FREE4LIB

Feasible **RE**covery of critical raw materials through a new circular **E**cosystem **FOR** a **Li-Ion B**attery cross-value chain in Europe

Clustering workshop “Production of raw materials for batteries from European resources”

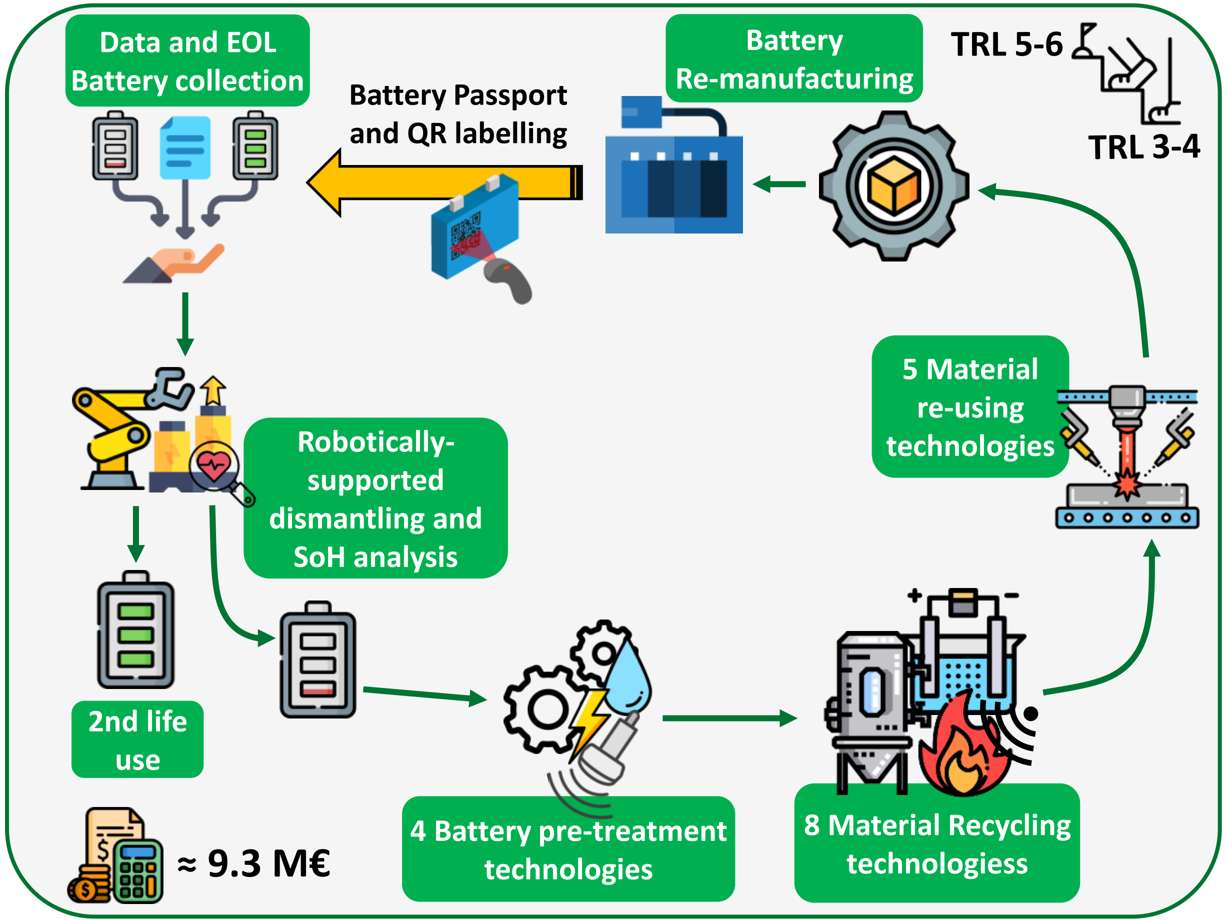
BSc. Juan Castro - FREE4LIB Coordinator

Project Summary

- **Create feasible and holistic recycling processes** by analysing and evaluating data from LIBs.
- **Develop sustainable and efficient technological solutions for recycling different Li-battery chemistries and material re-using** based on intelligent process design to optimise its scale up.
- **Recover higher amount of resources from spent LIBs to use as secondary raw materials in new batteries** based on a sustainable transferability model to improve vertical integration on manufacturing.
- **Design for Recycling** (DfR) of new Li-batteries
- **Create a Battery Passport and battery recycling modelling platforms**
- **Research on 21 technologies covering the entire Li-ion battery value chain**

Consortium of **22 partners** from **7 different countries**, coordinated by CARTIF:

CARTIF, CSIC (Consejo Superior de Investigaciones Científicas), EUROBAT, Fondazione Bruno Kessler, Uni Graz, eurecat, ALIÉNOR (Tailor-made solutions), ITL (Istituto sui Trasporti e la Logistica), IREC, Fraunhofer IWKS, EID, TORRECID, Sakarya University, ACCUREC Recycling GmbH, WATT4EVER (Batteries for life), Politecnico di Milano, erion, AVL, next move, NESSTEC (New Energy Storage Systems & Surface Technologies), Lurederra centro tecnológico, AIMPLAS, recyclia



