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Critical Raw Materials Recovery from EV Batteries

A Comprehensive Material Flow Analysis of Europe's Recycling Value Chain

MSc thesis

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A Comprehensive Material Flow Analysis of Europe's Recycling Value Chain

by

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Abstract

The energy transition relies heavily on lithium-ion batteries (LIBs), which are crucial for electrifying transportation and supporting renewable energy integration. However, the concentration of LIB production in China and other countries poses supply chain risks for the European Union. To mitigate these risks, the EU has introduced circularity targets as part of the 2023 Battery Directive. Despite the growing focus on these targets, significant knowledge gaps remain regarding their feasibility and the comprehensive tracking of other strategic materials. This study addresses these challenges and forecasts the circularity of critical raw materials in European EV batteries by 2040. Following a detailed review of LIB recycling processes and Europe's recycling capacities, the study performs a bottom-up assessment of material recovery efforts, evaluating the feasibility of meeting the EU's circularity targets.

Acknowledgments

I am profoundly grateful to my partner, Anneloes, for her endless encouragement, and understanding throughout this journey. Her patience, love, and unwavering belief in me have been invaluable, offering me the strength and financial support to return to university at a later stage in life. It is through her consistent support that I have been able to pursue my long-standing aspiration of embarking on an academic career. Without her, none of this would have been possible. I would also like to extend my gratitude to my family and Anneloes' for their equally strong support.

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Adrien Perello-Y-Bestard
The Hague, September 2024

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BMS Battery Management System.

CE Circular Economy.
CTR Carbothermic Reduction.

dMFA dynamic MFA.

EOL End-of-Life.
ESS Energy Storage Systems.
EU European Union.
EV Electric Vehicle.

LCO Lithium Cobalt Oxide.
LFP Lithium Iron Phosphate.
LIB Lithium-ion batteries.
LMO Lithium Manganese Oxide.

MFA Material Flow Analysis.

NCA Nickel Cobalt Aluminum.
NiCd Nickel Cadmium.
NiMH Nickel Metal Hybrid.
NMC Nickel Manganese Cobalt.

PbA Lead-acid.

SAR Salt-Assisted Roasting.

TC Transfer Coefficient.

1

Introduction

1.1. Background

1.1.1. The battery value chain

The energy transition is driving a significant shift in the materials we use, with key technologies such as Electric Vehicles (EVs), solar panels or wind turbines requiring far more metals than their fossil fuel counterparts [1–5]. Batteries, and Lithium-ion batteriess (LIBs) in particular, are central to this transition, playing a critical role in electrifying land transportation, which accounts for 15% of global greenhouse gas emissions [6]. They also help mitigate the intermittency of renewable energy sources and ensure grid stability. Batteries for EVs and Energy Storage Systems (ESS) are projected to directly contribute nearly 20% of the CO2 emissions reductions needed by 2030 to reach net zero emissions and indirectly to another 40% [5]. LIBs have increasingly replaced other types like Nickel Metal Hybrid (NiMH), Nickel Cadmium (NiCd), and Lead-acid (PbA) batteries. They are expected to dominate the global market for the coming decades despite the emergence of alternatives [1, 5, 7–11].

China currently leads the global LIB value chain with over 80% of worldwide battery cell production capacity [5, 12] and this influence is expected to continue in the coming years [13]. Beyond cell production, China also controls a significant portion of component manufacturing and raw material refining [5, 14–18]. However, the concentration of supply chain activities is not confined to China. For instance, 90% of planned nickel refining facilities are located in Indonesia, and 70% of global cobalt mining occurs in the Democratic Republic of Congo [14, 15, 19].

The European Union (EU) is particularly vulnerable to supply chain disruptions due to this heavy concentration [20] and many constituents of LIBs are classified as critical or strategic raw materials. This includes copper, lithium, aluminum, manganese, graphite, nickel, cobalt, and phosphorus [1, 21, 22]. Additionally, competition among China, the U.S., and the EU for control over the battery value chain has intensified. The U.S. Inflation Reduction Act (IRA), passed in 2022, heightened concerns within the EU about its ability to ensure its industrial competitiveness. In response, the European Commission has introduced several regulatory measures, including the Net-Zero Industry Act, the Critical Raw Materials Act, and the Battery Directive [23–26]. The EU has also ramped up funding and subsidies for battery production and research [27–31], and recently imposed tariffs on Chinese electric vehicle batteries [32, 33].

1.1.2. The role of Circular Economy

European battery cell production capacity reached about 225 GWh in 2023, producing around 100 GWh of the 210 GWh demand for EV and ESS [12]. Announced gigafactories should increase capacity to about 2,000 GWh by 2030 [12, 34]. This growth sets Europe on track to meet its exponential demand for LIB cells which is primarily driven by EVs adoption [2, 5, 7, 35–37]. By 2025, domestic production could meet up to 72% of this demand, with full self-sufficiency potentially achieved by 2026 [12]. However, Europe's midstream value chain remains underdeveloped. This stage, which includes the production of key components like cathodes, anodes, and their precursors — the intermediate compounds used to manufacture electrodes — lags in development [12, 38–40].

Circular Economy (CE) strategies, such as recycling, offer a potential solution for securing future supply of components and raw materials, but timing will be crucial to successfully align supply with growing demand. On one hand, the EU's Clean Vehicle Directive mandates that all new cars sold by 2035 must be zero-emission [41]. On the other hand, operationalizing and scaling recycling facilities takes significant time, and their profitability depends on the availability of sufficient End-of-Life (EOL) materials, which are currently limited [37].

In this context, the 2023 Battery Directive provides a strategic framework to position the EU as a leader in battery circularity [25]. The directive supports the development of recycling markets by setting ambitious targets to be gradually implemented starting in 2025 (figure 1.1).

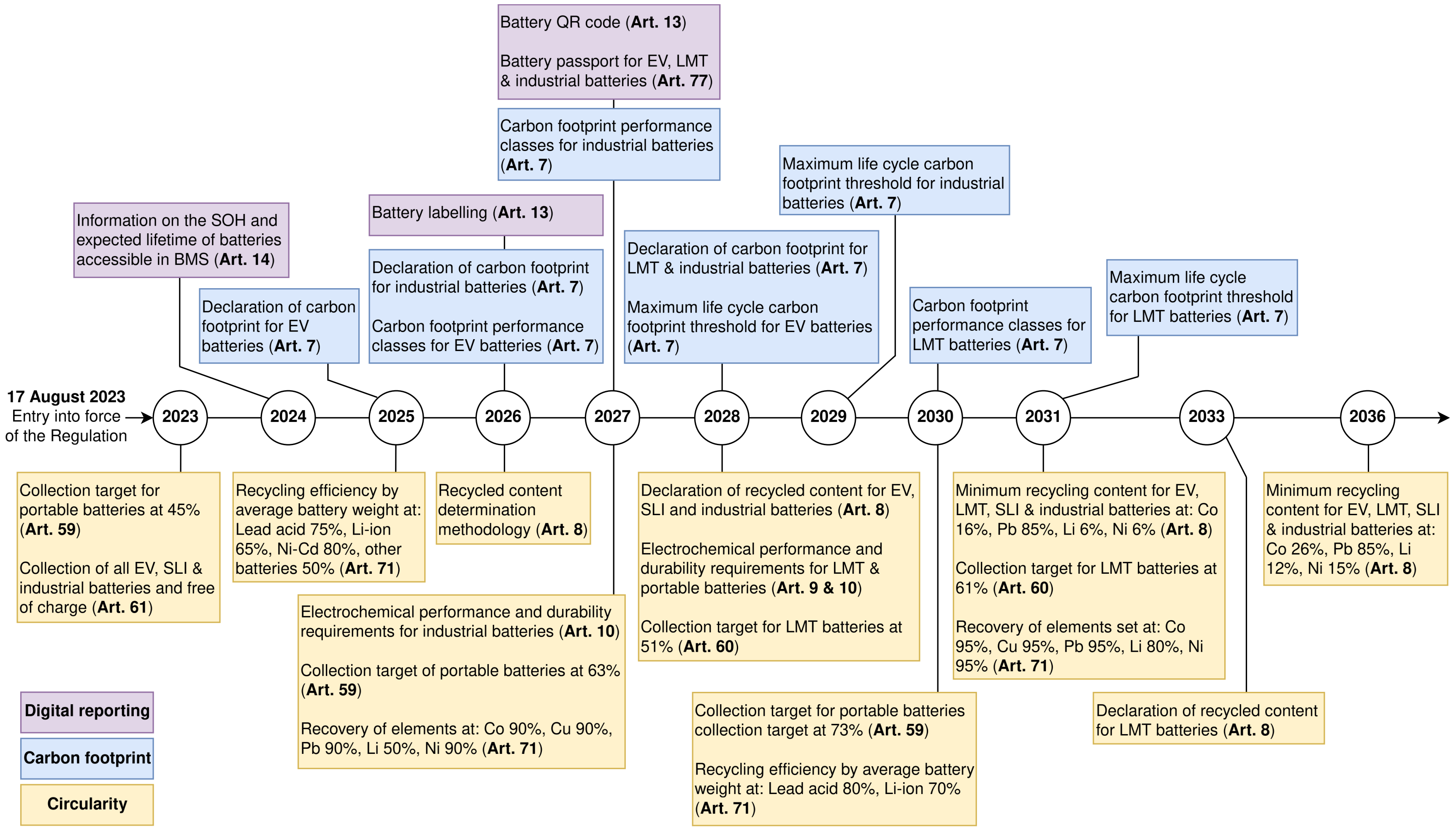


Figure 1.1. Main targets of the 2023 EU Battery directive.
EV = Electric Vehicle, LMT = Light Means of Transport, SLI = Starting, Lighting and Ignition, SOH = State of Health.

