EUROPEAN COMMISSION

HORIZON 2020 PROGRAMME - TOPIC H2020-LC-BAT-2019
Strongly improved, highly performant and safe all solid-state batteries for electric vehicles.

GRANT AGREEMENT No. 875189

SAFELiMOVE – Deliverable Report

<< D8.4 – Recycling study on SAFELiMOVE technologies>>

Deliverable No.	SAFELiMOVE D8.4	
Related WP	WP8	
Deliverable Title	Recycling study on SAFELiMOVE technologies	
Deliverable Date	2023-04-30	
Deliverable Type	REPORT	
Dissemination level	Public (PU)	
Written By	Davide Burlon (LCE) Massimo De Pieri (LCE)	2023-04-17
Checked by	Alexander Tesfaye (UMC) Abdelbast Guerfi (HQ)	2023-05-12 2023-06-01
Approved by	Maria Martinez (CICe)	2023-05-16
Status	Final	2023-06-16

Disclaimer/ Acknowledgment

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 875189. The information and views set out in this publication does not necessarily reflect the official opinion of the European Commission. Neither the European Union institutions and bodies nor any person acting on their behalf, may be held responsible for the use which may be made of the information contained therein.

Publishable summary

This document is the first output of research activities carried out by Life Cycle Engineering (LCE) in the SAFELiMOVE H2020 project. SAFELiMOVE consortium is engaged in designing, testing and delivering at prototype scale an innovative battery for e-mobility purposes, exploring the Solid State technology; within this framework, LCE is appointed to evaluate environmental performances and general sustainability of this innovative technology to understand what are benefits and loads compared to existing benchmark technologies delivering the same function. LCE research activities follow in the Work Package 8 - Industrialization perspective and Roadmap towards 2030: specific activities related to this deliverable are included in task 8.5 – Recycling.

Goal of task 8.5 is to focus on battery recycling technologies, providing comprehensive technology review and suggesting what the best technology for SAFELiMOVE battery might be in the early future. This deliverable is organized in 3 main modules.

Module 1 represents a general introduction to battery recycling research area: general considerations on battery architecture and components are reported, then a focus on battery chemistries is provided. Core section of module 1 is related to the European battery directive, considering limits, targets and thresholds introduced by this regulatory tool and strongly related to battery recycling.

In Module 2, details about each identified battery recycling technologies are reported. Per each technology, a deep description of the process is provided; details about critical aspects and promising features are reported as well to introduce the concept of "technological comparison" among each route.
First, mechanical recycling is investigated; this technique mainly implies only a mechanical separation of the battery, thus introducing the need for additional treatments afterwards. This process feature represents a significant shortcoming for the mechanical recycling, as either the quality of the output material is not ready to enter the market again and additional processes are needed to complete the recycling.
Pyrometallurgy is the second evaluated technology. It is well suited for traditional battery chemistries and a valuable quality of the output is achieved. On the other hand, high amount of energy is required (with an associated risk of high environmental footprint if not coupled with renewable energy sources) and innovative chemistries such as SAFELiMOVE one might generate issues in the process due to chemical nature of some components. In addition, only valuable metals are recovered with all other materials being lost or ending up in the slag (e.g. lithium) with little opportunity to be reclaimed.
Third technology is represented by Hydrometallurgy. Despite registering a high material demand, especially acids, solvents and water, this technology is well suited to handle a wide variety of battery chemistries in an almost stand-alone configuration (i.e., without the need for further treatments).
When no actual recycling is applied, but rather refurbishment techniques, the concept of "Direct recycling" is introduced. This is the fourth technology studied in this research. Direct recycling allows to recover a wide range of battery components with limited resource consumption, however relying heavily on manual operations due to its nature and the low technology readiness level. This recycling procedure is still in a development stage and only studied as a research topic nowadays.

Specific recycling information about the SAFELiMOVE battery components is introduced in the last section of Module 2, explaining what the main issues related to Lithium metal and hybrid electrolyte are.

Outcomes of the research are thoroughly reported in Module 3, where two main sections are introduced. First, the best battery recycling technology is suggested based on a multidimensional decision-making tool which considers several metrics which are relevant for the investigated topic. Namely, the following items are considered:

- Independency of the process
- Material recovery efficiency
- Process energy intensity
- Process material intensity
- Process industrial maturity

- Battery chemistry range
- Recovered materials quality.

The result of this rating is reported in a weighted average form and suggests Hydrometallurgy as the most promising recycling route for general batteries for e-mobility.
Then, specific considerations on SAFELiMOVE battery recycling are reported. As the project is reaching the prototype phase, detailed information will be available in further months; nevertheless, preliminary key findings related to recycling potential of the project battery are reported and explained. Finally, conclusions and recommendations are listed as final sections of this research activity.

Executive summary

This report constitutes deliverable 8.4 “Recycling study on SAFELiMOVE technologies” of SAFELiMOVE project. The activities presented here, performed during the project period, refer to WP8 “Industrialization perspective and Roadmap towards 2030” and more specifically to task 8.5 “Recycling”.

No deviations from planned timings or planned objectives occurred for task 8.5. The task was successfully completed, and all goals achieved.

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1 List of Abbreviations

EC	European Commission
EV	Electric Vehicle
GHG	Greenhouse gases
LiB	Lithium-ion battery
SSB	Solid-state battery
LiM	Lithium metal
LFP	Lithium Iron Phosphate battery
LCO	Lithium Cobalt Oxide battery
LMO	Lithium Manganese Oxide battery
NMC	Lithium Nickel Manganese Cobalt Oxide battery
NCA	Lithium Nickel Cobalt Aluminium Oxide battery
LLZO	Lithium Lanthanum Zirconium Oxide electrolyte
LATP	Lithium aluminium Titanium Phosphate electrolyte
PEO	Polyethylene Oxide
PP	Polypropylene
PA	Polyamide
BMS	Battery Management System

2 Introduction

This report constitutes deliverable 8.4 “Recycling study on SAFELiMOVE technologies” of SAFELiMOVE project. The activities presented here, performed during the project period, refer to WP8 “Industrialization perspective and Roadmap towards 2030” and more specifically to task 8.5 “Recycling”.

Main purposes of the task are:

- a literature analysis on existing recycling process for the known materials already included in current batteries relevant for SAFELiMOVE technologies, focusing on the commercial Li-ion batteries, to draw a technology review identifying most promising recycling routes to be explored in the project.
- a literature analysis focused on non-conventional materials focusing on SAFELiMOVE battery type, thus investigating challenges for solid electrolyte and lithium-metal anode recycling.
- definition of most promising recycling processes respecting the European Directive (2006/66-EC) on batteries recycling, showing that recycling threshold (>50% wt.) of material recovery and valorization can be reached for the technology developed in SAFELiMOVE project.

The report follows the same structure as the above-mentioned task purposes. In addition, as an introduction to the more technical chapters, a focus on the new battery proposal from the European Commission (to replace Battery Directive 2006/66-EC) is provided, to give an overview on the future policy framework for battery recycling indicated by Europe.

SAFELiMOVE Deliverable 8.4 on battery recycling addresses key challenges in the framework of electric mobility sector. A rapid market growth for electric vehicles (EVs from now on) is happening to meet European and global targets of transport sector GHG-emissions reduction. A huge number of waste EVs, thus of EV batteries, to be treated is then foreseen in the next years, posing a serious challenge for the whole waste management sector. According to (1) indeed, around 8 million tons of waste from lithium-ion batteries (LiBs) are expected to be generated in 2040. However, this aspect offers several opportunities to the whole electric mobility chain as well. Spent EV batteries are to be thought as a valuable source of materials for new battery production, as well as a potential still-exploitable source of electrical energy.

According to the waste management hierarchy concept (2), well applicable for EV batteries, reuse should be preferred to recycling. The latter can be a resource- and energy-intensive process whose environmental impacts are not negligible. Reuse, instead, allows extracting maximum economic value and minimizing short- to medium-term environmental burdens thanks to further applications (e.g. for stationary applications) other than the one the battery was formerly manufactured for.

However, even batteries properly reused will reach the state where no further value can be extracted from them. At this end-of-life stage, recycling must be the inevitable fate for all batteries (2). Adopting processes as efficiently as possible, recycling is able to turn waste batteries into valuable sources of materials. This would both translate into a reduction of critical life-cycle stages environmental impacts (mining and manufacturing) of the former battery and into tackling elements and raw materials depletion issue. While there are still controversies on whether lithium-ion battery raw materials supply will be critical in next years for resources scarcity (3), it is well known that materials needed for battery production come from very few countries in the world. As representative figures:

- Australia, Chile, and Argentina own around 90% of global lithium resources (4).
- More than 70% of global cobalt production comes from Congo (1).
- In 2017, only 32 countries worldwide accounted for the production of raw materials needed for lithium-ion batteries manufacturing (4).