Project duration: from September 2022 to September 2026

Funded by the European Union

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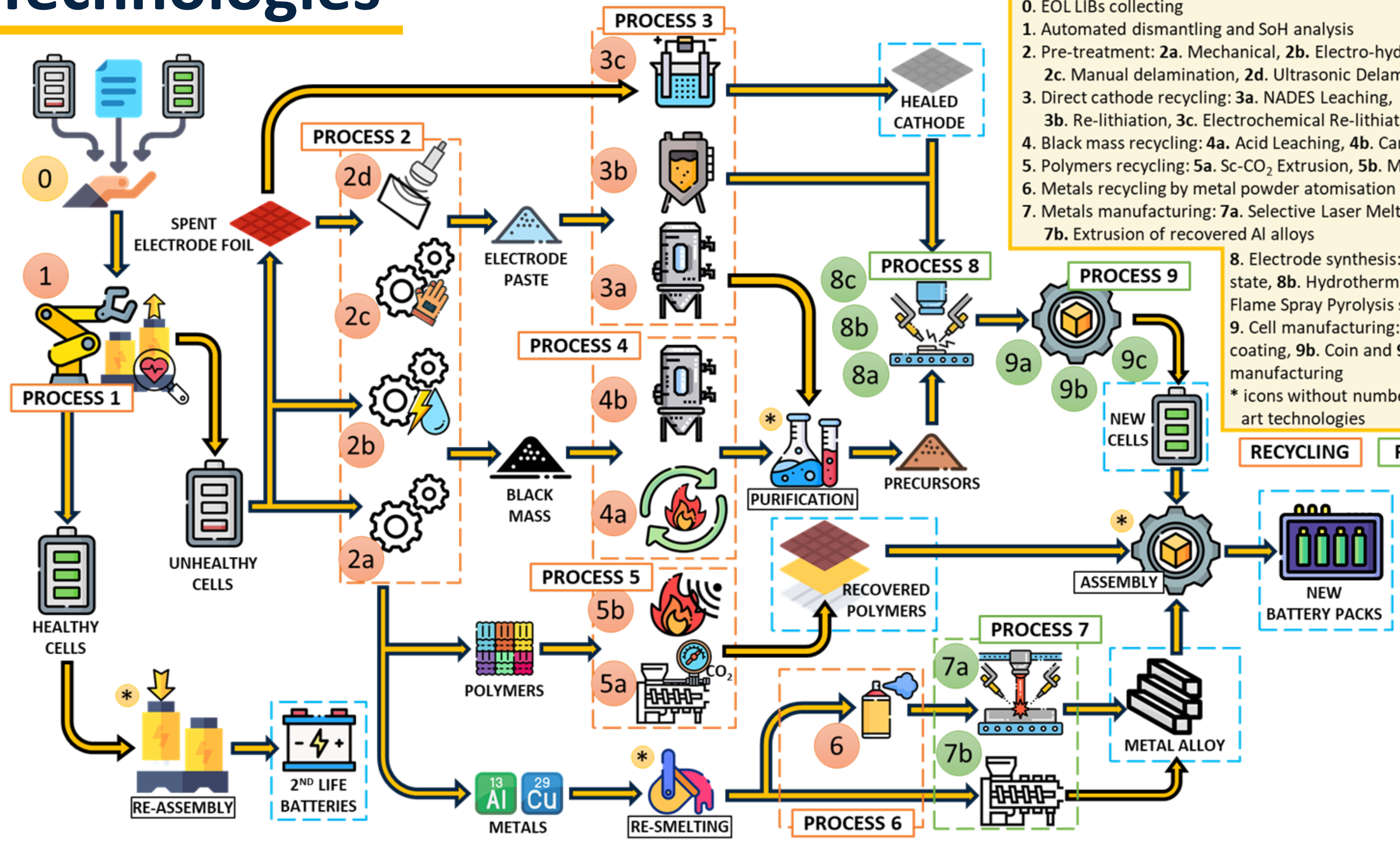
FREE4LIB

Raw Materials Week - Brussels, 14th November 2022

Juan Castro - Circular Economy Researcher

TECHNOLOGY CENTRE CARTIF

Technologies



0. EOL LIBs collecting
1. Automated dismantling and SoH analysis
2. Pre-treatment: **2a.** Mechanical, **2b.** Electro-hydro, **2c.** Manual delamination, **2d.** Ultrasonic Delamination
3. Direct cathode recycling: **3a.** NADES Leaching, **3b.** Re-lithiation, **3c.** Electrochemical Re-lithiation
4. Black mass recycling: **4a.** Acid Leaching, **4b.** Carboreduction
5. Polymers recycling: **5a.** Sc-CO_2 Extrusion, **5b.** MW Pyrolysis
6. Metals recycling by metal powder atomisation
7. Metals manufacturing: **7a.** Selective Laser Melting and **7b.** Extrusion of recovered Al alloys
8. Electrode synthesis: **8a.** Solid state, **8b.** Hydrothermal and **8c.** Flame Spray Pyrolysis synthesis
9. Cell manufacturing: **9a.** Electrode coating, **9b.** Coin and **9c.** Pouch cell manufacturing
* icons without number are state of art technologies