These include improving **recycling efficiency** (70% of the battery weight by 2030), increasing **material recovery** rates (95% for nickel, cobalt, and copper, and 80% for lithium to be recovered by 2031), and ensuring **recycled content** in new batteries (26% for cobalt, 15% for nickel, and 12% for lithium in new batteries by 2036). The regulation also aims to foster innovation and competitiveness by setting environmental and social standards through supply chain due diligence requirements and the implementation of a Digital Battery Passport.

1.2. Problem definition

1.2.1. Addressing CRM circularity

The strategic importance of LIB, coupled with the critical materials they contain, has drawn considerable interest from the scientific community. Many quantitative studies have been conducted to evaluate the potential for resource recovery from recycled batteries. These analyses cover a range of geographical regions, including global assessments [42–54], some with regional details [55–57], as well as focused studies on China [58–67], the U.S. [65, 68–71], and Europe [72–89].

Despite this extensive research, the feasibility of achieving the ambitious European circularity targets remains underexplored. Only three studies address or provide sufficient data to estimate the recycled content targets, and these assume that the directive's material recovery rates will be met [72, 74, 75]. Additionally, assessments from industry consultants, such as the Boston Consulting Group, lack transparency in their methods and assumptions [77]. The other directive targets are simply ignored. Furthermore, very few studies cover the whole range of strategic materials for Europe. Copper is often overlooked [55–57, 72, 77, 80, 83, 84, 86, 87, 89], similarly for graphite [55–57, 72, 75–78, 83, 84, 86, 87, 89], Manganese [55, 72, 75, 77, 78, 84, 86, 87, 89], and Aluminum [55–57, 72, 75, 77–80, 83, 84, 86, 87].

1.2.2. An incomplete picture of EU recycling

These gaps can partly be attributed to some studies focusing on specific European countries, which choose not to capture the transnational complexities of the LIB recycling supply chain [78, 79, 83, 85]. More concerning, however, is the oversimplification of the European recycling infrastructure itself. For example, only two studies differentiate between pyrometallurgy and hydrometallurgy recovery routes [73, 76], and they rely on outdated data that does not reflect the rapid advancements in the market [90, 91]. The criticism can be extended to many recent papers that track industrial recycling capacities [92–109]. Moreover, capacities are often compared without accounting for the type of feedstock being processed [110]. For example, the capacity of standalone plants that handle both mechanical pre-treatment and hydrometallurgy typically refers to the tonnage of spent batteries, whereas standalone hydrometallurgy plants process only crushed electrodes (i.e., black mass). Directly comparing these capacities tends to overestimate the hydrometallurgical facilities. While accurate and up-to-date data on recycling capacities and technologies exists, much of it is locked behind paywalls [111–113]. This limitation hampers efforts to fully assess the actions required to achieve circularity targets and blocks a comprehensive evaluation of the trade-offs between various recycling routes, particularly when considering profitability, environmental impacts, and circularity together. This challenge is even more critical given that an amendment mechanism is in place to adapt to ongoing developments in the battery industry.

1.2.3. Tracking multiple constituents

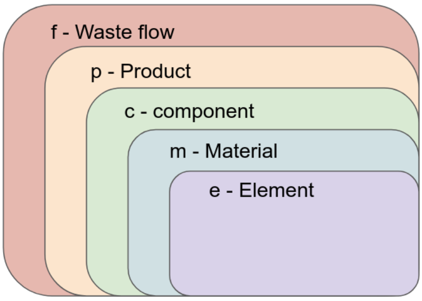


Figure 1.2. Hierarchical conceptualization of composition relationships.

The quantitative studies also reveal a broader issue: a narrow vision of circularity. CE aims to “keep products, components, and materials at their highest utility and value at all times” [114]. This implies the ability to assess the quantity and quality of various products, components, and materials and to understand how they mix along waste streams. In other words, circularity demands the tracking of constituents across multiple composition layers (figure 1.2) — a practice that is rarely implemented.

Resource management studies typically rely on Material Flow Analysis (MFA), a mathematical framework based on the mass balance principle [115, 116]. However, reviews of MFA in waste management reveal that analyses are typically conducted either at the chemical element level or the product level [117–121]. Even when both levels are considered, the focus tends to shift towards one or the other [122]. This in turn can limit the ability to capture the full complexity of waste streams. Many authors have pointed at data availability, reliability, or the effort required to collect it as a major hindrance in conducting MFA studies, especially concerning mass fractions and transfer coefficients [118, 122–124]. While big data techniques and the implementation of digital passports show potential, the lack of robust frameworks capable of simultaneously tracking multiple compositional levels continues to be a challenge. This shortfall hinders the transition from traditional waste management strategies to more systemic circular business models that integrate reverse supply chains.

One of the most widely used tools for steady state MFA is STAN (subSTance flow ANalysis) [125–127]. It allows for mathematical calculations, including data reconciliation, as well as result visualization through Sankey diagrams. However, it can not consider multiple data aspects, such as accounting for more than product-element relationships, simultaneously handling multiple chemical elements, or integrating environmental and economic data. For dynamic MFAs, most models rely on custom-made code. In 2020, Pauliuk and Heeren introduced ODYM (DYnamic Material systems) to specifically confront these limitations [128, 129]. ODYM is an open-source Python-based modeling framework that can handle large datasets, and multiple substances and jointly consider product, component, alloy, and chemical element levels. It is the only tool currently capable of addressing these specific requirements [130]. A review of the articles that reference the ODYM framework also reveals that it has yet to be applied to a system with this degree of granularity.

1.3. Research question

Based on the problem definition, the main research question is: **What is the projected circularity of critical raw materials in European EV batteries by 2040?** The study narrows its focus to EV batteries, as they represent the primary source of LIB demand. It includes the EU27 countries, along with the UK, Norway, and Switzerland given their significant recycling capacities and to avoid having to account for intra-European import-export dynamics.

To address this research question, the following sub-questions are formulated:

1. What quantity and types of end-of-life batteries are expected to be generated in Europe by 2040 due to the rise in EV adoption?
2. How is European LIB recycling capacity expected to evolve, and to what extent could it manage the projected battery waste?
3. What quantities of critical materials could be recovered from European end-of-life EV batteries?
4. Can the recycling targets outlined in the 2023 EU Battery Directive be realistically achieved?

Chapter 2 provides an in-depth overview of the design and recycling of LIBs, while chapter 3 builds on this by integrating a dynamic MFA to assess EOL LIBs, along with a novel modeling framework for tracking multiple composition layers. The results and their implications are discussed in chapter 4.