A stable and sustainable supply chain is then of paramount importance, especially for EU countries. The development of reliable and efficient recycling processes, and thus of a strong industry based on battery recycling, should be a key driver for Europe to reduce its dependency on foreign countries regarding raw materials supply for the mobility sector. Due to the rapidly increasing number of EVs that will be purchased in Europe, in the next year several waste batteries will be available to be exploited as source of valuable materials in EU regions.

3 European Directive on battery recycling and new proposal

Since 2006, batteries and waste batteries have been regulated at EU level under the Batteries Directive 2006/66/EC. The Commission proposed to revise this Directive in December 2020 due to new socioeconomic conditions, technological developments, markets, and battery uses. By means of this proposal, The Commission indicated mandatory requirements for all batteries (i.e., industrial, automotive, electric vehicle and portable) placed on the EU market. Requirements such as use of responsibly sourced materials with restricted use of hazardous substances, minimum content of recycled materials, carbon footprint, performance and durability and labelling, as well as meeting collection and recycling targets, are essential for the development of more sustainable and competitive battery industry across Europe and around the world (5).

Despite being introduced after SAFELiMOVE project start (which occurred in February 2020), a focus on this proposal is of paramount importance to understand EU future requirements in the battery field.

The current European Directive most relevant limitation is that it does not mention specifically neither lithium (ion) rechargeable batteries nor their use for electric mobility. Such batteries are handled by the Battery Directive as “other batteries”, different from lead-acid and nickel-cadmium batteries. Regarding recycling at end of life, for the “other batteries” category, only a generic “50 % of average weight” is indicated as minimum recycling process efficiency. This threshold is the one targeted by SAFELiMOVE project, as declared in the project proposal.

In addition, the end-of-life stage of batteries is addressed by the current regulatory framework through the Batteries Directive. There are currently no legal provisions in the EU that cover other aspects of the production and use phases of batteries, such as electrochemical performance and durability, GHG emissions, or responsible sourcing. In line with the ‘one-in-one-out’ principle 10, the proposed Regulation should replace the current Batteries Directive.

3.1 General overview and main contents

For the reasons explained above, in April 2019, the Commission published an evaluation of the Batteries Directive, carried out by stakeholders’ consultation and subsequent setting of targets by impact assessment evaluation (6).

The main needs expressed by **representatives from industry** were for:

- a stable regulatory framework that ensures investment certainty
- a level playing field that enables the sustainable production of batteries
- the efficient functioning of recycling markets to increase the availability of quality secondary raw materials.

The main concerns expressed by **representatives from civil society** were on the need for sustainable sourcing and for applying the principles of the circular economy to the battery value chain.

Thanks to the impact assessment evaluation, 13 measures were identified to address the above-mentioned problems. Within each of the 13 broad policy measures, several sub-measures were considered. These sub-measures are in many cases alternative to each other (e.g. for Measure 3, collection-rate targets for portable batteries can be either 65% or 75%, but not both), whereas in other cases they are designed so that they can be cumulative and/or complementary (e.g. for Measure 13, a battery 'passport' for industrial batteries works in addition to information obligations) (6).

To facilitate the analysis, the proposal groups the sub-measures into four main policy options, which are compared against a business-as-usual scenario. These four options are set out below.

- Option 1, business-as-usual, is an option that keeps the Batteries Directive, which mostly covers the end-of-life stage of batteries, unchanged. For the earlier stages in the value chain, there is currently no EU legislation in place and so this will remain unchanged.
- Option 2, the medium level of ambition option, is an option which builds on the Batteries Directive, but gradually strengthens and increases the level of ambition. For the earlier stages in the value chain for which there is currently no EU legislation, the proposed change is to bring in information and basic requirements as a condition for batteries to be placed on the EU market.
- Option 3, the high level of ambition option, is an approach that is a bit more disruptive, but still within the limits of what is technically feasible. It entails, for example, setting limit values and thresholds to be complied with within a set deadline.
- Option 4, the very high level of ambition option, includes measures that would go significantly beyond the current regulatory framework and current business practices.

Table 1 presents an overview of the different sub-measures included in the policy options, with the preferred option based on the impact assessment highlighted in green.

As indicated in the proposal, the Commission's preferred option is a combination of Option 2 and Option 3. The combination chosen provides a balanced approach in terms of effectiveness (achievement of the objectives) and efficiency (cost-effectiveness).

Table 1 - Options for the identified measures (*green = preferred option; light green = preferred option pending a revision clause)* (6)

Measures	Option 2 - medium level of ambition	Option 3 - high level of ambition	Option 4 – very high level of ambition
1. Classification and definition	New category for EV batteries Weight limit of 5 kg to differentiate portable from industrial batteries	New calculation methodology for collection rates of portable batteries based on batteries available for collection	/
2. Second-life of industrial batteries	At the end of the first life, used batteries are considered waste (except for reuse). Repurposing is considered a waste treatment operation. Repurposed (second life) batteries are considered as new products which have to comply with the product requirements when they are placed on the market	At the end of the first life, used batteries are not waste. Repurposed (second life) batteries are considered as new products which have to comply with the product requirements when they are placed on the market.	*Mandatory second life readiness*
3. Collection rate for portable batteries	65% collection target in 2025	70% collection target in 2030	75% collection target in 2025

Measures	Option 2 - medium level of ambition	Option 3 - high level of ambition	Option 4 – very high level of ambition
4. Collection rate for automotive and industrial batteries	New reporting system for automotive, EV and industrial batteries	Collection target for batteries powering light transport vehicles.	Explicit collection target for industrial, EV and automotive batteries
5. Recycling efficiencies and recovery of materials	Lithium-ion batteries and Co, Ni, Li, Cu: Recycling efficiency lithium-ion batteries: 65% by 2025 Material recovery rates for Co, Ni, Li, Cu: resp. 90%, 90%, 35% and 90% in 2025 Lead-acid batteries and lead: Recycling efficiency lead-acid batteries: 75% by 2025 Material recovery for lead: 90% in 2025	Lithium-ion batteries and Co, Ni, Li, Cu: Recycling efficiency lithium-ion batteries: 70% by 2030 Material recovery rates for Co, Ni, Li, Cu: resp. 95%, 95%, 70% and 95% in 2030 Lead-acid batteries and lead: Recycling efficiency lead-acid batteries: 80% by 2030 Material recovery for lead: 95% by 2030	/
6. Carbon footprint for industrial and EV batteries	Mandatory carbon footprint declaration	Carbon footprint performance classes and maximum carbon thresholds for batteries as a condition for placement on the market	/
7. Performance and durability of rechargeable industrial and EV batteries	Information requirements on performance and durability	Minimum performance and durability requirements for industrial batteries as a condition for placement on the market	/
8. Non-rechargeable portable batteries	Technical parameters for performance and durability of portable primary batteries	Phase out of portable primary batteries of general use	Total phase out of primary batteries
9. Recycled content in industrial, EV and automotive batteries	Mandatory declaration of levels of recycled content, in 2025	Mandatory levels of recycled content, in 2030 and 2035	/
10. Extended producer responsibility	Clear specifications for extended producer responsibility obligations for industrial batteries Minimum standards for PROs	/	/
11. Design requirements for portable batteries	Strengthened obligation on removability	New obligation on replaceability	*Requirement on interoperability*
12. Provision of information	Provision of basic information (as labels, technical documentation or online) Provision of more specific information to end-users and economic operators (with selective access)	Setting up an electronic information exchange system for batteries and a passport scheme (for industrial and electric vehicle batteries only)	/

Measures	Option 2 - medium level of ambition	Option 3 - high level of ambition	Option 4 – very high level of ambition
13. Supply-chain due diligence for raw materials in industrial and EV batteries	Voluntary supply-chain due diligence	Mandatory supply chain due diligence	/

For this deliverable purposes, measures 2, 4, 5 and 9 are of main interest and will be presented more in detail below, directly quoting the proposal (6):

