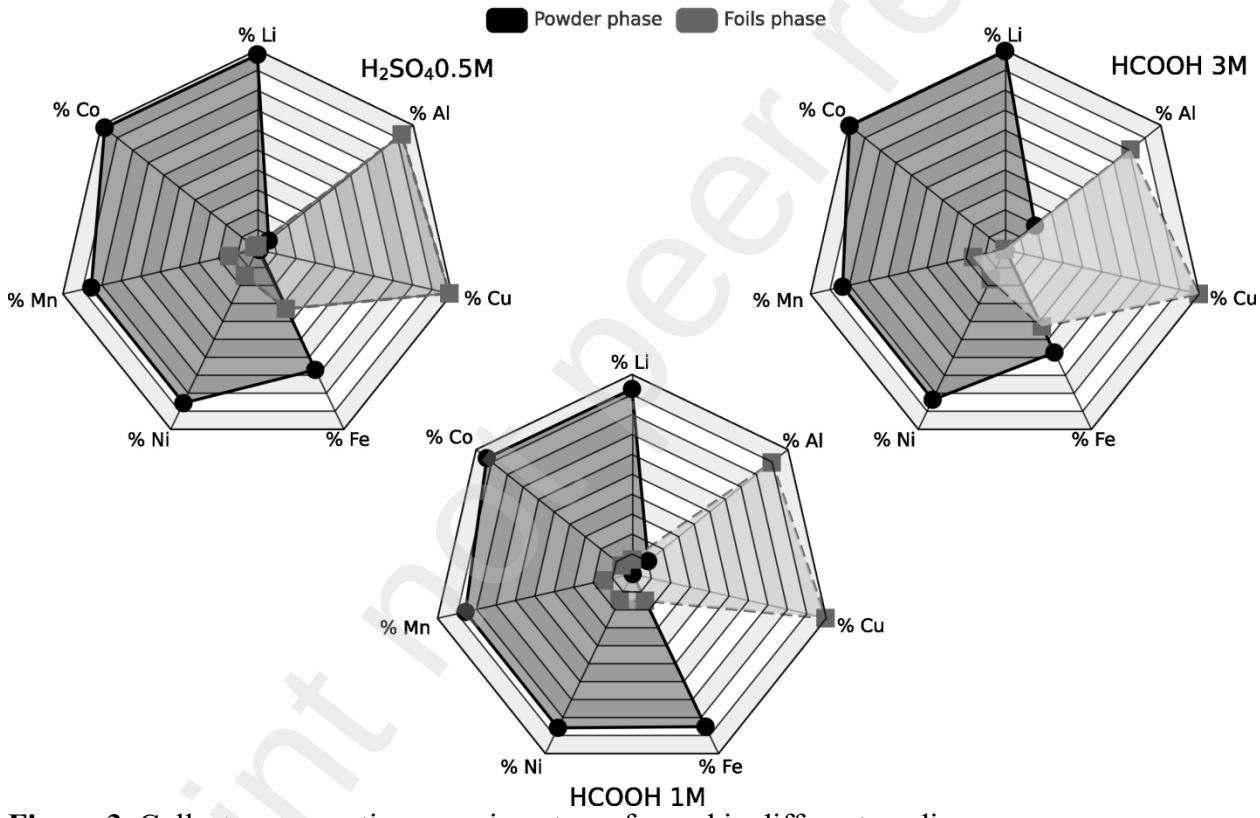



**Figure 3.** Collectors separation experiments performed in different media.

### 3.3 Active materials acidic leaching

Because of the insolubility of Co(III) species, the leaching step often includes a strong acid and a reductive reagent, such as hydrogen peroxide, sugar or other organic compounds, to obtain only Co(II) species (Li et al., 2010). Between common reagents, formic acid was chosen for its

good reducing activity and high availability. Furthermore, it was already used for the separation of black mass from current collectors, resulting in a less reagents consumption.

The leaching experiments were conducted using four different solutions: a) 1M HCCOH, 4M HCl; b) 2M HCCOH, 4M HCl; c) 3M HCOOH, 4M HCl; and d) *aqua regia* for comparison. The results are reported in the radar plots in Figure 4, in which are shown the percentage of aqueous Li, Co and Ni ions and the overall leaching efficiency (reported as "total").