2

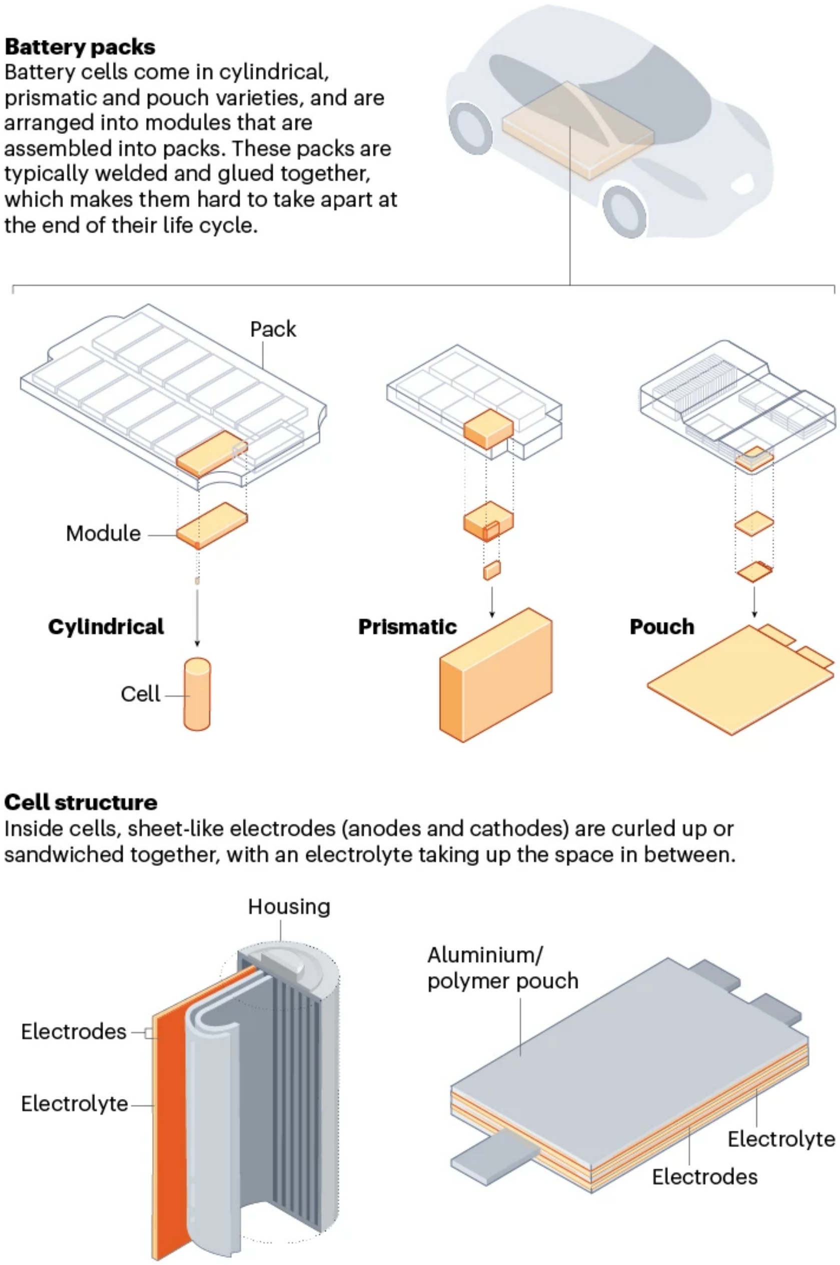
Overview of LIB recycling

2.1. Battery design

2.1.1. LIB structure

LIBs used in EVs are organized into a hierarchical structure comprising cells, modules, and packs (figure 2.1). The cell is the fundamental unit, containing the essential battery components. Multiple cells are grouped to form modules, which are then assembled into a battery pack [131, 132]. The pack is the final assembly that integrates with the EV and includes additional components such as a Battery Management System (BMS) to monitor and ensure the battery's performance and safety.

Within each cell, the **anode** and **cathode** serve as the positive and negative electrodes. The anode is typically composed of graphite, while the cathode is made of metal oxides like lithium nickel manganese cobalt oxide or lithium iron phosphate. These materials are known as **active materials** because they participate directly in the electrochemical reactions that enable the storage and release of electrical energy. During charging, lithium ions move from the cathode to the anode, and during discharging, the process reverses [7]. A porous polymer membrane known as the **separator** separates the anode and cathode. This separator, often made from polyethylene (PE) or polypropylene (PP), prevents short circuits by allowing lithium ions to pass through while physically keeping the electrodes apart [133]. The **electrolyte**, a conductive liquid or gel, facilitates the movement of lithium ions between the electrodes and is typically composed of lithium hexafluorophosphate (LiPF6) dissolved in organic carbonate solvents such as ethylene carbonate (EC) or dimethyl carbonate (DMC) [134, 135]. Additives in the electrolyte enhance its performance and longevity by improving conductivity and stability [136]. **Current collectors** conduct electrical current to and from the electrodes. They are usu-



Battery packs
Battery cells come in cylindrical, prismatic and pouch varieties, and are arranged into modules that are assembled into packs. These packs are typically welded and glued together, which makes them hard to take apart at the end of their life cycle.

Cell structure
Inside cells, sheet-like electrodes (anodes and cathodes) are curled up or sandwiched together, with an electrolyte taking up the space in between.

Figure 2.1. Typical structure of EV battery system. Source: [132].

ally made of aluminum for the cathode and copper for the anode [137]. Finally, **binders** help to hold the active materials of the electrodes together and adhere them to the current collectors [138]. Polyvinylidene fluoride (PVDF) is commonly used for the cathode, while styrene-butadiene rubber (SBR) and carboxymethyl cellulose (CMC) are employed for the anode [139, 140]. All these components are enclosed in a steel or aluminum casing which provides structural integrity and protection. Figure 2.2 illustrate the detailed structure as described above.

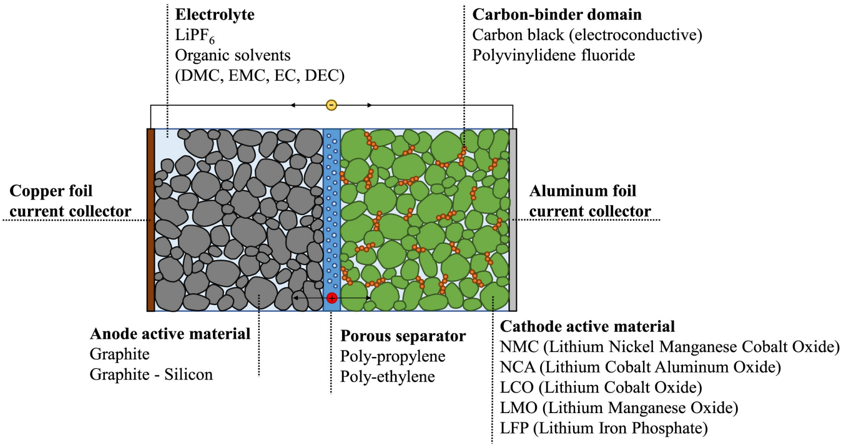


Figure 2.2. Schematic model of lithium-ion batteries. Source: [101].

2.1.2. LIB chemistries

LIBs are usually distinguished by their cathode chemistries. The most commonly used chemistries are LCO, LMO, LFP, NCA, and NMC [7, 37, 56, 141]:

Lithium Cobalt Oxide (LCO) batteries are a well-established technology, known for their specific energy typically ranging from 150 to 190 Wh/kg, making them a popular choice for portable electronic devices. However, their tendency to overheat and experience thermal runaway, combined with a relatively short life cycle of 500 to 1,000 full cycles, means they are rarely used in EV and ESS applications

Lithium Manganese Oxide (LMO) batteries offer a longer cycle life, ranging from 1,000 to 1,500 cycles, and significantly better thermal stability than LCOs batteries. Since LMOs do not require critical or costly metals beyond lithium, they are also more affordable. Their ability to deliver high power makes them suitable for applications that demand quick bursts of energy, though their specific energy is lower than other LIBs, typically between 100 and 150 Wh/kg. LMOs were initially used in the first mass-market EVs but are now primarily found in low-cost products like electric two-wheelers, power tools, and low-range EVs.

Nickel Cobalt Aluminum (NCA) batteries are known for their high specific energy, typically between 200 to 250 Wh/kg which makes them particularly suitable for EVs. However, they come with drawbacks, including higher production costs and challenges related to structural and thermal stability. These issues can lead to decreased performance and a shorter lifespan, typically averaging between 1,000 to 1,500 full cycles.

Nickel Manganese Cobalt (NMC) offer a good balance across key parameters such as specific energy (150 to 220 Wh/kg), performance, durability (1,500 to 2,000 cycles), and cost. This has made it the most popular choice in the automotive industry. However, NMCs are also are sensitive to overcharging and therefore require a BMS to prevent thermal runaway. Within the NMC category, several formulations reflect the current effort to minimize cobalt content in the cathode. This include: NMC111 (33% nickel, 33% manganese, and 33% cobalt), NMC523 (50%, 20%, and 30% respectively), NMC622 (60%, 20%, 20%), NMC811 (80%, 10%, 10%) and NMC955 (90%, 5%, 5%)

Lithium Iron Phosphate (LFP) cells are notable for their excellent thermal stability, even at high temperatures, which enables their direct integration into battery packs. They also demonstrate high durability, with a lifespan of up to 2,000 full cycles while retaining their performance. LFPs are relatively cost-effective due to the absence of nickel and cobalt. However, they have relatively low specific energy, ranging from 90 to 160 Wh/kg, which can restrict their application in EVs compared to NMC or NCA.

2.2. Recycling processes

The recycling route for LIBs begins with the collection of end-of-life batteries. Spent batteries can be repurposed or recycled for material recovery. The recycling process involves a series of steps that typically include pre-treatment, pyrometallurgy, and/or hydrometallurgy, though there are many possible variations [110, 142, 143]. The sequence and depth of these processes can differ significantly, depending on the specific approach used at the recycling facility. A description of these processes is provided in this section.

2.2.1. Sorting and echelon utilization

Sorting may be necessary to separate batteries by chemistry, shape, or size, depending on the recovery process used at the recycling plant. At this stage, it is also possible to diagnose the State-of-Health (SoH) of the batteries for re-use in other applications [100, 140]. If material recovery is prioritized over reuse, the batteries may first be deactivated.