- **Measure 2** on second-life for industrial and EV batteries recognizes that there are trade-offs between promoting the development of second-life batteries on the one hand, and recycling on the other. The Commission concluded that a combination of Option 2 and Option 3, whereby specific end of waste criteria including a state of health check are set that batteries have to fulfil in order to be sent to repurposing or remanufacturing, will provide the most appropriate way forward. This approach is aimed to encourage the repurposing and remanufacturing of batteries while ensuring that waste batteries undergo proper treatment in line with EU waste legislation and international agreements.
- **Measure 4** on collection rate for industrial and EV batteries does not set collection targets as the ones for portable batteries. This is because of the "new" product flow in the market and need to first develop the "available for collection" methodology for it. Because of this, option 2 on creating a new reporting system for automotive and industrial batteries is the preferred path. Option 3 is proposed to be re-assessed through a review clause.
- **Measure 5** on recycling efficiencies and material recovery sets targets for 2025 based on what is currently technically feasible (option 2) and targets for 2030 based on what will be technically feasible in the future (option 3). Table 2 below summarises the targets set in the two different time frames, for the sole lithium-ion batteries, regarding recycling efficiencies and materials recovery:

Table 2 - Recycling processes and materials recovery efficiencies (6)

Li-ion batteries	2025	2030
Recycling efficiency	65%	70%
Cobalt recovery	90%	95%
Nickel recovery	90%	95%
Lithium recovery	35%	70%
Copper recovery	90%	95%

Measure 9 on recycled content in industrial and EV batteries sets two complementary options, namely bringing in a mandatory declaration of recycled content in the short term (option 2) and setting mandatory targets for recycled content for lithium, cobalt, nickel and lead in 2030 and 2035 (option 3). More in detail, as cited in article 8 of the proposal, industrial batteries, electric vehicle batteries and automotive batteries with internal storage and a capacity above 2 kWh that contain cobalt, lead, lithium or nickel in active materials shall be accompanied by technical documentation demonstrating that those batteries contain a minimum share of cobalt, lead, lithium or nickel recovered from waste present in active materials in each battery model and batch per manufacturing plant. In

- Table 3 below, the minimum limits are indicated, for the two different time frames:

Table 3 - Industrial/EV batteries minimum recycled content per material (6)

	From 1st January 2030	From 1st January 2035
Cobalt	12%	20%
Lead	85%	85%
Lithium	4%	10%
Nickel	4%	12%

Such measures should contribute to providing a predictable legal framework that would encourage market players to invest in recycling technologies.

3.2 Chapter VII – End-of-life management of batteries

Chapter VII of the proposal is entirely devoted to the end-of-life management of batteries, both on technical aspects and on responsibilities of parties. A summary of the main topics addressed is provided in this document.

3.2.1 Parties' obligations

Articles 49, 50, 51 and 52 set obligations for the different actors involved in all the value chain, regarding collection of waste EV batteries:

- **Battery producers** shall take back, free of charge and without an obligation on the end user to buy a new battery, nor to have bought the battery from them, all waste electric vehicle batteries of the respective type that they have made available on the market for the first time in the territory of that Member State. For that purpose, they shall accept to take back waste automotive batteries, industrial batteries and electric vehicle batteries from end-users, or from collection points provided in cooperation with licensed distributors of automotive/EV batteries, EoL vehicle treatment/recycling facilities, public authorities/third parties carrying out waste management on their behalf.
- **End users** shall discard waste batteries separately from other waste streams, including from mixed municipal waste. Waste batteries incorporated in vehicles or appliances and that are not readily removable by the end-user, shall be discarded by the end user in accordance with the Directives 2000/53/EC and 2012/19/EU, where applicable.
- **Operators of waste treatment facilities** shall hand over waste batteries resulting from the treatment of end-of-life vehicles to the producers of the relevant batteries or producer responsibility organizations acting on their behalf.

3.2.2 Treatment and recycling targets

Article 56 of the proposal clearly states that collected batteries shall be neither landfilled nor incinerated. However, unlike portable batteries, for EV batteries no collection rate threshold is indicated in the proposal currently (as already mentioned in Measure 4 in Table 1). Article 57 indicates that all waste batteries collected shall enter a recycling process, whose recycling efficiencies and levels of recovered materials shall comply with the limits indicated in Table 2. It is also indicated that recycling may be undertaken outside the Member State concerned or outside the European Union, provided that the shipment of waste batteries follows the proper European Commission Regulations.

3.2.3 End-of-life information

Producers or producer responsibility organisations acting on their behalf shall make available to end users and distributors all the information regarding the prevention and management of waste batteries with respect to the types of batteries that the producers supply within the territory of a Member State. In addition, regarding requirements to repurposing and remanufacturing, Independent operators shall be given access to the battery management system of electric vehicle batteries with internal storage with a capacity above 2 kWh, on equal terms and conditions, for the purpose of assessing and determining the state of health and remaining lifetime of batteries.

From the moment that a battery model is supplied within the territory of a Member State producers shall make available electronically, upon request, to waste management operators carrying out repair, remanufacturing, preparing for re-use, treatment and recycling activities, as far as it is needed by those operators to carry out those activities, the information on the processes to ensure the dismantling of vehicles and appliances in a way that allows the removal of incorporated batteries.

4 Battery Recycling Technologies

In this chapter, the technology review on EV batteries recycling processes is provided, result of the scientific literature analysis performed.

The first part focuses on state-of-the-art processes, thus on processes already developed and/or relevant in an industrial perspective. It must be noted that such processes are developed to handle mainly lithium-ion EV batteries. Although being different from SAFELiMOVE technology, lithium-ion battery is currently the most common technology for electric mobility. In addition, several components of Li-ion battery can be found in the solid-state SAFELiMOVE battery as well. Hence, an analysis on current recycling processes can provide fruitful knowledge for the development of a proper recycling pathway for solid state batteries.

The second part focuses on non-conventional key components present in SAFELiMOVE technology, namely the solid electrolyte and the lithium-metal anode, offer a technology review on existing recycling processes for such components.

4.1 State of the art

The recycling techniques for LIBs are still under development, and there is currently no technology available (each technology has certain advantages and disadvantages) that would permit the recovery of all elements from used batteries. Furthermore, there are relevant losses in the current technological innovation, and, at the same time, battery chemistry is always evolving. Therefore, recycling requires continuous advancement according to the material use, battery system design, and manufacturing process (7).

So far, four main typologies of LiB recycling processes were studied:

- Mechanical recycling
- Pyrometallurgical recycling
- Hydrometallurgical recycling
- Direct recycling

Each technology has its own characteristic and is suitable to recover different battery material/components. By different combinations of the above-mentioned processes, recycling processes were also developed at industrially-relevant scales. Several recycling industries were established in Europe, a list of the most important will be provided further in the document.

4.1.1 Mechanical recycling

Mechanical processing involves the use of different techniques to liberate, classify, and concentrate materials without altering their chemistry (8). Companies performing such technology separate and recover battery components (as shown in Figure 1) to be further distributed to other recyclers and/or metal producers. In Europe, for instance, mechanical recycling is operated by Akkuser (Finland) and Batrec

(Switzerland) companies. They both perform mechanical separation of black mass (mixture of cathode and anode active materials), current collectors (Al and Cu foils) and other battery components (printed circuit boards, casing, cables, etc.) that are further sold to specialized recyclers.

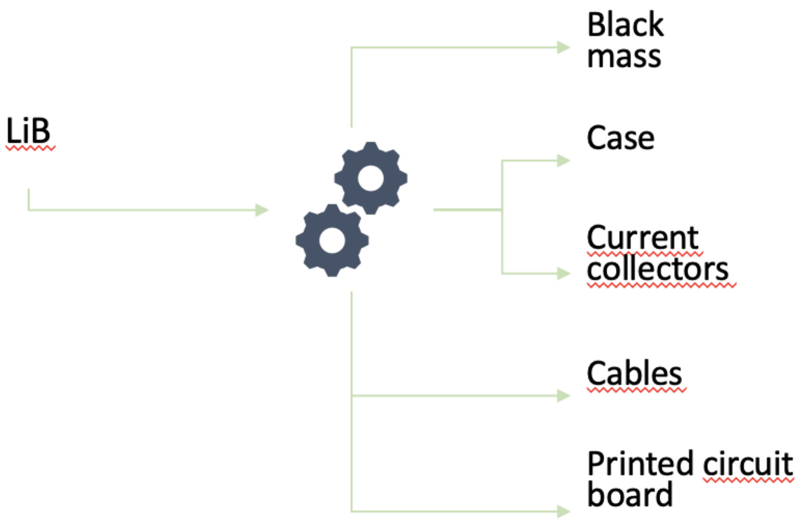

Figure 1 - Mechanical recycling simplified process flow diagram

While it is of paramount importance to thoroughly separate battery components, to allow further recycling processes as efficient as possible, this type of recycling is not sufficient to actually recover materials and components from the former battery, ready to be reused for new batteries manufacturing. When mechanical recycling processes are coupled with subsequent recycling processes (pyrometallurgy, hydrometallurgy, direct metallurgy), they are called **mechanical pre-treatments**. Their importance will be analysed more in depth in chapter 4.1.5.