The proposed leaching solutions showed better efficiency increasing the amount of reductive agent from values of 92%, 87%, and 82% for Li, Co, Ni respectively, in case of 1M HCOOH (case a), to 98%, 97%, and 98% in case of 3M HCOOH (case c). Also, the overall metals leaching efficiency increased from 90% to 98%, meaning that only less than 2% of all metal ions are lost to the leftover graphite phase (see Table S4).

The previous choice of 3M HCOOH allows to use of a one-pot process to separate the collector first (with slightly more than 3M HCOOH solution) and then, after the removal of the Al and Cu foils, adding a proper quantity of 37% HCl to the solution reaching 4M for the leaching process. This results in a desirable less chemicals consumption and milder conditions than the state-of-the-art (e.g. *aqua regia*). Furthermore, it is known that hydrochloric acid has a very good leaching activity towards each kind of common batteries' chemistry, such as LCO, NMC, NCA, and LFP, resulting a versatile reagent.(Joulié et al., 2014; Li et al., 2024)

At the end of this step, the proposed process was able to detach the powders (i.e. mixed metal oxides and graphite) from the current collectors, separate the latter, and obtain an aqueous solution with metals ions using an acid leaching. This solution, containing mainly Li, Co, Ni and Mn ions, can either treated to recovery each metal.

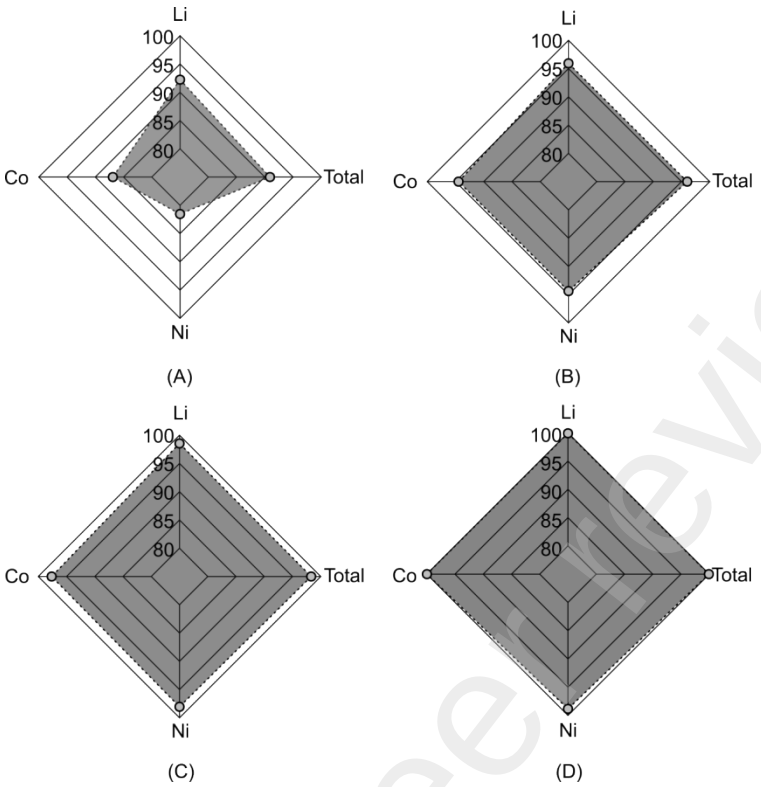



**Figure 4.** Materials leaching process comparison radar plots. Case A) 1M HCCOH, 4M HCl; case B) 2M HCCOH, 4M HCl; case C) 3M HCOOH, 4M Hcl; case D) *aqua regia*.

### 3.4 Transition metal precipitation and recovery of lithium

Lithium ions can be separated by taking advantage of their behavior to produce high soluble salts compared to the other metal ions. Among the many precipitating reagents, we chosen sodium sulfide because the alkaline sulfides are soluble while transition metal sulfides are very insoluble. Furthermore, sulfide salts can be easily changed into sulfate salts, which are common Co and Ni commercial forms, following the same approach of the oxygen-sulfuric acid process used in the mining industry for ore leaching and salt refining.

After the adding of sodium sulfide, lithium and sodium ions remained in the liquid phase, while any other transition metal ion precipitated and filtered easily.