Echelon utilization refers to the practice of re-purposing spent EV batteries for secondary applications [144–147]. When the battery falls below 70-80% of its initial capacity, it is no longer safe and reliable enough for EV traction. However, they do retain significant economic value and can therefore be used for other applications that have lower safety requirements than EVs, leading to the extension of the overall battery lifetime. One of the most common examples of secondary use is stationary Energy Storage Systems (ESS) for grid stabilization and peak shaving.

2.2.2. Deactivation and dismantling

Deactivation

Deactivation and discharging are terms used interchangeably in the literature but they refer to distinct processes. Discharging involves draining the battery's stored energy, while deactivation (or stabilization) includes a broader set of safety measures designed to neutralize potential hazards due to chemical reactions or thermal runaway. Without precautions, mechanical treatment could lead to the release of hydrogen fluoride, a highly toxic substance [148–151], or cause short circuits between the anode and cathode which can produce heat and ignite the flammable electrolyte solvent [100, 134, 149–152].

Common methods for **discharging** batteries include immersion in conductive salt solutions or electrical short-circuiting [100, 140, 150, 152–154]. The aqueous solution prevents lithium from reacting with the electrolyte, while short-circuit releases the remaining charge as heat (energy recovery is technically feasible but impractical and uneconomical at large-scale) [149, 150, 155]. Alternative **deactivation** techniques can be employed either in conjunction with or instead of discharging. Deactivation typically includes physical or chemical alterations of the electrolyte via thermal treatment. This is usually done either by pyrolysis (at 450°C - 600°C, in the absence of oxygen) or by cryogenic freezing (-175°C to -200°C, using liquid nitrogen) [100, 140, 149, 150]. Pyrolysis, in particular, can also be applied after mechanical treatment to enhance material recovery and is discussed in more detail further.

Many recyclers opt against discharging or deactivation due to the high capital and equipment costs, as well as the technical complexities of scaling the process. Instead, they proceed to mechanical treatment in an inert atmosphere, such as nitrogen, argon, or carbon dioxide to reduce handling risks. This limits exothermic reactions from lithium-air interactions and prevents the release of toxic gases [100, 140, 152–154].

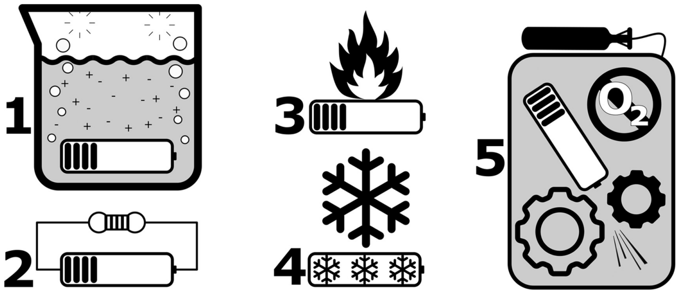


Figure 2.3. Deactivation techniques: 1. Salt solution, 2. Electrical short-circuiting, 3. Thermal treatment, 4. Cryogenic treatment, 5. Inert atmosphere. Source [156]

Dismantling

Dismantling entails the physical teardown of battery components and can be carried out at the pack, module, or cell level. This process benefits mechanical treatment by allowing for simpler separation and yields purer products, but it first requires deactivation to ensure worker safety. Dismantling is difficult to automatize because of safety requirements and the lack of standardized design. The heavyweight of EV batteries, together with the variety of shapes and sizes, make it challenging to develop a universal automated system [139, 150, 157, 158]. The introduction of the battery passport under the EU Battery Directive could help address some of these challenges. Meanwhile, facilities either rely on manual disassembly, which is time-consuming and exposes workers to health and safety hazards, or bypass this stage altogether [100, 148, 149, 159, 160].

2.2.3. Mechanical pre-treatment

The objective of mechanical pre-treatment is to produce black mass, a concentrated mix of battery electrodes. This process involves separating spent batteries into several distinct fractions: the black mass itself, metallic fractions (ferrous containing iron from the casing, and non-ferrous aluminum and copper from both the casing and the collectors), and a plastic fraction. Pre-treatment is specifically designed to maximize the recovery of black mass, as it contains valuable raw materials such as lithium, cobalt, and nickel. The key steps are comminution and separation, with optional post-processing steps performed either in-between or after.

Comminution

Comminution involves the crushing and grinding of batteries to liberate the electrode materials [149, 153]. This process can be performed either as dry or wet crushing. In dry crushing, no liquid is used, while wet crushing involves the deactivation solution mentioned earlier [100, 150]. For additional safety, dry crushing can be conducted under inert and/or cryogenic conditions. Notably, cryogenic conditions facilitate the electrode separation and improve the yield of the black mass [150]. Various crushing techniques exist, ranging from low-speed rotary shears to high-speed impact crushing or water jets [155]. Wet crushing has several advantages: it suppresses dust generation and acts as a fire retardant by absorbing heat generated during crushing. However, wet crushing also has drawbacks. It requires chemical reagents and generates wastewater, both of which add costs and have environmental impacts. The influence of these methods on the efficiency of subsequent processing stages is debated. Some argue that dry crushing leads to higher surface oxidation, which hampers the efficiency of separating the metallic fraction [100, 150]. Others contend that wet crushing causes electrode materials to pass through screens too quickly before fully separating from their collector foils, complicating further purification [140, 160].

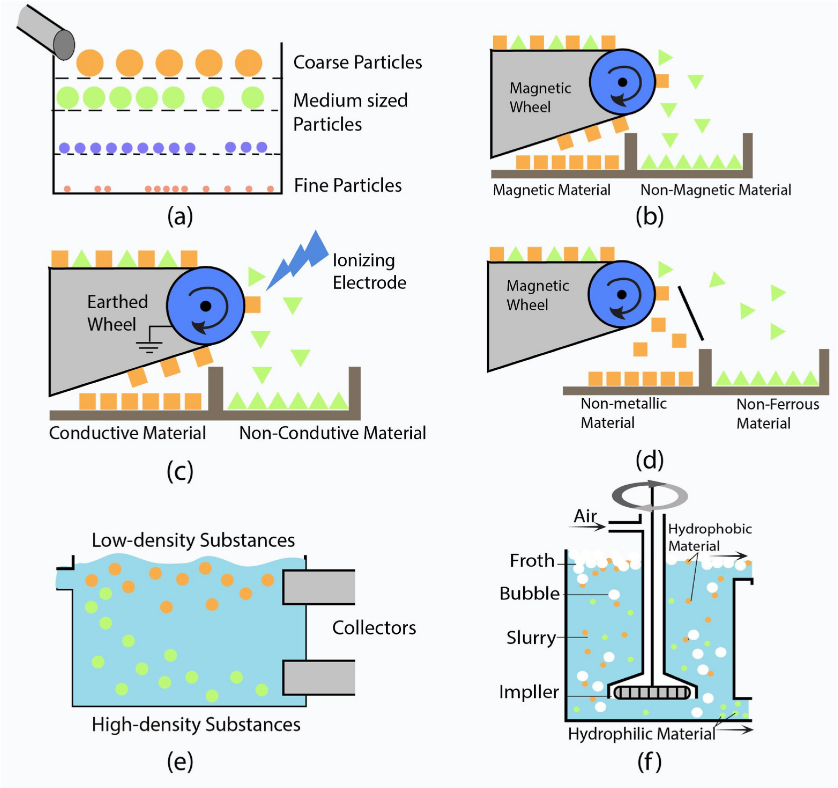


Figure 2.4. Separation techniques: a. Sieving, b. Magnetic separation, c. Electrostatic separation, d. Eddy current separation, e. Gravity / density separation, f. Froth flotation. Source [100]

Separation

Following size reduction, materials are separated based on their physical properties. The process often begins with **sieving**, which classifies particles by size into three main fractions [100, 140, 150–152, 154]: i) a coarse fraction that primarily contains metals (iron and aluminum) and polymers from the casing, as iron tends to resist shredding and plastics can form lumps at room temperature; ii) a fine fraction (the black mass), which contains electrode materials such as lithium, cobalt, manganese, nickel, and graphite; and iii) a middle fraction, consisting of current collectors like aluminum and copper foils, along with separators, and some active materials. **Magnetic separation** is then used to further sort out ferromagnetic and paramagnetic materials (i.e., steel and aluminum) from the coarse materials. [100, 140, 155]. Electrical properties are also utilized through techniques such as **electrostatic separation** or **eddy current separation** [100, 140, 150, 152, 155]. Electrostatic separation uses an electric field to distinguish between conductive materials (i.e., aluminum, copper) and non-conductive ones. Conductive materials are attracted or repelled based on their ability to hold a charge. It works best for fine particles and when the materials are dry. Eddy current separation targets non-ferrous metals (also aluminum and copper in this case) by applying rapidly changing magnetic fields. These fields create eddy currents in the metals, which generate opposing magnetic forces that push the metals away from the rest. This method is more effective for sorting midsize to larger particles. **Gravity**, and **air (pneumatic) separation** employs water or air tables to separate materials based on their density or aerodynamic properties [100, 140, 152, 153, 155]. They typically distinguish lighter organic materials from heavier ones, such as separating the plastic separator from metal electrodes and copper current collectors.