Table 4 - Advantages and disadvantages of mechanical recycling processes

Pros	Cons
Separation of components and materials	Further treatment steps needed to obtain materials to be used for new battery production
High share of battery components recovered	

4.1.2 Pyrometallurgy

Pyrometallurgical recycling process uses a high-temperature furnace to reduce the component metal oxides to an alloy of Co, Cu, Fe and Ni, by the so-called ***reductive smelting*** process. The process is already established commercially for consumer LiBs and, thanks to its flexibility in battery feedstock, allows the recycling of batteries based on different chemistries (2). Umicore recycling plant (located in Belgium, able to treat around 7000 t of batteries per year) is based on pyrometallurgical process and is suitable for both lithium-ion and nickel metal hydride batteries (1). Another advantage of pyrometallurgy is that it requires little to no pretreatment methods (most often shredding or crushing) to prepare batteries for recycling (1). In addition, as the metal current collectors – especially aluminium – favours the smelting process, pyrometallurgy can accept directly whole cells or modules, without the need for a prior passivation step (9).

Outcomes of the pyrometallurgical process are the metallic alloy fraction, slag and gases. The gaseous products include volatile organics from the electrolyte and binder components and flue gases from polymers combustion. Electrolytes and plastics combustion provide energy to the process, thus reducing the external energy demand required for the process. This means that the recovery of electrolytes and plastics (approximately 40–50 per cent of the battery weight) is not considered in pyrometallurgical recycling. To obtain the separated metals, the metal alloy has to be separated through hydrometallurgical processes (see section 4.1.3). The slag typically contains aluminium, manganese and lithium, which can be later obtained by further hydrometallurgical processing, but can be directly sold to other industries as well (e.g. cement industry). Lithium recovery from smelting slag is indeed not an efficient process as for the valuable metals in the alloy, requiring a large amount of energy (9).

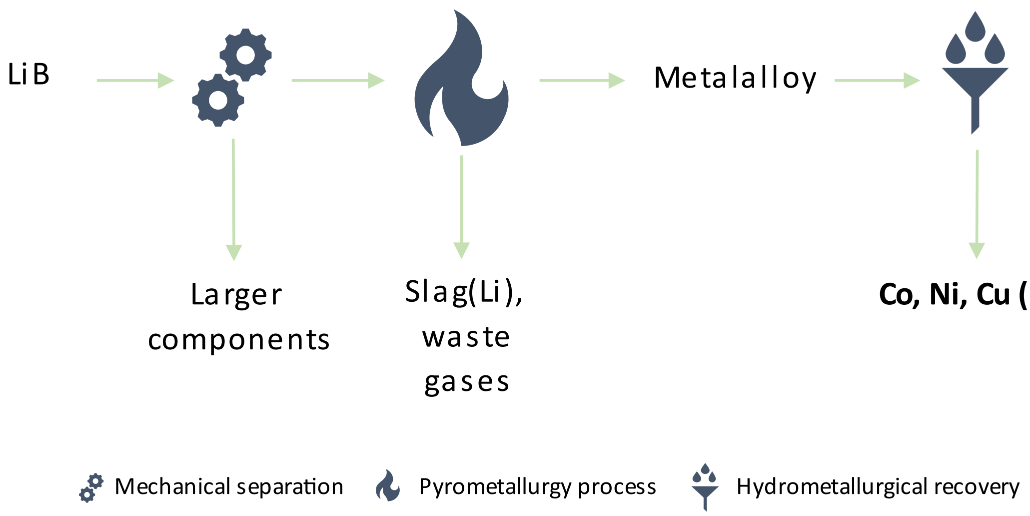

Figure 2 - Pyrometallurgical recycling simplified process flow diagram

Battery recycling nowadays is driven mainly by recovering valuable metals, such as cobalt and nickel. This is why pyrometallurgy is the most frequently used recycling process despite its high energy demand, limited numbers of materials recovered and environmental burdens (such as production of toxic gases). In addition, for the effective metals recovery further leaching processes are needed, meaning significant chemical reagents and water consumption.

In addition, even though technically suitable for all types of batteries, pyrometallurgy would not be economically sustainable for those cathode chemistries with low (or absent) valuable metals content, such as LFP batteries. Due to their favourable characteristics of safety, stability and affordability, LFP-based batteries are foreseen to cover a relevant share in EV batteries market. Hence, alternatives to pyrometallurgy must be studied and implemented to ensure proper levels of materials/components recovery, where at least an effective Lithium recovery is ensured.

Table 5 - Advantages and disadvantages of pyrometallurgical recycling processes

Pros	Cons
High flexibility to different battery chemistries	Except for metals, other battery materials poorly recovered or completely discarded
Little to no pre-treatments required	Unable to accept “cheaper” cathode formulations (e.g. LFP)
High share of valuable metals (Co, Ni, Cu) recovery	High energy demand and environmental burdens linked to toxic gas emissions
Mature technology developed at industrial scale	

4.1.3 Hydrometallurgy

Various hydrometallurgy techniques were developed in recent times for recycling cathode active materials of different chemistries LiBs, such as LCO, LMO, NMC, NCA and LFP, to recover not only valuable metals such as Co, Ni and Mn, but also Li (10). Hydrometallurgical recycling comprehends three main steps: methods use primarily aqueous solutions to extract such metals from LiBs. Extraction (also referred to as *leaching*) is often carried out aqueous solutions of H_2SO_4 and H_2O_2, although HCl, HNO_3, and organic acids including citric and oxalic acids are commonly used (1).

- The first extraction stage, the *leaching*, consists in the dissolution of valuable metals by acid or basic agent in an oxidizing or reducing medium in leaching tanks [55,57]. This is often performed by means of aqueous solutions containing H_2SO_4 and H_2O_2, although HCl, HNO_3, and organic acids including citric and oxalic acids are commonly used (1).
- The second stage of *impurity removal* by solid-liquid separation, which clarifies the leached solution by filtration or centrifugation.
- The last stage is then devoted to the final recovery of valuable metals in hydroxide or metal salts. This process includes, for example, solvent extraction, electrochemical techniques, selective precipitation, and separation by ion exchange resins (10). Cobalt is usually extracted either as the sulfate, oxalate, hydroxide or carbonate, and then lithium can be extracted through a precipitation reaction forming Li2CO3 or Li3PO4 (2).

Thanks to the overall high purity of metals recovered, they can be re-used not only for remanufacturing the original cathode materials, but also in a wide range of other applications (2).

It is important to notice that battery pre-treatments are mandatory for an effective hydrometallurgy recycling, to separate the active components from all other battery parts (e.g. modules packaging and cells current collectors) and obtaining the so-called *black mass* used as feed for hydrometallurgical processes. Such pretreatment methods can be electrical, mechanical, or thermal. Prior LiBs discharging and passivation is needed to improve safety and workability, while mechanical treatments such as sieving and magnetic separation are useful to separate the different material streams and to improve recovery efficiency.

Recycling methods being currently developed rely on hydrometallurgy to a larger degree, without involving previous pyrometallurgy steps, in part also because the cost of facilities to implement the methods is smaller. Examples of implemented processes, based on hydrometallurgy, in Europe are Recupyl in France and LithoRec in Germany, with the former developed at an industrially relevant scale and relying on the sole hydrometallurgy (10).