To recover lithium from this solution, we chosen the precipitation with carbonate method because it is a well-known process, it requires alkaline condition as well as the sulfides

precipitation meaning less reagents consumption, and it could be easily reproduced at the lab scale. However, the lithium recovery from brine or hydrothermal springs is a hot topic in the scientific community and many methods are described in the literature.(Ghasem, 2024; Murphy and Haji, 2022; Xiang et al., 2025; Zhang et al., 2021)

The solution was easily separated from the solid, and its volume was reduced to about one third by evaporation leading the precipitation of part of sodium chloride, that can be reused for other purposes. The concentrated solution was then heated again to 90°C and 5 grams of $K_2CO_3$ were added obtaining the precipitation of $Li_2CO_3$. The recovered salt weighted 1.92 g with a yield of 85% respect the total amount of lithium recoverable, and a purity of 94.6% due to the contamination of $Na^+$ and $K^+$ ions. Assuming a full-scale plant, yield and purity can be improved thanks to some industrial devices and tricks, such as recirculating the liquid part again into the precipitation process. This way no lithium would be lost and less potassium carbonate and water would be needed.

### 3.5 Metal sulfides dissolution and separation

The precipitated sulfides contained valuable metals like cobalt, nickel, manganese, iron, and trace of copper and aluminum. The leaching and refining processes of Ni and Co sulfide ores are well established at the large scale because are used in the mine industry. The air-ammonia, oxygen-sulfuric acid and chlorine leaching are the most common processes use for the separation and refining step.(Moats and Davenport, 2014)

To mimic the oxygen-sulphuric acid method at the lab-scale, metal sulfides(ca. 10 g of precipitate) were treated at room temperature with ca. 15 ml of a 25%wt. hydrogen peroxide and sulfuric acid solution resulting in the metal sulfate solution immediately. Then, the metal sulfates solution pH was adjusted to 4.3, as reported in the experimental section, and treated with a 0.3 M

Cyanex272 kerosene solution to separate and recovery cobalt sulfate resulting a yield of 81% and a purity of 97%, which is in line with the literature data,(Strauss et al., 2021; Swain et al., 2008) proving the effectiveness of the method. Furthermore, the other metal ions (such as Ni and Mn) can be recovered by adjusting the pH to appropriate value, as reported elsewhere.(Cheng et al., 2016; Flett, 2005)

To support the sustainability of this choice, we evaluate the atom economy efficiency (AEE) of the reaction (see Tabel S5). Using hydrogen peroxide 30 wt.%, the theoretical AEE is ca. 28%, while we found an actual value of 24% for the reported conditions without any optimization. This result suggest that using a different oxidant reagent, such as $O_2$ instead of $H_2O_2$ 30wt.%, could improve the AEE towards the maximum value of 68%. However, this method seems fine for our purpose to proof the recovering of metal sulfates at the lab-scale. Furthermore, this proves the technical applicability of the proposed process with established treatments in industrial plants and supports its possible use with low capital investments.

### 3.6 LCA of the recycling process

In table 1 are reported all the environmental impact categories calculated by the LCIA method EF3.0 on the proposed process. The rows 1-6 contain environmental impacts of the recycling steps illustrated in Figure 1 as percentage of the overall impact values, which are collected in row 7. On the other hand, rows 8 – 14 contain the environmental credits from all secondary material as percentage of the overall avoided environmental burdens of the process, which are available in row 15. Each column of the table is associated to one of the environmental impact categories proposed by the EF3.0. Red cells are used to express the impact of the recycling steps (the higher the impact of the step, the brighter the shade of red) and green for environmental credits of the materials (the higher the credit from the material, the brighter the shade of green).

Row 16 contains the net environmental impact of the recycling process that have been calculated by balancing the overall impacts and the credits (values in row 7 + values row 15). In row 16, green numbers are used for the results having a negative sign to highlight that the avoided burdens are higher than the impacts, whereas red is used to indicate the categories for which the impacts are higher than the credits.

First of all, it is important point out the environmental neutral impact of the Step 1 (0% for each environmental category) because it does not require appreciable electricity and the materials (metal scraps) can be reused an unlimited number of times. This evidence represents a great advantage compared to other common pretreatment methods, for example heating at 250 °C and freezing with liquid nitrogen require a lot of energy and produce $CO_2$ while discharging in salts solution needs a lot of water and produces slugs.(Gao et al., 2024)

On the other hands, Steps 2, 3, and 5 are responsible for the highest environmental burden for the process.(Alipanah et al., 2021)