PROCESS 1
Automated dismantling, SoH analysis and 2nd life batteries

PROCESS 2
Pre-treatment

PROCESS 3
Electrode recycling

PROCESS 4
Black Mass recycling

PROCESS 5
Polymers recycling

PROCESS 6
Metals recycling

PROCESS 7
Metals manufacturing

PROCESS 8
Electrode Synthesis

PROCESS 9
Cell & Battery Pack Manufacturing

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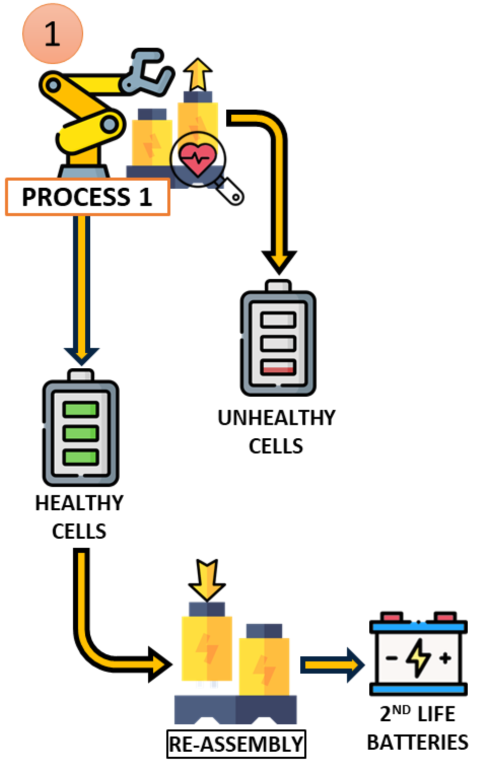
Recycling Technologies

1. Dismantling and SoH analysis



Reduction of risks associated with LIBs handling and manipulation, due to the presence of residual stored energy is the aim of **Robotically-supported dismantling of battery packs technology**. Dismantling will happen down from the battery pack to the module level by integrating and validating the robotized tasks to accelerate and facilitate the dismantling process.

State of Health tool will be developed to determine cells remaining lifetime with as little effort and time as possible. I will improve existing experimental methods by using and/or combining machine learning (such as *Neural Networks, Support Vector Regression or Fuzzy Logic technologies*), and model-based methods (such as *electrochemical models (ECM) techniques*).

Unhealthy cells will be processed by the pre-treatment step and healthy cells will be re-assembly for use on a **2nd life battery pack**.

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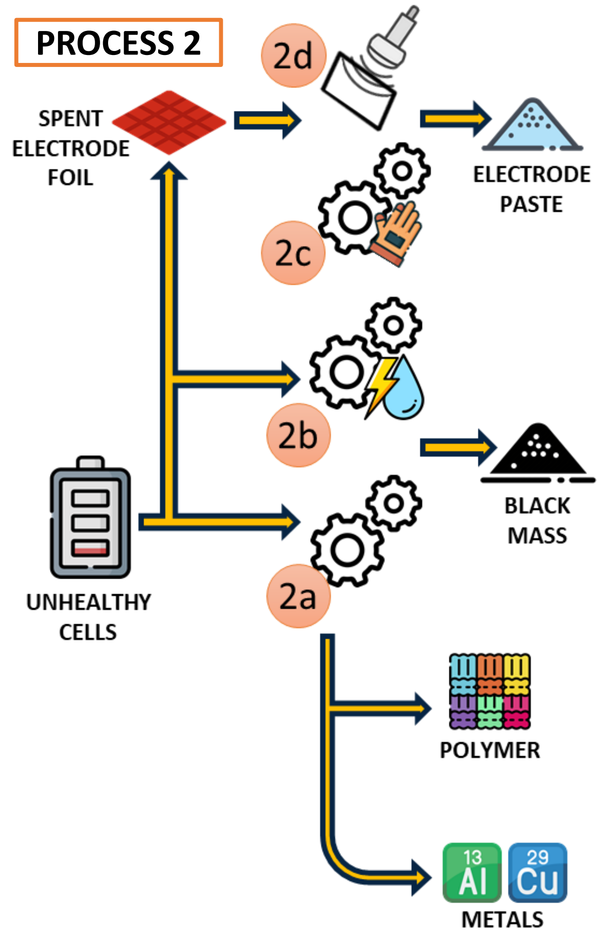
Recycling Technologies