Advanced treatments

Comminution and separation alone are insufficient to fully detach the cathode materials from the aluminum collectors [150, 152, 160, 161]. The strong adhesive properties of PVDF cause some active materials to remain attached to the collectors or clumped together. This persistent binding complicates the extraction in subsequent hydrometallurgical processes and increases the consumption of chemical reagents [150, 160, 162]. To address this challenge, two broad categories of techniques can be employed: thermal and chemical treatments (see figure 2.5) [139, 140, 149–153, 162, 163]. In contrast, the anode collector can be more easily removed from the anode active material because the bond between graphite and copper foil is relatively weak [140, 150, 162]. Graphite usually remains mixed with the cathode active material in the black mass. Methods such as froth flotation can be used to separate it - ideally after PVDF removal - but are still confined to the lab scale [140, 151, 152, 155].

Thermal treatment involves various techniques that differ in temperature, atmospheric conditions, and oxidation processes. It is typically performed at temperatures below 600°C to avoid significant degradation of graphite and damage to the aluminum foils [140, 150, 154, 160]. For deactivation purposes, lower temperatures are sufficient, as electrolytes evaporate be-

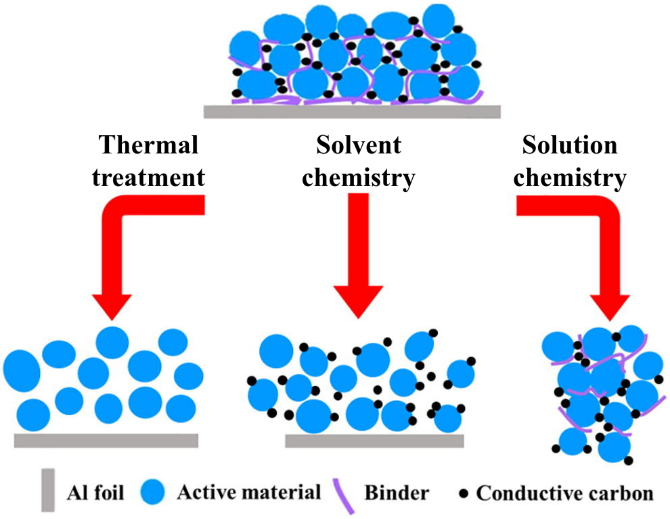


Figure 2.5. Advanced mechanical pre-treatments. Adapted from [163]

low 200°C [152]. For post-mechanical treatment, higher temperatures are needed to decompose the organic binder and weaken the adhesion between the cathode active material and the collector [140, 150, 152, 153, 160]. This is because PVDF only begins to decompose at 350°C. The presence of oxygen (roasting or calcination) accelerates PVDF breakdown and lowers the required temperature but also causes graphite oxidation and increased carbon emissions. In contrast, pyrolysis requires higher temperatures (500–600°C) to decompose PVDF, but better preserves the graphite structure [150–152, 160]. All thermal treatments release fluorinated gases which besides atmospheric pollution also lead to equipment corrosion, and higher temperatures increase these emissions [150, 160, 162]. Techniques such as vacuum pyrolysis or the use of additives like calcium oxide or molten salts can help mitigate these environmental impacts by neutralizing fluorinated emission or lowering temperature requirements [92, 139, 150, 154, 162].

Chemical methods provide alternatives to thermal treatment by selectively targeting the cathode active material, the aluminum collector or the PVDF [139, 150, 151, 153, 160, 162]. **Acid solutions** dissolve the active material and extract the valuable metals while **alkali dissolution** specifically targets the aluminum foil, dissolving it without affecting the cathode material which remains intact for further processing. In contrast, **solvent**

chemistry dissolves the PVDF itself. Solution chemistry (acid and alkali) is less commonly used because it can create impurities that complicate metal recovery and generate wastewater. Solvents have the advantage of preventing the release of fluorinated compounds and can be recovered through distillation. However, their high viscosity makes separating dissolved materials challenging. They are also expensive and toxic, requiring special safety measures.

2.2.4. Pyrometallurgy

Smelting

Pyrometallurgy can be broadly categorized into high-temperature smelting, carbothermic reduction, and roasting. **Smelting** recovers valuable metals by heating the battery materials to temperatures above their melting point to facilitate the separation of the metals in the liquid phase (typically between 1250°C and 1500°C) [92, 102, 103, 151, 159]. The process unfolds in two main stages: the thermal deactivation phase to evaporate electrolytes and prevent explosions due to pressure buildup, followed by a high-temperature phase where the materials melt and separate into distinct phases. During smelting, organic material (electrolytes, plastics, graphite, etc.) and aluminum serve as reducing agents, aiding in the conversion of metal oxides to pure metals and providing additional fuel to reduce the overall energy requirements. The process produces two main outputs: a metallic alloy composed of heavier metals, such as cobalt and nickel, which settle at the bottom, and slag, which contains lighter elements like lithium and manganese. Additionally, flue gases are released during the process [103, 164].

Carbothermic reduction and roasting

Carbothermic Reduction (CTR) and roasting, on the other hand, are more energy-efficient processes. These methods are exothermic, generating their own heat during the reaction, whereas smelting relies on an external heat source. This enables them to function at lower temperatures, generally ranging from 600°C to 1000°C [92, 103, 159, 165, 166]. They are typically applied after mechanical pre-treatment, where the active cathode material is heated in the presence of an external reducing agent (e.g., coke, charcoal) to reduce metal oxides to their metallic states.

Outputs and recent developments

In all cases, hydrometallurgy is required to refine the alloy output and obtain pure metals [73, 151, 159, 164]. However, the number of substances reclaimed is limited. Indeed, recovering lithium is challenging because it ends up in the slag and although it can technically be extracted using hydrometallurgy, this approach is not economically attractive [101, 103, 164, 167]. As a result, slag is currently often sold as a raw material for the construction industry. The difficulty is exacerbated by the shift in battery manufacturing towards chemistries with reduced cobalt content. These shifts reduce the economic viability of pyrometallurgy for battery recycling [101, 103, 167]. Recent advancements have introduced **Salt-Assisted Roasting (SAR)** to address these challenges [92, 159, 166, 168–170]. In this process, salts are mixed with the battery materials to transform lithium compounds into more volatile and water-soluble forms. These lithium compounds are carried away with the flue gases and, as the gases cool, they condense and are collected in the flue dust.

2.2.5. Hydrometallurgy

Hydrometallurgy is gaining momentum. It efficiently recovers high-purity, battery-grade metal salts and can process most constituents of the black mass [171]. It is also an essential step following pyrometallurgy. The approach operates at relatively low temperatures and involves aqueous chemistry, beginning with leaching to dissolve metals into solution. These dissolved metals are then selectively separated, concentrated, and purified, ultimately converting them into battery precursors.

Leaching

Leaching is a process that uses chemicals, known as leaching reagents, to dissolve solid metals and convert them into a liquid form called leachate, where the metals exist as ions. This conversion is necessary for recovery methods like solvent extraction or chemical precipitation to work effectively. Leaching methods can be divided into organic and inorganic acid leaching, alkali leaching, and bioleaching. Acid leaching involves using hydrogen ions in a low-pH solution to interact with metals, while alkali leaching utilizes hydroxide ions (OH^-) in a high-pH solution. Bioleaching makes use of bacteria, fungi, or archaea to dissolve metal ions but is still far from industrial applications and is therefore not described in more detail [163, 165–167, 172–174].

Inorganic acids are widely used in industrial applications due to their rapid reaction rates, high leaching efficiency, and low costs. They can dissolve a wide range of metals efficiently [163, 166, 172, 175]. However, this often results in lower selectivity, as they can dissolve multiple metal ions simultaneously, making subsequent separation more complex [172, 176, 177]. Additionally, they present notable challenges, including the emission of toxic and corrosive gases such as sulfur oxides, nitrogen oxides, and chlorine gas. The process also generates acidic effluent, which requires resource-intensive treatment to neutralize and remove contaminants [163–165, 167, 173–175].

On the other hand, **organic acids** are biodegradable, generally easier to recycle, and produce less toxic and corrosive waste [164–166, 172]. Although many organic acids are weaker than inorganic acids, some achieve leaching efficiencies comparable to those of inorganic acids thanks to their chelating properties. This property allows organic acids to form multiple bonds with metal ions, enhancing their selectivity. It also enables them to function as both leaching agents and precipitants, simplifying the process of leaching and metal ion separation into a single step [163, 165, 166, 175]. However, they tend to be slower and more costly, which poses challenges for their use at an industrial scale [166, 167, 172, 173]. Additionally, their environmental benefits are questioned when evaluated from a life-cycle perspective [175, 178].