Depending on the impact category under evaluation, the percentage contribution of single steps varies significantly. For instance, “Human toxicity, cancer” is mostly affected by Step 5 while “Human toxicity, non-cancer”, “Ozone depletion” and “Resource use, minerals and metals” by Step 3. Step 2 instead is critical for the categories “Climate change” and “Water use”. The environmental burden of Step 2 can be mostly associated to the usage of formic acid, while the impact of Step 3 to hydrochloric acid. The consumption of hydrogen peroxide represents the main environmental impact contributor concerning Step 5. Despite the environmental impact results, the choice of the reagents in the proposed process was not trivial, but it was driven by decreasing the impacts. Indeed, the formic acid was used both for Step 2 and 3, while the hydrogen peroxide was used to mimic the oxygen-sulphuric acid process and showed an atom

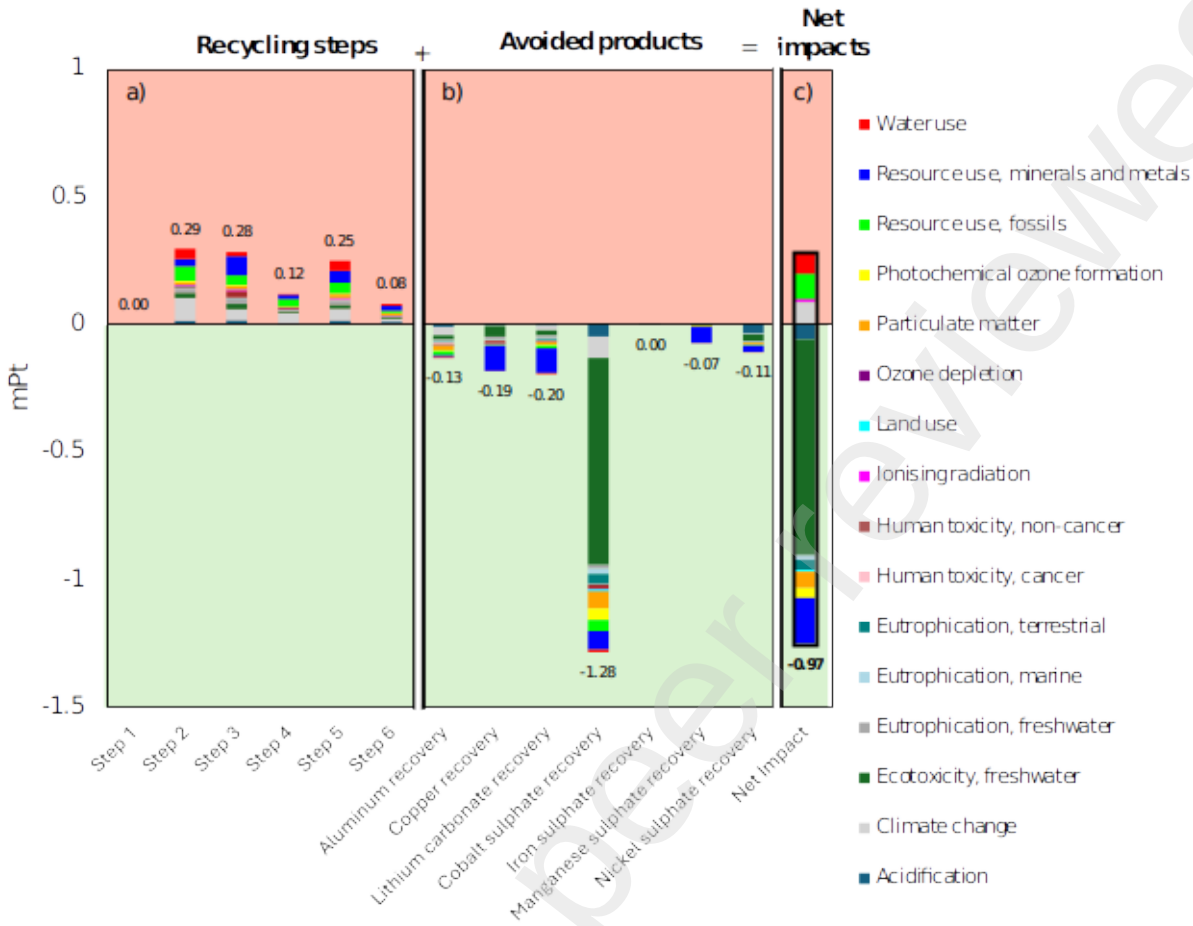
economy ratio similar to the theoretical one, as reported before. On the other hands, these important results suggested the direction to improve the sustainability of the process.(Alipanah et al., 2021) In addition, Steps 4 and 6 slightly contribute to the overall environmental impacts of the process for all categories. Such low impacts are made possible by the reuse of waste carbonates (produced with lithium-carbonates) to replace potassium carbonate for the pH control. Concerning the avoided environmental burdens, we observe that for most of impact categories assessed with EF3.0 the recovery of cobalt sulfate tuns out as the material responsible for avoiding the highest environmental impact. However, for some specific categories, relevant environmental credits are given by secondary aluminum (i.e. for "Human toxicity, cancer") as well as secondary copper and lithium carbonate (i.e. for "Eutrophication, freshwater" and "Resource use, minerals and metals") and nickel sulfate (i.e. for "Acidification"). Observing the results available in row 16, it can be observed that the majority of the impact categories show a negative sign (10 out of 16), which means that the recycling process presented here turns out to be less impacting than the production of raw materials. This evidence entails that the environmental benefits of the proposed recycling process are higher than the impacts. Such result demonstrates that the recycling process described in this paper, which is based on a raw chemical approach and treats unsorted EoL-LIBs, shows a great potential in mitigating most of the environmental impacts associated with the extraction of critical raw materials involved in the batteries industry. Indeed, the proposed process showed the recovery of metals (Li, Co, Ni, Cu, Al, etc) with high yield, good avoided environmental burdens, and methods which can be easily moved from the lab to the pilot and industrial scale.