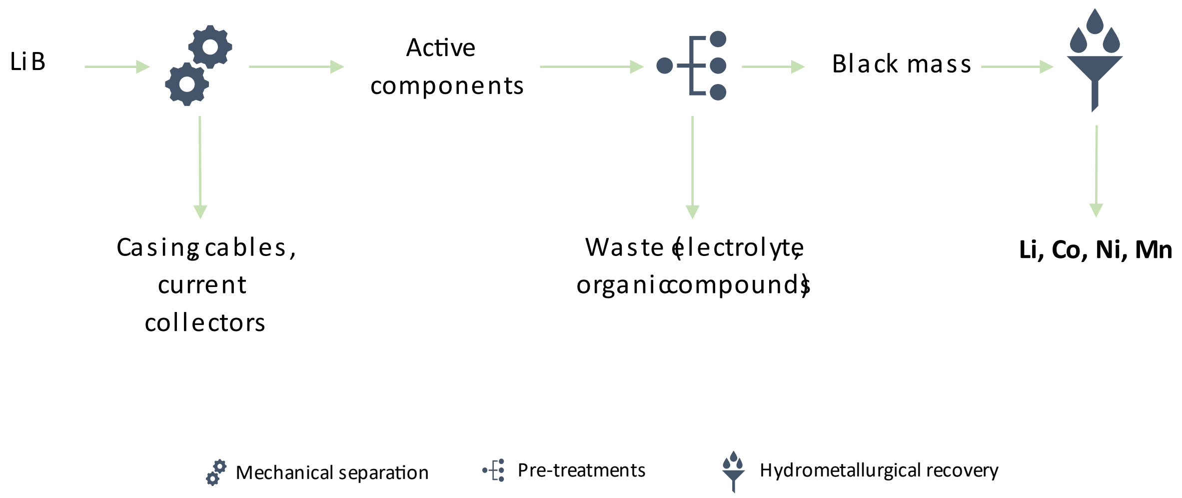

Figure 3 - Hydrometallurgical recycling simplified process flow diagram

Hydrometallurgical recycling techniques present several advantages such as high recycling process efficiency, lower energy consumption than pyrometallurgical methods and pureness of the final products (10). There is however huge consumption of chemical reagents and water, to be eventually purified (2). In addition, for the process to work properly, it is necessary that LIBs be subjected to previous physical pretreatments, to thoroughly separate battery parts for the active components to be in the ideal conditions to be treated. However, this aspect is an opportunity for recyclers to recover a larger share of battery components, not only the active ones, if properly retrieved once separated. According to (2), mixing the anode and cathode materials at the start of the recycling process hampers the metals recovery by hydrometallurgy. A method in which anode and cathode assemblies could be separated prior to mechanical or solvent-based separation would greatly improve material segregation. This is why entire disassembly of batteries is one of several key areas where designing for end-of-life recycling promises to have a real impact. However, thermal treatments are indeed investigated to remove graphite, for instance, and more in general all the organic components. A more detailed focus on pre-treatments will be provided in this document in section 4.1.5.

Table 6 - Advantages and disadvantages of hydrometallurgical recycling processes (10)

Pros	Cons
High flexibility to different battery chemistries	Pre-treatments necessary
High efficiency of materials recovery	Extensive use of chemicals and high volume of waste water
High purity of products	Graphite and organic materials not recovered
Lack of emissions	High operating costs

4.1.4 Direct Recycling

The removal of cathode or anode active materials from the electrode for reconditioning to be reused in a remanufactured LIB is known as direct recycling. In principle, metal-oxide cathode materials can be reincorporated into a new cathode electrode with little changes to the crystal morphology of the active material. Lithium content is to be reactivated by replenishment to compensate for losses due to degradation of the material during battery use (2). Avoiding complex and expensive purification steps, direct recycling could be suitable for lower-value cathodes chemistries such as LMO and LFP. Direct recycling consists indeed of physical and chemical steps with relatively low materials and energy demand:

- reactivation of cathode material may require the addition of chemical reagents, but in lower amount than for hydrometallurgy (1).
- calcination may be required to regenerate generate the new battery materials, energy requirements are likely to be lower than for either pyrometallurgical or hydrometallurgical methods (1).

For its intrinsic nature, direct recycling has the important advantage that, in principle, all battery components can be recovered and re-used after further processing (with the exclusion of separators) (2). The need for physical separation of cathode/anode active materials translates into the opportunity of complete recovery of other components, such as casing, cables and current collectors. Nonetheless, while there is substantial literature regarding the recycling of the cathode component from spent LIBs, research on recycling of graphite anode is limited, owing to its lower economical value. Nevertheless, the successful demonstrations of physically-separated graphite anodes reuse in new batteries was proven, with similar properties to that of pristine graphite (11).

Despite the several advantages of direct recycling, however, considerable obstacles hamper its actual penetration in industrial environments. The efficiency of direct recycling processes is strongly related with the battery state of health. In case of low state of charge or severe damages to the active components, direct recycling may be neither advantageous nor feasible. Potential issues may arise handling different cathode chemistries as well. For maximum efficiency indeed, direct recycling processes should be tailored per each cathode formulations, requiring specific processes per type of cathode materials (2). Future production of batteries should rely on a reduced number of available cathode chemistries. Otherwise, significant manual labour would be required to cope with many different cathode materials. Automation is currently being studied for battery recognition, sorting, and disassembly, however with limited scope and volume (1).

Moreover, direct recycling route for cathode treatments is highly sensitive to contamination by other metals, such as aluminium, characterized by poor electrochemical performance (2). Comminution pre-treatments requiring a high degree of comminution form fine particles of Al and Cu, which are difficult to separate from components to be reactivated, with active materials with poorer electrochemical performances more likely to be obtained (2). Separation of the materials streams before mechanical sorting is hence recommended.

As already mentioned, direct recycling is not commercialised yet. Currently, only some published processes from recycling plant, such as the USA facility of OnTo Technology, are available. However, number of studies on direct recycling is constantly increasing over the years, highlighting the interest on this technology.

Table 7 - Advantages and disadvantages of direct recycling processes (10)

Pros	Cons
Potential recovery of all battery components	Pre-treatments and separation steps necessary
Suitable for “cheaper” cathode formulations	Mix of materials reduces process quality
Lower energy and reagents demand	Generally lower quality of products
Lack of emissions	Not developed industrially yet

4.1.5 Importance of pre-treatments

Current design of cells makes recycling extremely complex and neither hydro nor pyrometallurgy currently can ensure recovery of pure material streams to be easily reused for battery manufacturing purposes (2).

Pre-treatment of spent LIBs is hence critical to maximise the number of recyclable materials, improve efficiency and quality of outputs, especially in the case of hydrometallurgy and direct recycling routes, other than reducing processes energy consumption (7). Performed alone, pre-treatments are not sufficient (as already explained in chapter 4.1.1 about mechanical recycling), but when coupled with further recovery processes they provide real value to the recycling chain.

Such pre-treatments are used for the separation of active materials from the battery casing, separator, current collector, electrolyte, additives, connections and for their preparation for further processing to recover valuable materials. A complete pre-treatment can be thought as a seven-step process (10):

- LiBs **discharging**, usually performed by soaking the cells for 24 hours in aqueous solutions.
- **Dismantling**, often operated manually to remove larger elements such as modules casing and cables. Automated disassembly processes, that would allow faster operations, are now studied.
- **Comminution** step, to reduce active materials in very small particles, is essential for hydrometallurgy recycling while it is not recommended for direct recycling, as already explained before.
- **Classification** (e.g. by sieving) may be operated to allow the separation of particles with different sizes, affecting the overall efficiency of recycling process.
- **Separation**, which is strongly affected by particles' size, is performed by different techniques, each one able to separate different components/materials. The most-commonly used techniques are: *magnetic separation* to remove iron-containing components; *eddy current* separation to split electrical conductors from non-conductive materials providing a thorough separation between Al/Cu and Co/Li; *electric field separation* to divide charged or polarised parts from the rest of the crushed mass.
- **Dissolution**, used mainly to separate residue of active materials from current collectors, kept together by binders.
- **Thermal treatments**: to remove organic materials, carbon conductive agents and binders still present in the black mass, that would hamper the leaching processes of metals recovery. *Pyrolysis* is preferred over incineration for the lower temperatures required. In addition, pyrolysis can be coupled with other treatments such as ultrasonication or microwaving.

4.1.6 Most relevant recycling companies worldwide

A list of the main companies operating LiB recycling worldwide is provided in Table 8 below, summarising what indicated in previous chapters. Peculiar characteristics such as type of battery accepted, type of process, main recoveries and losses are provided, as well as the geographical indication and the technology level, are provided. It is possible to notice that Europe plays a relevant role with several plants operating or developing processes for LiB recycling.