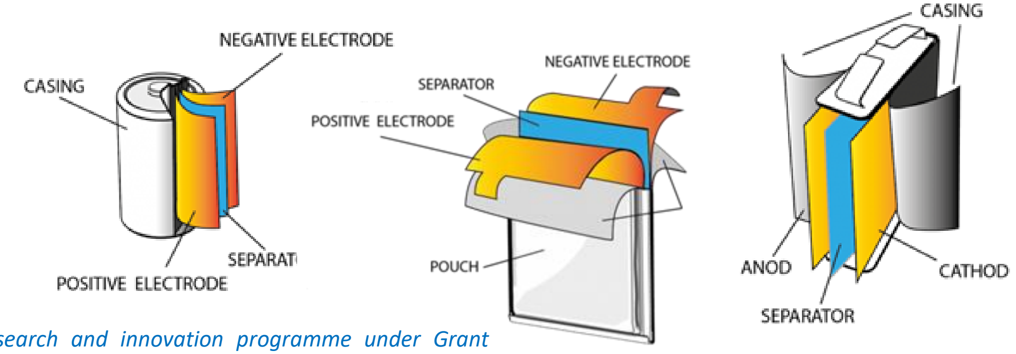
2. Four pre-treatment technologies



Mechanical pre-treatment (2a) and **Electro-hydro fragmentation (EHF) (2b)** for the black mass, Cu/Al foils, ferrous/steel fractions and polymer separation and delivered to further recycling processes.

Electrode Delamination under 2 different pre-treatment techniques: **Manual delamination (2c) and Ultrasonic delamination (2d)** for the application of electrode direct recycling technologies depending on the subsequent recycling process. It consists of a two-step process, (i) electrical cell deactivation followed by (ii) a non-breaking of cells to open, manually at laboratory scale and optimised to implement with larger equipment to obtain enough electrode materials for its further recycling step.

For cylindrical, pouch and prismatic cells



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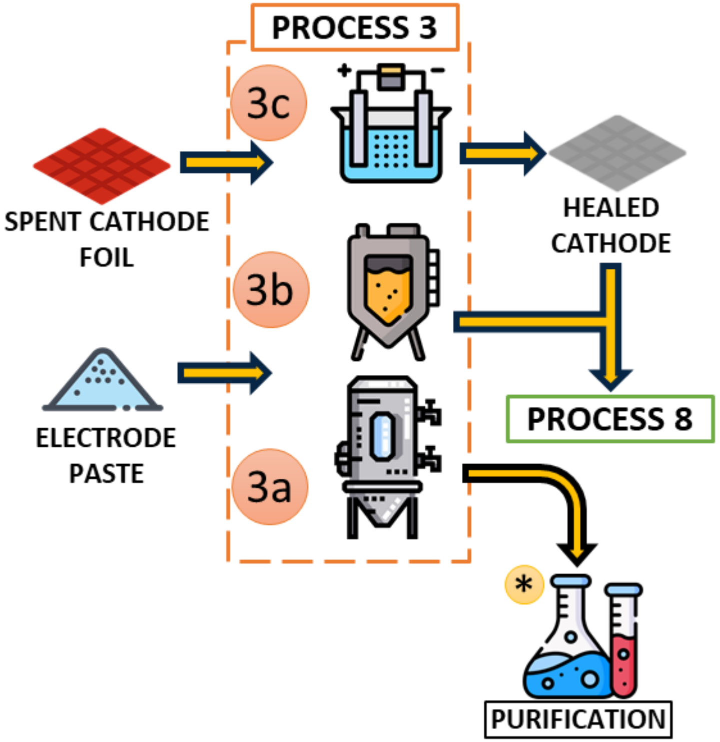
Recycling Technologies

3. Electrodes Direct recycling



A hydrometallurgical recycling process for major metal components of the recovered cathode paste. A leaching step based on **Natural Deep Eutectic Solvent (NADES) leaching (3a)** followed by a selective precipitation step to recover valuable compounds.

After ultrasonic delamination of LIB electrodes, the active material (electrode powder) recovered will be restored to its original performance by **hydrothermal re-lithiation (3b)** followed by a short annealing step without destroying electrode morphology.

After the mechanical separation of cells, without delamination steps, an **electrochemical re-lithiation (3c)** process is applied directly to the cathode foils.

Depending on the direct recycling process, the following step will be a purification (for NADES leaching) or an electrode synthesis process (for re-lithiation).

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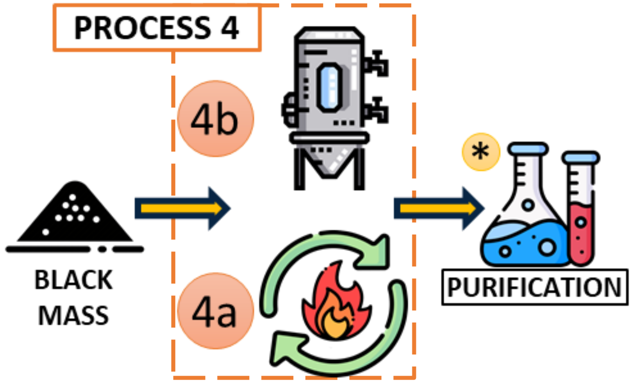
Recycling Technologies

4. Black Mass recycling



Carboreduction of black mass (4a) will be based on a pyro-metallurgical low-temperature thermal treatment to recover Lithium by a carboreduction process. On the other hand, rest of metal oxides will be found at slag stream which under physical separation and/or hydrometallurgical process will be recovered too, depends on slag composition.