The discussion on table 1 allows to remark on the balance between potential environmental advantages and burdens of the recycling process presented here as well as on the most critical

environment hot-spots of the process. Also, the discussion highlights that such considerations depend on the impact category under consideration, and therefore it is not possible to get a general outcome from them. Through the normalization and weighting of the impact categories results, it is possible to calculate a single score, which is expressed as millipoints (mPt), that summarizes all the indicators and was illustrated in Figure 5. The chart is composed of three parts, representing a) the impact single scores of the recycling steps, b) the single score of avoided burdens from secondary materials recovery (also called "avoided products"), and c) the balance between the impact of recycling and the credits from secondary materials recovery (also called "net balance"). Overall, it is again demonstrated that the most impacting recycling phase results to be Steps 2, 3, and 5, whose environmental burdens are very similar with each other. Also, Figure 5 confirms the considerations that have been done regarding Steps 1, 4 and 6, showing a moderate single score environmental impact.

**Table 1.** Environmental impact characterization of waste batteries recycling process, including the percentage impacts of recycling steps (red cell), the percentage avoided burdens of secondary materials recovery (in green cells) and the balance between impacts (in red letters, with positive sign) and credits (in green letters, negative sign).

| | | Acidification | Climate change | Ecotoxicity, freshwater | Eutrophication, freshwater | Eutrophication, marine | Eutrophication, terrestrial | Human toxicity, cancer | Hjuman toxicity, non cancer | Ionising radiation | Land use | Ozone depletion | Particulate matter | Photochemical ozone formation | Resource use, fossils | Resource use, minerals and metals | Water use |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Row 1 | Step 1 | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% |
| 2 | Step 2 | 23% | 39% | 29% | 28% | 28% | 30% | 10% | 7% | 32% | 40% | 20% | 22% | 28% | 33% | 15% | 38% |
| 3 | Step 3 | 28% | 19% | 30% | 35% | 28% | 28% | 18% | 71% | 34% | 29% | 44% | 26% | 25% | 22% | 40% | 15% |
| 4 | Step 4 | 8% | 17% | 10% | 8% | 9% | 9% | 6% | 15% | 8% | 7% | 14% | 7% | 11% | 16% | 9% | 3% |
| 5 | Step 5 | 25% | 20% | 23% | 22% | 24% | 23% | 61% | 5% | 22% | 19% | 11% | 27% | 26% | 24% | 25% | 37% |
| 6 | Step 6 | 16% | 5% | 8% | 7% | 11% | 10% | 5% | 2% | 4% | 5% | 11% | 18% | 10% | 5% | 11% | 7% |
| 7 | **Impacts** | **4,77E-2** | **8,84E+0** | **1,48E+2** | **3,10E-3** | **5,96E-3** | **5,98E-2** | **7,15E-9** | **4,30E-7** | **1,31E+0** | **2,84E+1** | **2,04E-6** | **2,84E-7** | **1,95E-2** | **1,35E+2** | **1,55E-4** | **1,42E+1** |
| 8 | Aluminum | 10% | 23% | 2% | 15% | 7% | 6% | 42% | 13% | 27% | 1% | 22% | 17% | 9% | 23% | 2% | 21% |
| 9 | Copper | 5% | 2% | 4% | 24% | 3% | 3% | 22% | 34% | 4% | 1% | 2% | 3% | 3% | 3% | 27% | 3% |
| 10 | $Li_2CO_3$ | 7% | 13% | 2% | 22% | 15% | 8% | 11% | 9% | 10% | 8% | 15% | 10% | 6% | 6% | 26% | 21% |
| 11 | $CoSO_4$ | 42% | 59% | 89% | 30% | 73% | 82% | 18% | 44% | 54% | 89% | 58% | 61% | 76% | 64% | 20% | 51% |
| 12 | $FeSO_4$ | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 0% | 1% | 0% | 0% | 0% | 0% | 0% | 0% | 0% |
| 13 | $MnSO_4$ | 3% | 1% | 0% | 1% | 1% | 0% | 1% | 0% | 2% | 0% | 1% | 3% | 1% | 2% | 17% | 1% |