Table 8 - Most relevant recycling processes worldwide

	Country	Scale	Feed	Pre-Processing	Methodology	Main recoveries	Secondary recoveries	Losses
Umicore	UE (Belgium)	Industrial	LiB, NiMH	Dismantling	Pyro + Hydro	Co, Ni, Cu, Ni sulfates, Li2Co3	Slag	Electrolyte, plastic, graphite
Recupyl	UE (France)	Industrial	Primary Li, LiB	-	Mech + Hydro	Li2CO3 LiCO2 Li3PO4 Co	Fe, Al, Cu, MeO, C	Electrolyte, graphite
Accurec	UE (Germany)	Industrial	LiB	Sorting, dismantling	Mech + Pyro + Hydro	Li2CO3 Co-alloy	Metallic alloy	Electrolyte, plastic, graphite

Akkuser	UE (Finland)	Industrial	LiB	Sorting	Mech	Black mass, Fe	Non-ferrous metals	Plastic
Retriev	USA/CAN	Industrial	Primary Li, LiB	Dismantling	Mech + Hydro	Li2CO3 MeO	Fe, Cu, Co, Al	Plastic
LithoRec	UE (Germany)	Emerging	LiB	Discharge, manual disassembly	Mech + Pyro + Hydro	Li2CO3 MeO	Al-Cu, plastic fractions	Electrolyte
Battery Resourcers	USA	Emerging	LiB	Discharge	Mech + Pyro + Hydro	Li2CO3 NMC(OH)2	Ferrous metals	Electrolyte
OnTo	USA	Emerging	Primary Li, LiB	Discharge, dismantling	Direct recycling	Refurbished cathode powder	Fe- and non-fe metals	Binder

4.2 Focus on SAFELiMOVE-specific battery materials recycling

Although several recycling technologies are available for LiBs, the transferability to solid-state batteries (SSB), as the SAFELiMOVE one, is not immediate. Even though cathode chemistries are the same, differences in used materials and fabrication technologies require other procedures to make solid-state battery components recyclable (12). Main challenges consist in handling the solid electrolyte and, especially in the case of SAFELiMOVE battery, the lithium metal anode. Recycling of SSB has not been industrially developed yet, thus a literature review was performed focusing on solid electrolyte and lithium metal anode recycling processes. Even though current literature on this topic is not that vast, more interest is increasingly being put on it, with recent studies demonstrating viable concepts to be applied to SSB recycling (13).

4.2.1 LiM recycling – opportunities and challenges

Lithium metal is mainly explored as potential solution in solid state batteries due to the higher energy density which can be achieved in such configuration. On the other hand, the adoption of Lithium metal introduces a series of battery handling issues which can significantly affect the recycling potential at the end of life. This section is devoted to explain why Lithium metal recycling is considered as a hard challenge compared to LiB and to explore which are the current best practices and technologies to effectively recycle this material.

Main concern associated with solid state batteries is the presence of Lithium in metallic form. In the SAFELiMOVE configuration, battery anode is mainly composed by Lithium metal. Unlike Lithium-ion batteries, solid state ones present several concerns in terms of handling at the end of life, as the Lithium metal is rather reactive; in addition to this issue, the physical condition of the Lithium tend to generate handling troubles due to the sticky attitude of the Lithium metal adopted for the anode (14). Furthermore, Lithium metal anodes are prone to break up in small particles which can generate troubles for the afore-mentioned reasons.

These considerations lead to the first conclusion that a conventional mechanical treatment (such as shredding as described in 4.1.1) is not applicable as the gluey nature of Lithium metal would quickly seize up the whole equipment. On the other hand, due to the high reactivity, water-based shredding is not applicable either due to the risk of flares and explosions associated with the Lithium metal reacting with atmosphere gases. To avoid all the issues described before, a controlled atmosphere is required to limit Lithium reactivity and therefore allow Lithium metal shredding. This technique is not yet commercially available at large scale due to the high implementation costs coupled with a low market availability of spent Lithium metal battery anodes. The potential market penetration for solid state batteries will be the driver

variable forcing recycling technologies to stand up and explore unconventional routes bringing them to a reasonable degree of industrial maturity.
At the time being, it must be transparently stated that Lithium metal anodes pose a serious question for what concerns end of life recovery, as the conventional battery recycling processes (still not very competitive for LiB and in ramp-up phase) are not designed to handle such kind of material.
Interesting studies such as (12) highlight the possibility to apply thermal treatments coupled with CO_2 input flows to obtain Li_2CO_3 which is a valuable precursor to other battery components. This technology appears to be even more promising when coupled with captured CO_2 from atmosphere or other emitting sources; although this cannot be seen as an effective Carbon adsorption sink, this combination may reduce the costs in case waste CO_2 becomes available at competitive market value. Main concern of these exploratory techniques is that the problem of separating Lithium metal handling its sticky and reactive nature is still not solved yet.
As for hydrometallurgy techniques, which are usually well suited to handle several kinds of chemical compounds, some concerns still endure. In particular, according to (13), Lithium metal can violently react in the leaching solution provoking dangerous reactions and limiting the overall process efficiency. Research on primary batteries (non-rechargeable batteries based on lithium-metal anode) highlights the effectiveness of Lithium metal thermal treatments (around 400 °C) prior to dissolution in water to reduce energy release up to 70% (15), providing high yield and purity of Li_2CO_3 obtained.
To sum up, there are currently no mature technologies designed to recover Lithium metal in an effective way. This can be due to the lack of offer, which limits both investments and R&D activities in this field, but the chemical nature of Lithium metal introduces a series of complex issues which might not be solved by extensive research only. Controlled-atmosphere shredding and aqueous solution treatment coupled with a prior thermal treatment to reduce energy release seems the sole promising way to separate and potentially recover Lithium metal. However, such process stays only at research state due to very high implementing costs, energy demand and limited knowhow concerning overall efficiency.

4.2.2 Solid electrolyte vs liquid electrolyte

Due to its liquid nature, electrolyte recovery in state-of-the-art LIBs is not possible. During pre-treatments indeed, the liquid electrolyte is usually washed away, or it evaporates when thermal treatments are applied. Not only it is challenging to handle, but also the liquid electrolyte would hamper subsequent recycling processes of other components due to its high reactivity. In addition, liquid electrolyte removal poses several risks regarding ecotoxicity: when evaporating or being removed, fluorine- or phosphorous-containing off-gases can generate, that are extremely toxic.

Despite several challenges related to them, SSBs as the SAFELiMOVE one are an opportunity regarding electrolyte recovery and recycling. The solid form allows an easier handling of the electrolyte, while eliminating any risk of off-gas toxic emissions. Furthermore, the significant economic value of materials composing the solid electrolyte (especially in the case of oxidic and composite electrolytes, where materials such as La, Ti, Zr are involved) is an additional driver to push its recyclability (13). Nonetheless, any polymeric part of the solid electrolyte would be removed in most cases. This is mostly because of its lower economic value and because its presence would hinder the hydrometallurgical steps of materials recovery. Furthermore, while providing opportunities for higher shares of battery recycling compared to liquid-electrolyte LIBs, handling the solid electrolyte poses several challenges that hence require complex procedures and processes.

The first obstacle is due to the very nature of the materials involved. To ensure proper stability, solid electrolytes require sintering between battery components, making it impractical to physically separate cathode materials from electrolyte ones. However, mechanical separation step is necessary to obtain a black mass suitable for further recovery processes, as no other practical methods currently exist at industrial-scale levels. (13) Further research should hence focus on effective separation of solid electrolytes materials from black mass active materials, that would allow an efficient materials recovery from the different battery components at the same time.

Provided that a proper separation of electrolyte materials is performed, **a combination of hydrometallurgy and direct recycling** seems the best way for solid state battery materials recovery. Proper adjustments must be however operated to copy with solid electrolyte materials complexities and sintered interfaces with cathode materials (13). Pyrometallurgy recycling method would not be sustainable for electrolyte recycling. Only the recovery of cobalt and nickel would be maximized, with all other materials ending up in the slag making them hard to be recovered, even valuable elements such as La and Zr.

Hydrometallurgical methods represent a promising route, especially regarding oxide-based solid electrolytes, taking advantage of the know-how already developed for the techniques developed to recover cathode materials. Elements such as La, Zr and Ti show indeed a considerable difference in solubility than cathode materials (such as Co, Ni and Mn), making the selective precipitation in hydrometallurgy solution possible with careful selection of reaction conditions (13). However, to effectively dissolve oxidic electrolytes strong acid solutions must be used. Environmental concerns may arise due to the adoption of such reagents and particular care must be taken with the waste solution. When it comes to polymer-based electrolytes, even though they may be soluble aqueous acid solutions for hydrometallurgy, they can alter the viscosity of the solution, with a potentially negative effect on co-precipitation processes.

An example of developed recycling path for solid state batteries (at prototype level), based on hydrometallurgy, is proposed by (12) and showed in Figure 4 below. A solid-state cell with LLZO electrolyte, Lithium metal anode and NMC811 cathode was studied, very close in composition to SAFELiMOVE cell. A multi-step hydrometallurgical procedure was developed allowing the recovery of as much materials as possible, not only from cathode but also from anode and solid electrolyte.