Hydrometallurgical technology, in this case, **acid leaching of black mass (4b)** (organic and inorganic acids). Following a purification process will be needed to carry out selective separation of each metal oxide leached previously.

Different **purification of leachate techniques** will be studied like non-extraction subsequently process to directly re-use leachate concentrates of Ni, Mn and Co as precursors without previous individual separation of each one.

On the other hand, to maximize the recovery target of metal oxides, co-extraction by **Flame Spray Pyrolysis** routes and wet routes will be carried out. The final chemistry of the co-extraction and the presence of impurities will be adjusted and assessed according to the synthesis route.

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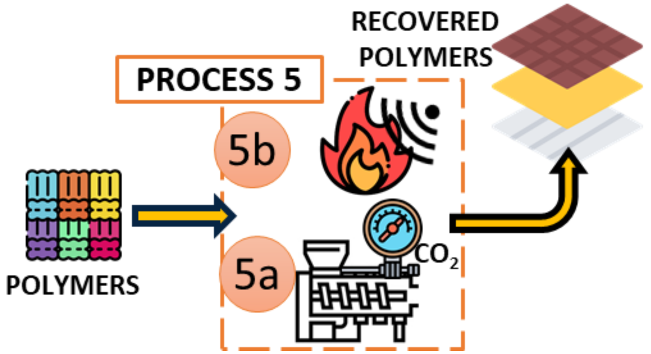
Recycling Technologies

5. Polymers recycling



Screening the different types of polymeric materials and sorting them in different streams for proper polymers valorisation routes such as (i) Separation based on electrostatic and triboelectric properties; (ii) Elutriation air system for separation based on density; (iii) Continuous conveyor equipment with NIR, optical VIS and hyperspectral camera to identify polymers depending on their chemical structure (i.e. PP, PE, ABS, PET…).

Decontamination of recovered thermoplastics by sc-CO_2 (5a) will be implemented as an innovative extrusion technology in the reprocessing of thermoplastic materials (i.e. PP, ABS) with the aim of removing volatile and contaminant substances.

Upcycling of thermoplastic compounds for material re-use will be researched aiming at improving thermoplastic compounds formulation performance by adding speciality additives such as compatibilizers, impact modifiers, stabilizers, coupling agents, chain extenders or fillers or reinforcing fibres.

Regarding thermoset materials, classified as non-reusable for battery packs assembly, will be harnessed by **Thermoset recovery (microwave-assisted pyrolysis technology) (5b)** which accepts waste from multiple sectors, making it a versatile technique

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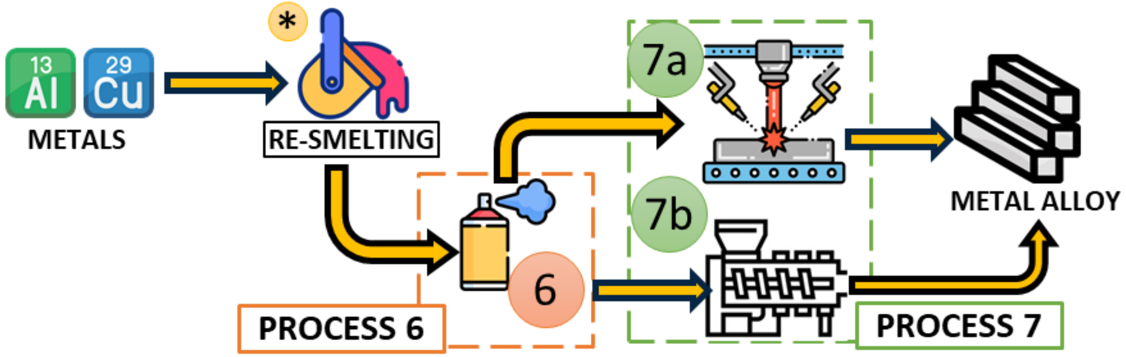
Recycling & Re-use Technologies

6. Metals recycling 7. Metals Manufacturing



Separated metals from the pre-treatment process will be smelted to obtain new cases for FREE4LIB LIBs.

Two refined routes will be studied in order to add alloying elements and pure aluminium to get the desired composition according to Battery Packs requirements

After smelting, the study of the powder atomisation production by gas powder atomisation and **centrifugal powder atomisation (6)** will be carried out, as well as the composition and optimal printing parameters to finally obtain an alloy with good mechanical characteristics, at least similar to AlSi10Mg.

Then, two routes of alloy forming processes will be defined by **Extrusion of recovered Al alloys (7b)** and powder metallurgy focusing on **Additive Manufacturing (AM) by Selective Laser Melting (SLM) technology (7a)**.