| 14 | $NiSO_4$ | 33% | 3% | 3% | 7% | 2% | 2% | 6% | 1% | 2% | 1% | 3% | 6% | 6% | 3% | 7% | 2% |
| 15 | **Credits** | **-1,01E-1** | **-5,00E+0** | **-2,07E+3** | **-3,00E-3** | **-1,60E-2** | **-2,26E-1** | **-9,65E-9** | **-5,34E-7** | **-4,67E-1** | **-9,55E+1** | **-3,92E-7** | **-6,51E-7** | **-5,04E-2** | **-5,14E+1** | **-3,09E-4** | **-2,20E+0** |
| 16 | **Net impacts** | **-5,32E-2** | **3,82E+0** | **-1,92E+3** | **9,96E-5** | **-1,01E-2** | **-1,66E-1** | **-2,50E-9** | **-1,05E-7** | **8,41E-1** | **-6,71E+1** | **1,65E-6** | **-3,67E-7** | **-3,10E-2** | **8,38E+1** | **-1,54E-4** | **1,20E+1** |
| 17 | Unit | mol H$^+$eq | kg $CO_2$ eq | CTUe | kg P eq | kg N eq | mol N eq | CTU h | CTU h | kbqU-235 eq | Pt | kg CFC11 eq | Desease inc. | kg NMVOC eq | MJ | kg Sb eq | $m^3$ depriv |




435 **Figure 5.** Normalization and weighting of results: a) impact single scores of each recycling step,
436 b) single scores of avoided burdens, c) net single score impact value of the recycling process.

437 On the other hand, focusing on the avoided environmental burdens, Figure 5 underlines the
438 huge advantage of recovering cobalt sulphate, especially in terms of "Ecotoxicity, freshwater"
439 for which the single score indicator receives a drastic benefit. However, also the recovery of
440 aluminum, copper, and lithium carbonate are highly relevant in contributing to avoid further
441 environmental impacts.

442 The overall positive and negative impacts are +1.02 and -1.98 respectively, resulting in a
443 negative net environmental impact. This means that the overall balance between the
444 environmental categories results favorable and, in the end, the environmental credits provided by

the categories showing results with a negative sign (especially "Ecotoxicity, freshwater") is higher than the impacts associated with positive environmental impacts (especially "Climate change", "Resource use, fossils" and "Water use"). This result further enhances the high environmental sustainability potential shown by the process described in this study over the positive evaluation provided in light of the results shown in table 1.

## 4. Conclusions

A full hydrometallurgical process able to recover more than 85% of lithium (95% purity), copper and aluminum current collectors, and half-processed sulfide material containing all other transition metals (Co, Ni, Mn, Fe) was described.

The process was based on a raw chemical approach inspired by circular economy principles to decrease its overall environmental impact. Its main strength is high flexibility, because it can treat any kind (LCO, NMC, NCA, LFP, etc) of mixed EoL-LIB provided by a real disposal plant, i.e. without any sorting step, thanks to the choice of versatile reagents. Then, the process is able to recover copper and aluminum current collectors and graphite as are, lithium as lithium carbonate, and all other transition metal ions as sulfides, which can be easily converted into sulphates as common commercial form.

The choice to base the process on a raw chemistry helps the scale up from the lab to industrial scale resulting in lower investments.

Furthermore, the life cycle assessment results in a negative overall impact showing the high environmental sustainability potential of the proposed raw chemical approach.

## ASSOCIATED CONTENT

**Supporting Information**.

The following files are available free of charge.

Supporting_Information.pdf

## AUTHOR INFORMATION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The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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