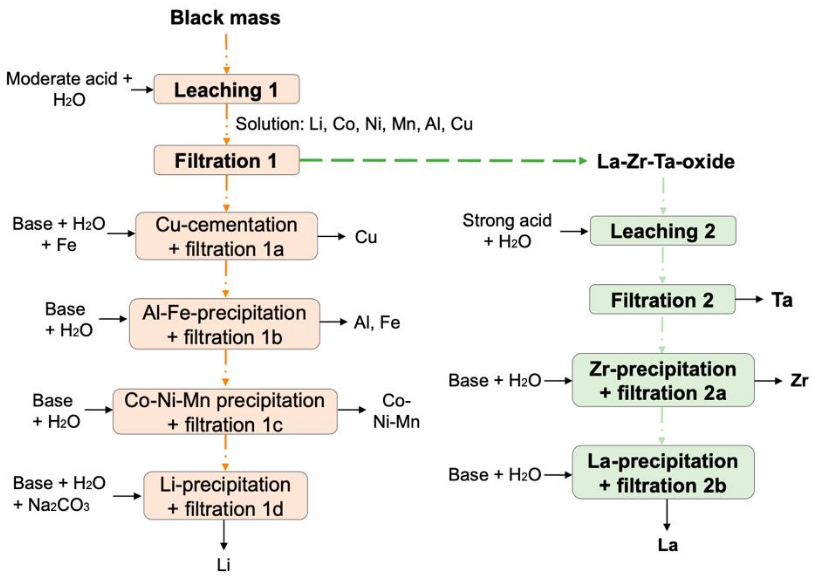

Figure 4 - Proposed hydrometallurgical recycling path for solid state battery (12)

It was found that direct recycling might also be possible for solid state batteries, adopting either hydrothermal regeneration or dissolution/precipitation processes with subsequent heat treatment (13). Regeneration using hydrothermal regeneration are being extensively studied for oxide-based cathodes, but also being explored for solid electrolyte due to their similar chemistry (15). Successful synthesis of LATP electrolyte using hydrothermal methods was indeed proved (16). Direct recycling of both electrolyte and electrolyte can be particularly advantageous for solid state batteries, where the two components are closely mixed or co-sintered. Dissolution/precipitation methods can be interesting when it comes to polymer-based solid electrolytes, e.g. made out of PEO. It is soluble in water and some polar solvents, such as acetonitrile, potentially providing an opportunity for separating it from the rest of the cell components or black mass (13). In case of hybrid electrolytes, such as SAFELiMOVE battery, a combination of the above-mentioned methods might be used to maximise electrolyte recovery.

4.3 Most promising battery recycling routes

This section is intended to provide synthesis of all considerations reported in previous chapters of this deliverable. Based on the outcomes of the battery recycling technology review, the following recycling processes have been identified for batteries for e-mobility:

1. Mechanical treatments
2. Pyrometallurgical processing
3. Hydrometallurgical processing
4. Direct recycling

Each of these routes presents pros and cons which have been thoroughly described within this document; as the main goal of this deliverable is to support SAFELiMOVE consortium in optimizing design for recycling of the final battery, a qualitative rating for the most suitable technology for generic battery recycling has been developed.
The rating scheme is designed considering 7 dimensions of relevant metrics, reported in Table 9. Each of these dimensions is evaluated with a qualitative rating from 1 to 3, where 3 represents the “good” value for the investigated metric while 1 represents a “poor” value of the metric. Each dimension is then associated with a weight factor which reflects its relevance according to the ultimate goal of the project. In such perspective, dimensions like material and energy intensity as well as industrial maturity have been considered with lower weight factor, as the goal of this study is to identify the most promising technology without considering market-related issues (such as material or energy costs). It must be noticed that energy intensity issue can be tackled by supplying renewable energy, thus limiting the environmental footprint of the recycling process.

Table 9 - Battery recycling score parameters

Dimension	Description	Weight factor
Recycling process coverage	Percentage of the recycling process which can be handled by the technology (i.e. how many other treatments shall be coupled with it to ensure proper recycling)	20%
Material recovery efficiency	How much battery material can be recovered by the target technology (considering as well quality and scarcity of the target materials)	20%
Process energy intensity	Amount of electric and thermal energy required to run the process (with direct effect on environmental footprint)	10%
Process material intensity	Amount of ancillary materials required to run the process (e.g. acids, water, solvents, etc.)	5%
Process industrial maturity	Technology readiness at market scale in a mature scenario where large amount of spent EV batteries becomes available	10%

Battery chemistry range	Range of battery chemistry where the technology is applicable	20%
Recovered materials quality	Quality of the material which is recovered by the technology	15%

Results of the application of the battery recycling rating is reported below; according to the rationale behind the index, higher value for a parameter means a better performance in the investigated dimension. Therefore, an intensity dimension marked with "3" identifies a technology with low consumption of resources. The overall score is then computed as weighted average and reported in Table 11.

Table 10 - technology recycling rating: dimension evaluation

Technology	Recycling process coverage	Material recovery efficiency	Process energy intensity	Process material intensity	Process industrial maturity	Battery chemistry range	Recovered materials quality
Mechanical recycling	1	2	3	3	2	3	1
Pyrometallurgy	2	2	1	3	3	1	2
Hydrometallurgy	2	3	2	1	2	3	3
Direct recycling	3	2	3	2	1	3	2

Table 11 – technology recycling rating: final assessment

Technology		Total score
Hydrometallurgy		2,5
Direct recycling		2,4
Mechanical recycling		2
Pyrometallurgy		1,85

According to the dimension described above, hydrometallurgy appears as the most appropriate recycling route for battery devoted to e-mobility purposes. Direct recycling presents as well a promising score, which is basically affected by a lower quality of the recycled material; this is due to the fact that direct recycling is a kind of refurbishment process which progressively lowers the quality of the components and implies a downgrade in operating conditions, other than being considerably affected by the quality of the incoming feedstock. These two technologies are by far the most appropriate when the North star metric is the quality of the recycled material and the independency of the process.

In such perspective, mechanical recycling and pyrometallurgy present lower performances. The former is affected by strong issues related to the additional processes required to obtain a good recycling material quality, while the latter is penalized by the small range of battery chemistries which can be treated. In particular, innovative chemistries (e.g. SAFELiMOVE one and other solid-state batteries) and emerging ones (Iron-phosphate) cannot be effectively treated via pyrometallurgy in its current configuration.

It shall be mentioned that this rating is a quali-quantitative score developed for research purposes in the framework of the SAFELiMOVE project. The aim is to quickly identify which is the best performer according to a set of parameters which relevance is assigned based on project goal and European strategies: different dimensions and different weight factors might then lead to completely different conclusions.

It can still be concluded that direct recycling and hydrometallurgy are the most promising routes considering output quality and applicability range; mechanical recycling simply cannot be applied alone, so the coverage and the independency are quite low, while pyrometallurgy is well-suited for a certain range of battery chemistries only, while energy intensity issues still endure.

4.4 SAFELiMOVE battery recycling procedure and outcomes

As final outcome of this deliverable, an evaluation on the potential share of SAFELiMOVE cell materials/components recovery is provided, adopting the best recycling technology illustrated in the previous chapter. Figure 5 below illustrates the SAFELiMOVE 1 Ah cell composition, as shared by the consortium technical partners. 1 Ah cell is considered as SAFELiMOVE benchmark cell for this deliverable, being the cell assembled by the consortium with the last available level of materials developed (2nd generation).

Already considering currently available technologies for battery recycling, SAFELiMOVE cell at end of life would be a valuable feed for recyclers, thanks to its NMC811 cathode chemistry. Even though cobalt content is lower compared to other NMC cathodes, the higher share of nickel (ca. 12% wt. of active cathode material) makes it suitable and profitable for pyrometallurgical processes. However, apart from Al current collector, only valuable metals in the cathode would be recovered.

Considering an enhanced process of hydrometallurgy tailored to accept solid state batteries, hence able to recover also catholyte components and oxidic electrolyte parts, the recovery rate would be considerably higher and compliant with the requirements of European directive (2006/66-EC), not only at battery level but already at cell level. A thorough hydrometallurgy process, coupled with proper pre-treatments, would allow the recovery of :

- Al current collector (ca. 6% wt.)
- entire cathode active material, not only valuable metals but also lithium (ca. 35% wt.)
- catholyte components, due to their nature similar to cathode materials (ca. 10%)
- Oxidic electrolyte parts (ca. 2% wt.)

hence going beyond the 50% wt. threshold indicated the EC proposal already at cell level (thus not considering BMS and module casing).