Alkali leaching is also gaining attention thanks to its selective properties which reduce the need for costly and complex separation steps [166, 167, 176, 178]. However, it is generally less efficient than acid leaching, requiring longer leaching times and higher concentrations to achieve effective results [175]. Sodium hydroxide (NaOH) is specifically employed to selectively remove aluminum, while ammonia-based reagents are used to leach cobalt, nickel, and copper [163, 166, 172, 176].

Separation

Separation of the different metal ions is essential after the leaching step. The two primary methods used for this purpose are solvent extraction and selective precipitation, which can be used either independently or in combination [164, 172, 174, 179]. Due to the complex composition of the leachate, multiple steps are often necessary to ensure effective separation and recovery. Novel approaches like co-precipitation and sol-gel methods have emerged to tackle these challenges.

Chemical precipitation separates the metals by converting them into an insoluble solid called a precipitate. It is done by adding a reagent, known as a precipitant, to the leachate. The precipitant reacts with the target metal ions, causing them to leave the solution phase and form a solid phase [165, 172, 173, 178]. This method can be used to separate metals or to remove impurities before the solvent extraction step. pH and temperature are adjusted to control the process and ensure optimal precipitation [164, 167, 180]. Achieving selective precipitation of individual metal ions can be challenging, often requiring a step-by-step approach [105, 174, 178]. Copper, Aluminum, and Iron are precipitated first one after the other at low pH values (<5). Nickel, Cobalt, and to a lesser extent, Manganese, are co-precipitated at moderate pH levels (~10) due to their similar chemical properties. Lithium is usually recovered last at higher pH values (~12) [165, 167, 173].

Solvent extraction, also known as liquid-liquid extraction, is a process that separates metal ions by transferring them between two immiscible liquid phases: the aqueous phase (leachate) and an organic phase. In this process, the aqueous solution is mixed with a solvent, known as the extractant, which has a high affinity for the target metal ions. The extractant forms a specific chemical complex with the metal ions, which is more soluble in the organic phase than in the aqueous phase. As a result, the metal ions migrate from the aqueous layer into the organic layer. The target metal ions are then typically recovered by precipitation or by electrolysis [163, 167, 173, 174]. Solvent extraction is well established due to its high recovery efficiency and ability to achieve high purity yields. The process is characterized by a short reaction time, low energy consumption, and can be operated continuously, but the operation remains complex and the solvent expensive [105, 165, 167, 173, 175]. Solvent extraction is commonly used to sequentially recover cobalt, followed by nickel, and lithium last, similar to the order of recovery in chemical precipitation [95, 163, 169, 175, 178, 181, 182].

Recent developments

Sol-gel and **co-precipitation** methods differ from traditional approaches by directly synthesizing cathode active materials or their precursors from the leachate. This one-step recovery (also called regeneration) eliminates the need for intermediate separation and extraction of individual metal ions, potentially reducing both costs and environmental impacts [163, 166, 174, 178]. As the name suggests, co-precipitation involves the simultaneous precipitation of multiple metal ions from the leachate, typically Nickel, Manganese, and Cobalt. The resulting co-precipitate is then mixed with a Lithium source (e.g., Li_2CO_3) and subjected to calcination to synthesize new active materials [163, 165, 173, 175]. The sol-gel method converts the leachate into a gel through hydrolysis, after adjusting the stoichiometry by adding specific metal salts and ammonium hydroxide. This gel is then heated to remove organic materials and solidify it into the desired cathode material [165, 166, 175, 178].

2.2.6. Direct recycling

Hydrometallurgy and pyrometallurgy decompose the cathode active materials into metal salts or high-value precursors, which are then used to remanufacture new cathodes. **Direct recycling** shortens the recovery route even further than the sol-gel and co-precipitation techniques described above. It focuses on directly restoring the crystal structure and electrochemical performance of the active materials [139, 163, 166, 175, 179]. It addresses defects such as lithium loss due to side reactions, the dissolution of metals (Ni, Mn, Co) into the electrolyte or their migration to the anode, and structural degradation like cracking that compromises the cathode's integrity [183–186].

Various methods can used to repair these defects but they all essentially involve introducing an excess lithium source and using physical or chemical techniques to reintegrate lithium ions into the cathode's crystal structure [176, 184–188]. For instance, chemical relithiation uses lithium compounds in a low-temperature solution, whereas hydrothermal relithiation involves high-pressure and high-temperature aqueous solutions to replenish lithium content. Solid-state sintering regenerates the cathode material by applying lithium salts and high-temperature heat to facilitate lithium reinsertion. Molten salt relithiation, instead, employs eutectic salt mixtures which have lower melting points than individual lithium salts. Lastly, electrochemical relithiation uses an electric field to drive lithium ions into the cathode material. Each of these methods is typically followed by heat treatment (annealing) to fully restore the crystal structure

Direct recycling presents several compelling advantages over pyro- and hydrometallurgy. It is simpler, more energy-efficient, and significantly reduces pollution. It is especially beneficial for low-value cathodes (e.g., LFP) which are less economically viable with conventional methods [163, 173, 189]. One of the most exciting advancements in this field is the possibility of **upcycling** NMC cathodes. This approach not only aims to regenerate the active material but also modifies its composition, shifting from low-nickel (e.g. NMC111, NMC532) to nickel-rich stoichiometries (e.g. NMC622, NMC811) [176, 179, 183, 185]. This could address the challenge of outdated chemistries, which can limit the effectiveness of direct recycling. The process typically involves adding and mixing additional nickel salts during the thermal stage.

Direct recycling is still in the research phase and has yet to demonstrate economic viability at an industrial scale. This viability is particularly sensitive to the fluctuating cost of lithium sources needed for relithiation processes [189]. Processing spent cathode materials with varying levels of lithium deficiency, coupled with the diversity in cell designs and chemistries, also remains challenging [166, 175, 185]. Additionally, mechanical pre-treatment methods introduce impurities, such as aluminum, copper, and iron, which can degrade the crystal structure (although, in small amounts, these impurities could also enhance electrochemical properties) [163, 166, 179, 186]. This further complicates scaling efforts while ensuring consistent, high-purity products. Consequently, manual disassembly is often preferred over mechanical pre-treatment before direct recycling or upcycling [175, 179, 184, 186]. Interestingly though, when accounting for the full costs, manual disassembly may be economically preferable over mechanical pre-treatment case [148].

2.3. European recycling supply chain

Building on the understanding of LIB recycling processes, a comprehensive mapping of existing and announced LIB recycling plants in Europe was conducted for the period 2010–2030 figures B.2 to B.4. The review

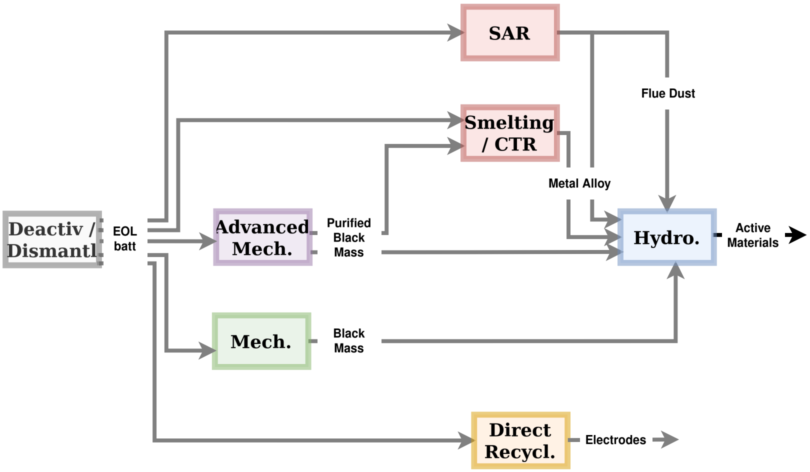


Figure 2.6. Overview of main processes in the European LIB recycling supply chain. For clarity, only flows between processes are represented on the diagram.

identified 100 plants operated by 63 companies, reflecting the most current market situation [90, 91]. The dataset was made available on Zenodo [190] and follows the Swave dataset format [109]. It covers information about each facility, including the company and other stakeholders involved, the location (both country and city), the status (operational, planning, stopped), the implemented processes, feedstock, output products, and the annual capacities. Additionally, it describes, to the best of the author's knowledge, which elements and components are being recovered. Figure 2.6 provides overview of the recycling processes included in the rest of the analysis.