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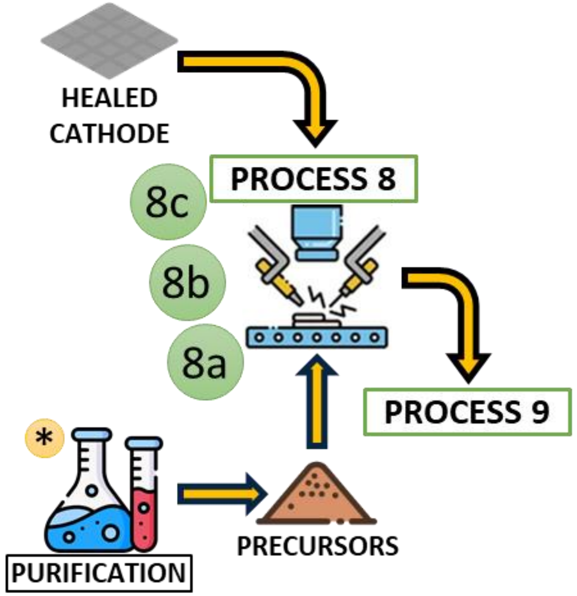
Material Re-use Technologies

8. Electrode synthesis



The synthesis of LMNO and NMC material from recycled raw materials will be studied by **Solid state and Hydrothermal electrode synthesis routes (8a & 8b, respectively)**. On the other hand, co-extraction of precursors will be taken by **Flame Spray Pyrolysis (FPS) technology (8c).**

Solid-state consists of dry grinding of the raw materials, calcination between 900 and 1.200 °C and subsequent grinding to obtain a powder between 5-10 µm and Hydrothermal route, starting from raw materials such as Li nitrate, nitrates/sulphates of nickel and manganese will be synthesized hydrothermally.

FPS consists of a one-step continuous process, offering scalability and liquid waste reduction advantages. A precursor mixture containing the metals of interest in the right reaction is atomized to create a spray of aerosol droplets that are ignited to form a flame. The precursors rapidly decompose followed by a phase transition to vapour and homogeneous reactions to form the oxide particles

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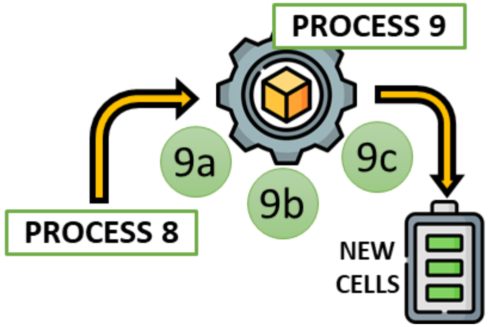
Material Re-use Technologies

9. Cell Manufacturing



New coating strategies to enhance the electrochemical performance of recycled materials will be developed. The materials will be optimized by means of PVD (Physical Vapor Deposition) coatings to enhance their conductivity, chemical/structural stability and Li^+ kinetics. **Electrode coating (9a)** will also improve charge-discharge efficiency.

After coating technology, **coin cell manufacturing (9b)** will use recovered redox-active materials and foils to process the electrodes to evaluate their performance at half-cell (vs. Li metal) coin cell level. The electrochemical performance of LMNO, NMC811, NMC622 and graphite will be evaluated. The materials should deliver capacities in the range of 135, 180, 160 and 350 $mAh \cdot g^{-1}$, respectively, at 0.1C for at least 100 cycles to validate their performance at the coin-cell level.

On the other hand, **pouch cells manufacturing (9c)** with commercial active materials and commercial foils; and recycled active cathode materials, graphite and aluminium and copper foils will be carried out for their comparison.

Produced pouch cells will be used for their assembly in new battery packs following the Design for Recycling (DfR) Guidelines promote in the project.

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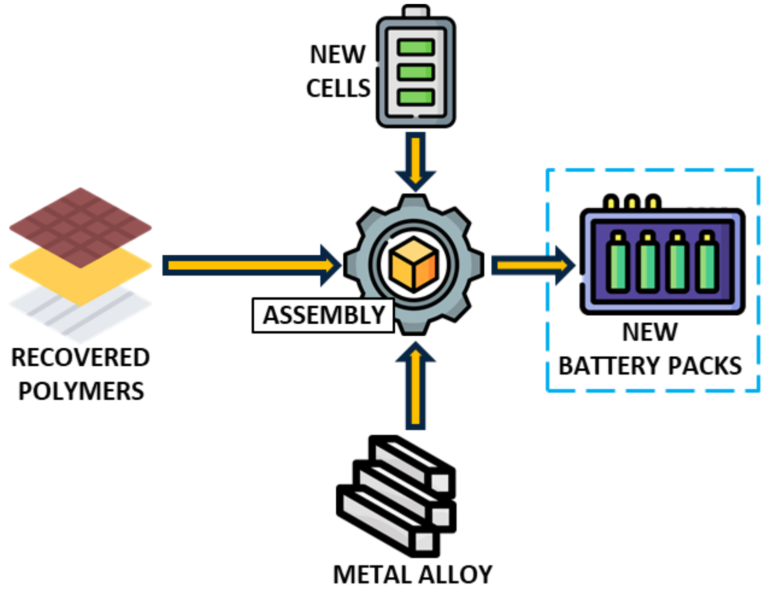
Material Re-use Technologies

New Battery Pack based on DfR



Design for recycling (DfR) guidelines will be developed based on: the compilation of lessons learnt from training workshops, EU legal requirements and recommendations, and results from LCA and SLCA. Its DfR will be used for 2nd life and also new battery packs.