If properly designed, cell packaging could be easily recycled as well. Here is assumed as not recyclable, as made out of an Al-coated multilayer of different polymers (PP+PA). However, the simple adoption of a single polymeric material (es. polyolefin) would make its recovery feasible.

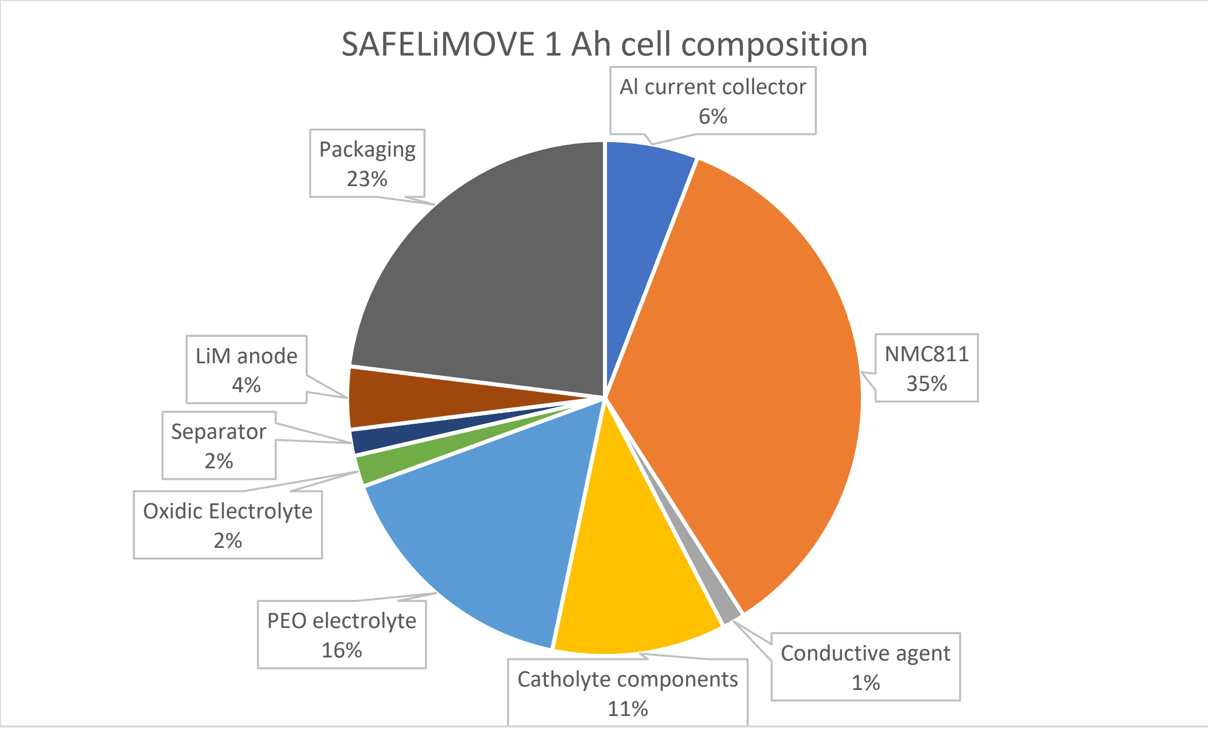

Figure 5 - SAFELiMOVE 1 Ah cell composition

Organic components such as conductive agents and polymeric electrolyte parts, as well as the lithium-metal anode, still pose challenges for their recovery in an effective way, even with more advanced recycling technologies being studied nowadays. However, some direct recycling techniques might allow the recovery of the polymeric part of the electrolyte as well, as already explained in chapter 4.2.2. This process should be coupled with further hydrometallurgy to recover the remaining oxidic part of the electrolyte, as well as all other active cell components.

5 Conclusions and Recommendations

The huge number of waste EV batteries foreseen for the incoming years, generated by the rapid EV market growth happening to meet global transport sector GHG-emissions reduction, poses not only serious challenges but also offers several opportunities to the whole electric mobility chain as well: spent EV batteries have the potential to become a valuable source of materials for new battery production, as well as a potential still-exploitable source of electrical energy.

Acknowledging this aspect, the European Commission proposed in 2022 an update of the current Battery Directive 2006/66-EC, indicating future policy frameworks for battery eco-design and recycling at end of life with particular focus on EV batteries.

In the meantime, battery recycling technologies are being both developed at industrially relevant scale and extensively investigated in the research field. According to the combined effect of recycling feasibility and final gain, currently only valuable metals such as cobalt, copper, steel, nickel, and aluminum are actually recycled by industrial processes. Those are commonly recovered as metallic alloys from pyrometallurgical processes or, in the case of large casing materials, during mechanical dismantling. As a result, industrial recycling processes are mainly driven by the cathode-chemistry market value, meaning that only certain cathode chemistries can be recycled in an economically sustainable way. Recycling processes based entirely on hydrometallurgy, coupled with proper mechanical and thermal pre-treatments, should guarantee a higher share of components recovery, also including lithium and less valuable materials, always ensuring a high purity. Currently such technology is not extensively adopted for the high cost and lower revenues from materials recovered compared to pyrometallurgy. However, with the huge number of batteries reaching the end-of-life stage foreseen for the incoming years and, more importantly, with the development of cathode chemistries free from heavy metals such as cobalt, hydrometallurgy process is likely to become the main recycling process. The concept of direct recycling is finding a strong interest in the research field. The opportunity to replenish battery materials instead of recycling them is an interesting aspect, proved to be technically feasible and to be able to provide batteries ready for further usage. However, this technology is currently developed only at pilot scale, due to some intrinsic challenges due to heterogeneity and complexity of processes involved.

All the above-mentioned recycling processes are developed focusing on currently used batteries for the automotive sector, namely lithium-ion batteries (LIBs). Even though similar to LIBs, solid-state batteries (SSBs) add challenges to their potential recycling, due to intrinsic features of their composition. Techniques to deal with SSB are being extensively studied. The solid electrolyte, especially if composed by oxidic materials, have the potential to be recovered by hydrometallurgy processes, however with a more extensive use of strong leaching agents and solvents, making the process even more complex. However, being similar to cathode materials, the oxidic solid electrolyte provide also opportunities for higher share of battery recovery, if proper leaching procedures are developed. This is not true for lithium metal, which nowadays represents an important obstacle for SSB recycling. Due to its stickiness, it is currently not recyclable, and, in addition, it hampers the recovery of other battery components. Further research on techniques and procedures to handle it in protected spaces is being however carried on, with prior thermal treatments showing promising results in reducing its reactivity in aqueous solutions.

Finally, to provide a synthesis of the literature review activity on battery recycling performed in this work, a qualitative rating for the most suitable technology for generic battery recycling has been developed, considering a metric of seven relevant parameters assessing the processes' most relevant hotspots. As a result, a recycling process based mainly on hydrometallurgy (coupled with proper mechanical/thermal pre-treatments) is considered the best option for battery components recovery, as a combination of efficiency, quantity and purity of materials recovered, flexibility of battery chemistries accepted and industrial feasibility. Such process is not only suitable for LIBs, but seems also the most appropriate for SSBs, offering promising opportunities for the recovery of solid electrolyte.

6 Risk Register

No additional risks identified other than the ones reported in the Risk Management Plan.

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Appendix A- Acknowledgement

The author(s) would like to thank the partners in the project for their valuable comments on previous drafts and for performing the review.

Project partners:

#	Partner	Partner Full Name
1	CICe	CENTRO DE INVESTIGACION COOPERATIVA DE ENERGIAS ALTERNATIVAS FUNDACION, CIC ENERGIGUNE FUNDAZIOA
2	SCHOTT	SCHOTT AG
3	UMICORE	UMICORE
4	HYDRO-QUEBEC	HYDRO-QUEBEC
5	SAFT	SAFT
6	RENAULT SAS	RENAULT SAS
7	TME	TOYOTA MOTOR EUROPE NV
8	IKERLAN	IKERLAN S. COOP
9	CEA	COMMISSARIAT A L ENERGIE ATOMIQUE ET AUX ENERGIES ALTERNATIVES
10	CIDETEC	FUNDACION CIDETEC
11	TUB	TECHNISCHE UNIVERSITAT BERLIN
12	RWTH AACHEN	RHEINISCH-WESTFAELISCHE TECHNISCHE HOCHSCHULE AACHEN
13	ABEE	AVESTA BATTERY & ENERGY ENGINEERING
14	LCE Srl	LIFE CYCLE ENGINEERING SRL
15	UNIRESEARCH BV	UNIRESEARCH BV