3

Methodology

This work integrates a dynamic MFA (dMFA) with the novel concept of a multilayer MFA, where the dMFA serves as the input for the latter (figure 3.1). The dMFA is a widely used approach for estimating EOL flows available for recycling [52, 55, 56, 79, 122]. In this study, the dMFA operates on two levels: the EV level and the battery level. At the EV level, a flow-based MFA estimates the stock of EVs based on sales forecasts and vehicle lifespans. At the battery level, the sales and EOL outflows of batteries are determined using a stock-driven model combined with battery lifetimes. Battery flows are initially expressed in energy units before being converted to mass units according to the energy density of each LIB chemistry. The resulting data, together with detailed composition information, are then utilized in a multilayer MFA. The multilayer MFA extends the concept of Transfer Coefficient (TC) to systems with hierarchical compositional layers. The conceptual framework for this approach is first developed, followed by its application to the recycling routes discussed in the previous chapter.

3.1. Battery stock and flow modeling

3.1.1. EV sales

To project future EV sales across Europe, including the EU27, Norway, Switzerland, and the UK, we employ regressions based on the Bass diffusion model, a well-established method for describing the adoption and diffusion of new technologies [7, 57, 72]. The Bass diffusion models are applied to historical data and forecasts from IEA, Statista, RMIS, and Forbes which extend through 2035 (figure A.5 in appendix) [191–194]. To address the uncertainty beyond 2035, additional sales forecasts from the scientific literature are incorporated to cover the period from 2035

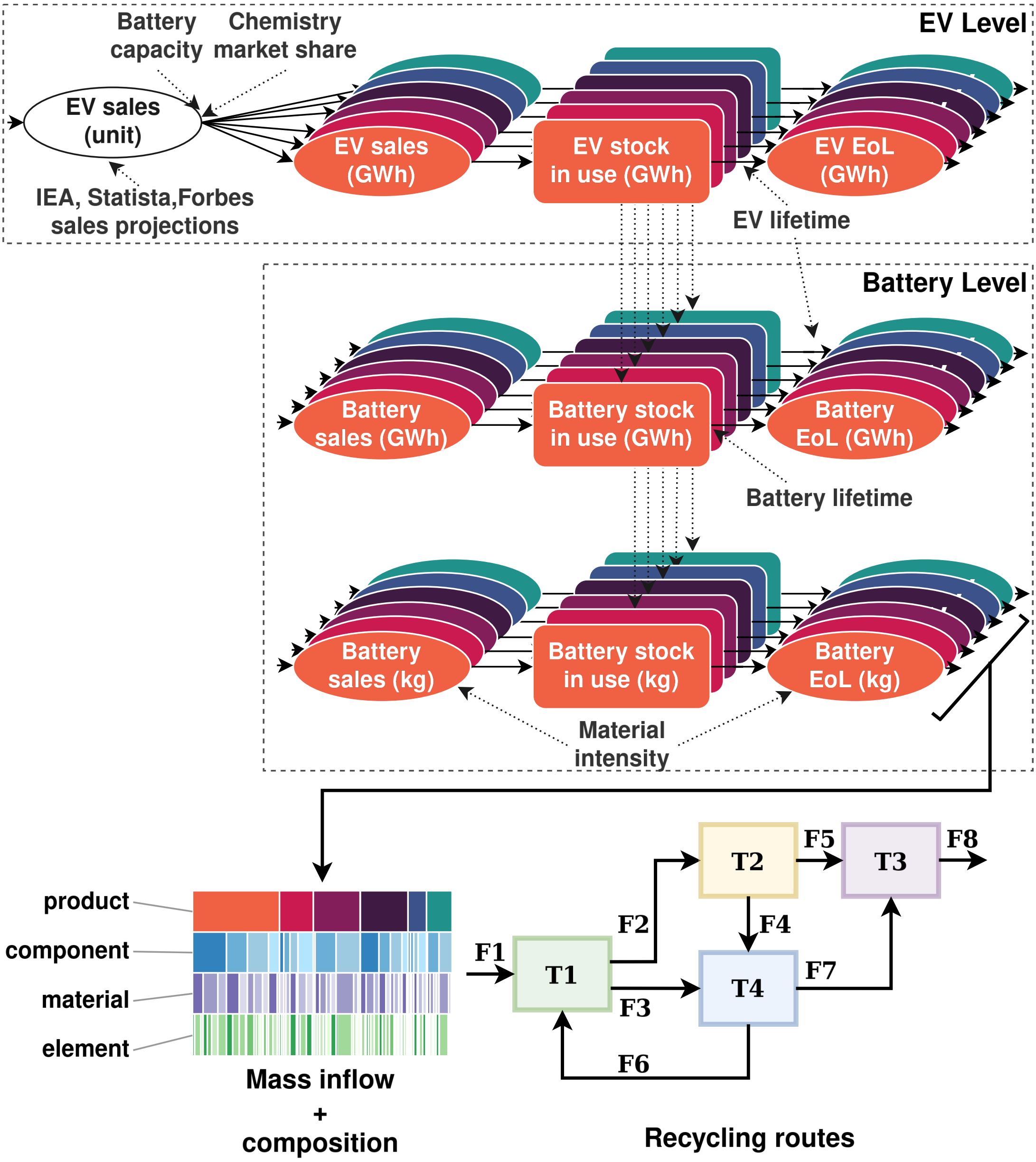


Figure 3.1. Conceptual outline of the dynamic MFA methodology used. Dashed lines represent influences between variables.

to 2040 [1, 72, 195]. Each sales regression is then scaled using a range of average battery capacities. These capacities are determined by combining projections of BEV and PHEV market shares with the battery sizes of various vehicle segments (small, medium, large, utility vehicles) and their anticipated market distributions (figures A.1 to A.3 in appendix). From these projections, low and high-demand scenarios are generated using the lower and upper bounds of all regressions.

3.1.2. Chemistry market shares

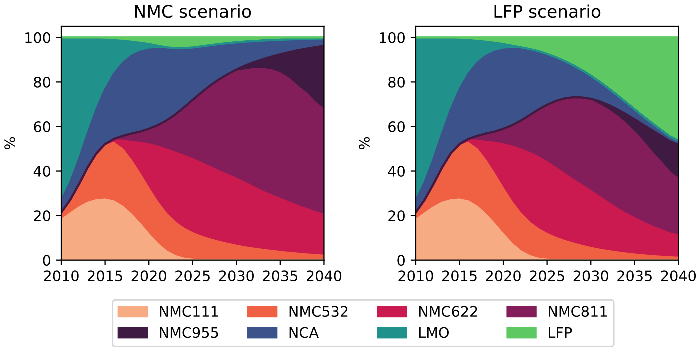


Figure 3.2. LIB chemistry market shares. The scenarios are adapted from [52] to reflect the European market.

EV sales are further segmented by battery chemistry according to market share forecasts (figure 3.2). The forecasts in this study are based on scenarios presented by Xu et al. [52], which are widely used in quantitative modeling [47, 55, 72, 196]. However, these world averages do not accurately represent the European market. LFPs are predominant in China, accounting for over 67% of EV battery capacity sales in 2023. In contrast, their adoption in Europe remains limited at 7% [5, 191]. To address this discrepancy, the scenarios are adapted to better reflect the European context, drawing on [48, 56, 57, 75, 78, 82, 87, 88, 197, 198].

While some studies consider the potential impact of emerging technologies, such as sodium-ion batteries, solid-state batteries, lithium-sulfur, or lithium-air batteries [37, 42, 47, 52, 53, 56, 87], many others focus exclusively on currently dominant battery types [45, 50, 57, 66, 72, 75, 78, 84, 88], and this work follows that approach. This simplifies the analysis of other parameters within the recycling supply chain. The choice is also supported by Wolf and Lüken's 2024 review of market projections [11]. Their review suggests that in the mid-term, sodium-ion batteries, with their relatively low energy density, will mainly be used in stationary applications or heavy-duty transport where their long cycle life is advantageous. Additionally, other innovative lithium chemistries are not expected to reach technological maturity until after 2030, a timeline that is often reflected in the scenarios from the studies referenced above.

3.1.3. Lifetimes and collection

Most studies adopt a conservative estimate for battery lifetime, typically aligned with the 8–10 year warranty period, resulting in an assumed lifespan of 8–12 years [58, 63, 64, 73, 74, 76, 79–89]. However, this approach has been criticized by Circular Energy Storage [199, 200], a consultancy specializing in end-of-life LIB market data, whose findings are also referenced by the International Energy Agency [191]. They argue that these conservative estimates not only contradict their real-world data, which suggests that vehicles retain enough value to last over 15 years but also fail to account for changes in usage patterns as vehicles age. While older cars may not be suitable for long-distance travel, they remain practical for shorter commutes and daily errands. This is consistent with the fact that the average annual distance traveled by cars in the EU is less than 15,000 km [201, 202], and modern EVs typically have a range exceeding 300 km [203], requiring only about 50 full charge cycles per year (well below the 125 full charge cycle implied by a 12 years lifespan).