Deliver **guidelines for FREE4LIB battery sustainable design** in accordance with eco-design principles, focusing on dismantling, recycling and re-assembly operations, aiming at reducing the environmental impact of the battery's life cycle, improving its recyclability, transport and usage phase, and optimising the EOL.

Deploy a new battery pack manufacturing procedure integrating FREE4LIB pouch cells (9c) and assembly with recovered materials (polymers and metals). **The aim is to assemble 3-5 battery modules and finally packs**.

Implement a **labelling/identification system** (scan QR labels) based on Battery Passport principles.

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TECHNOLOGY CENTRE CARTIF

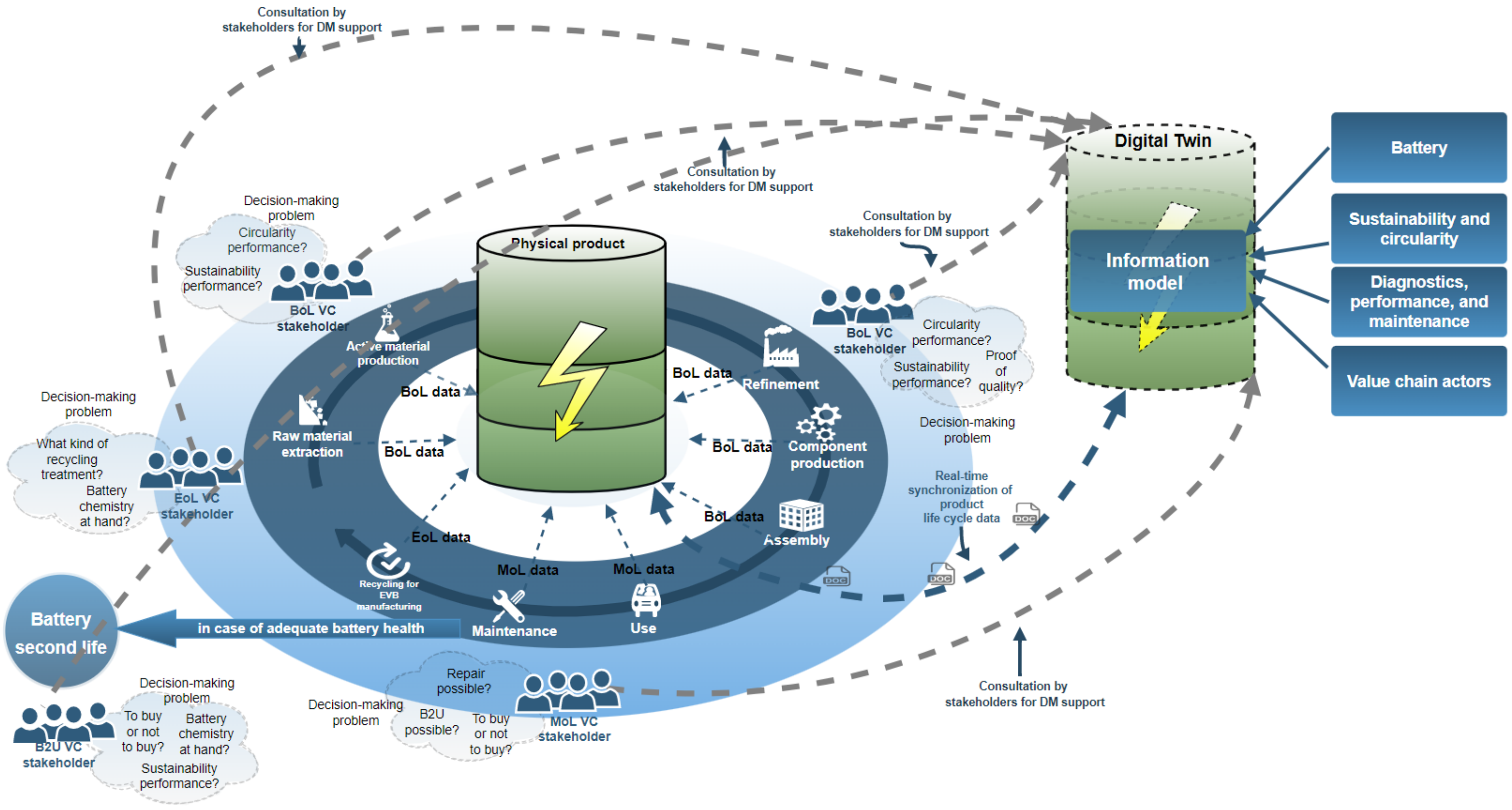
Battery Passports

Context and Objectives of the Battery Passports

- Context: transition towards sustainable circular electric vehicle battery value chains.
- Requirements: data to support value chain stakeholders in respective decision-making situations.
- Challenge: lack of data.
- Solution: Digital battery Passports as a data source.
- Objective: development of a digital Battery Passport concept for electric vehicle batteries.

The battery Passport platform

- Exploiting Data management and Blockchain technology
- Tracking LIBs between its BOL and EOL
- Explore the potential of a circular economy for traction batteries
- Platform will provide a digital identity (automatize processes, generate suggestions, report alerts, etc.)



Source: Berger, K., Schöggl, J.-P., Baumgartner, R.J., 2022. Digital battery passports to enable circular and sustainable value chains: Conceptualization and use cases. J. Clean. Prod. 353, 131492. https://doi.org/10.1016/j.jclepro.2022.131492

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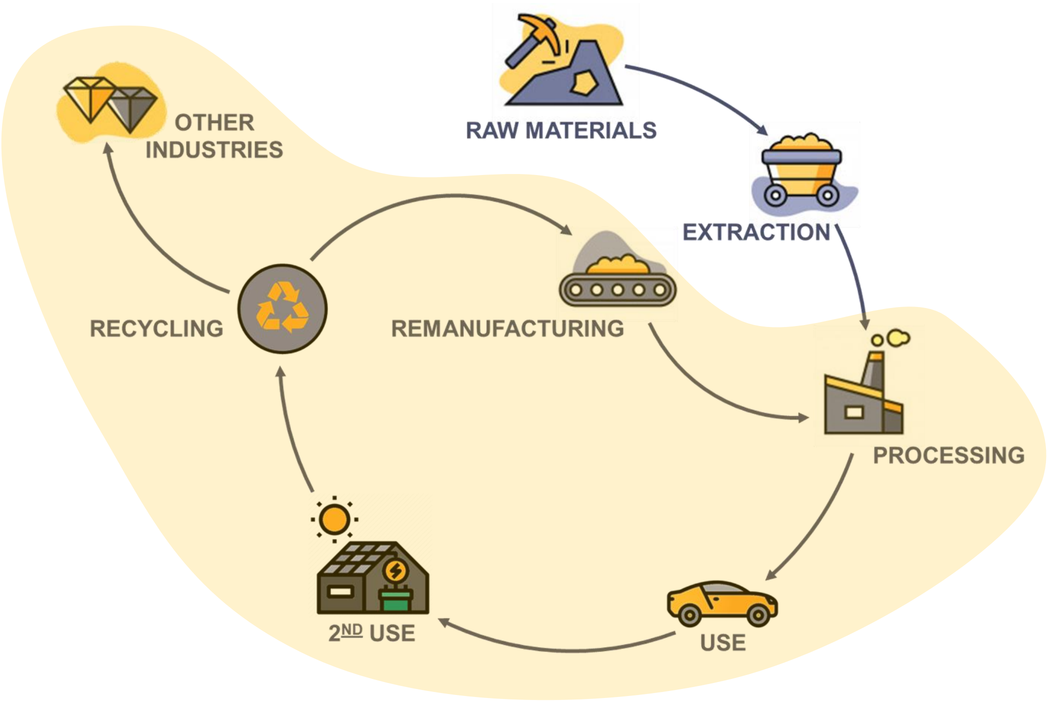
Impact Assessments



FREE4LIB's action zones in the LIB value chain

Environmental Life Cycle Assessment (LCA) for the main impact categories linked to the different recycling technologies. Thorough LCA thus identifying the main benefits/burdens to support the optimisation and further scalability of the technologies (as decision-making for the scale-up process).

Social Life Cycle and Systemic Sustainability Assessment (SLCA)

Techno-economic assessment (TEA) and Hazard and Operability (HAZOP) study of at least 8 of the scaled-up recycling technologies.

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C&D, Exploitation and Engagement Activities

Main goal

To lay down the basis for the **wide dissemination** of FREE4LIB's results, **market penetration** of the target KERs, and **general audience's sensitisation** of recycling solutions for Lithium-Ion Batteries.

Communication & Dissemination and Engagement actions

- **Strategic stakeholder mapping**
- Screening of **policy & regulatory framework** for specific actions
- Dedicated communication and actions for **consumer & citizens engagement**
- Coordination of **cooperation with European thematic networks, H2020 and Horizon Europe projects**
- Publication of **scientific articles** in journals, open science premises, and **presentations at conferences/congresses**
- **Awareness-raising & scientific popularization**
- **Knowledge information and exchange** with technologies communities and experts

Exploitation actions

- Identification of **new business models** (product selling and usage selling)
- **Implementation strategy** for each new business model (revenue streams, cost structure, etc.)
- **Exploitation audit** to characterise FREE4LIB exploitable results
- **IPR workshops**

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Thank you!

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