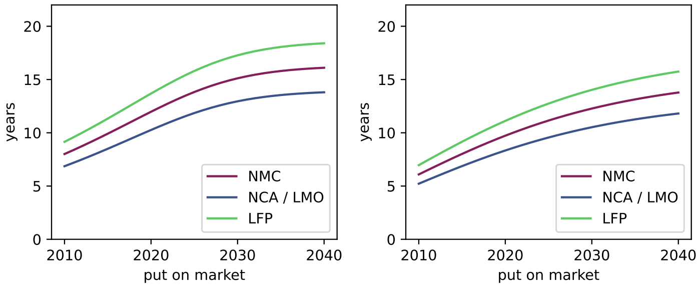


Figure 3.3. Average battery lifetime.

Although fewer studies model a longer battery lifespan — typically around

15–16 years [59, 69, 70, 72] - both conservative and longer estimates overlook the role of technological innovation. This oversight is significant, given that model time frames often extend up to 30–40 years, during which substantial improvements in battery technology are expected. Therefore, we align with studies that incorporate technological evolution into their projections [60, 61, 65, 67, 71, 75, 78]. Specifically, we assume a Weibull distribution with a shape parameter of 3.5 and model the average battery lifetime for NMC chemistries to increase from 8 years in 2010 to 16 years by 2035. Additionally, we consider a more conservative scenario where the average battery lifetime for NMC chemistries increases from 6 years in 2010 to 14 years by 2040. In both cases, LFP chemistries are modeled to last 15% longer, while NCA and LMO chemistries are projected to have lifetimes 15% shorter, reflecting the differences in their life cycles, as discussed in chapter 2. For vehicle lifetimes, we assume a baseline Weibull distribution with a shape parameter of 3.75 and a scale parameter of 19 to align with usage patterns reported in [204]. A more conservative Weibull distribution with a shape of 5.5 and a scale of 15 is also considered in the sensitivity analysis, based on data from Circular Energy Storage [205].

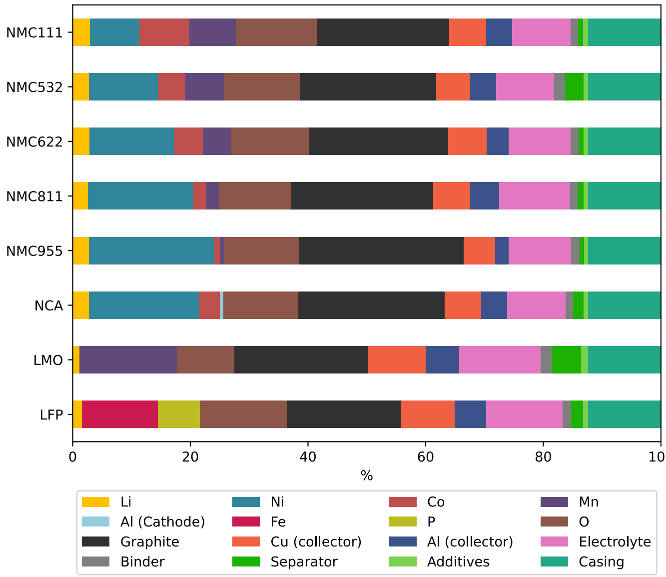


Figure 3.4. LIB composition. Source: [76, 103, 151, 206–210]

3.1.4. LIB composition

The various chemistries represent the top level of the composition hierarchy (i.e. the product layer). The component layer follows the description from figure 2.2. They are further divided into their material constituents (metal oxide, plastics, steel, solvents, etc.) and the corresponding chemical elements. A simplified representation is provided above (figure 3.4).

Performance improvements are expected to translate into changes in battery composition. For example, while anodes are predominantly made of graphite, silicon is increasingly being introduced as an additive to enhance performance, and its use is expected to grow in the future [7, 11, 37]. However, due to the lack of up-to-date data on these advancements, we assume a consistent composition for modeling purposes. We do account nonetheless for a decreasing material intensity per energy unit over time (see figure A.4 in Appendix).

3.2. Multilayer MFA

3.2.1. Conceptual framework

TC in simple MFA systems

In MFAs, TCs are a key concept used to quantitatively describe how materials flow through various stages of a system [125, 211, 212]. They represent the ratio of material transferred from one stage to another and are instrumental in identifying inefficiencies and bottlenecks in material handling and processing. Additionally, TCs are valuable for scenario modeling and impact prediction. They help evaluate the effects of changes such as the introduction of new technologies, modifications in operational practices, or adjustments to regulatory requirements. Figure 3.5 illustrates a simple MFA system with the mass transfer equations expressed below.

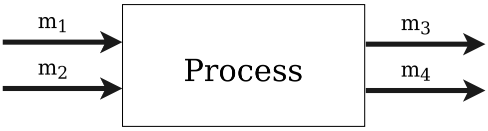


Figure 3.5. A simple mass transfer

$$\begin{cases} m_3 = a_{31}m_1 + a_{32}m_2 \\ m_4 = a_{41}m_1 + a_{42}m_2 \end{cases} \Rightarrow \begin{cases} -a_{31}m_1 - a_{32}m_2 + m_3 = 0 \\ -a_{41}m_1 - a_{42}m_2 + m_4 = 0 \end{cases}$$

Assuming that m_1 and m_2 are known (e.g., equal to y_1 and y_2), we get:

$$\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ -a_{31} & -a_{32} & 1 & 0 \\ -a_{41} & -a_{42} & 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} m_1 \\ m_2 \\ m_3 \\ m_4 \end{bmatrix} = \begin{bmatrix} y_1 \\ y_2 \\ 0 \\ 0 \end{bmatrix}$$

Extending TC to multi-layer MFA systems

Next, we examine a single-layer system where the layer accounts for all the chemical elements present in the flows. Figure 3.6 illustrates a system composed of two elements (E_1 in blue, and E_2 in yellow). The corresponding algebraic expression is given below assuming that m_1 to m_4 are known (equal to y_1 to y_4).

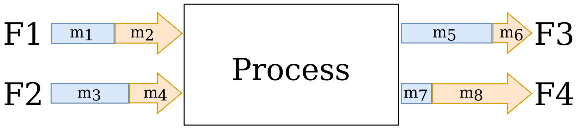


Figure 3.6. A single layer mass transfer where each flow is composed of 2 elements

$$\begin{cases} m_5 = a_{51}m_1 + a_{53}m_3 \\ m_6 = a_{62}m_2 + a_{64}m_4 \\ m_7 = a_{71}m_1 + a_{73}m_3 \\ m_8 = a_{82}m_2 + a_{84}m_4 \end{cases} \Rightarrow \begin{cases} -a_{51}m_1 - a_{53}m_3 + m_5 = 0 \\ -a_{62}m_2 - a_{64}m_4 + m_6 = 0 \\ -a_{71}m_1 - a_{73}m_3 + m_7 = 0 \\ -a_{82}m_2 - a_{84}m_4 + m_8 = 0 \end{cases}$$

$$\begin{bmatrix} 1 & & & & & & & \\ & 1 & & & & & & \\ & & 1 & & & & & \\ & & & 1 & & & & \\ -a_{51} & & -a_{53} & & 1 & & & \\ & -a_{62} & & -a_{64} & & 1 & & \\ -a_{71} & & -a_{73} & & & & 1 & \\ & -a_{82} & & -a_{84} & & & & 1 \end{bmatrix} \cdot \begin{bmatrix} m_1 \\ m_2 \\ m_3 \\ m_4 \\ m_5 \\ m_6 \\ m_7 \\ m_8 \end{bmatrix} = \begin{bmatrix} y_1 \\ y_2 \\ y_3 \\ y_4 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}$$

This can also be written as:

$$(\mathbf{I} - \mathbf{A})\mathbf{x} = \mathbf{y} \tag{3.1}$$

with $\mathbf{I}$ the identity matrix, $\mathbf{A}$ the technology matrix that records the TCs a_{ij} associated with the mass transfer from m_j to m_i, $\mathbf{x}$ the masses to be determined (i.e., the vector composed of each mass m_i), and $\mathbf{y}$ the inputs. The computational structure is thus similar to LCA and input-output analysis [213–215]. The parallel with the technology matrix from IO tables is further illustrated in figure 3.7.

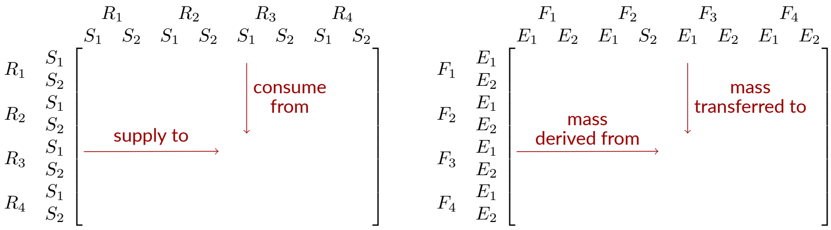
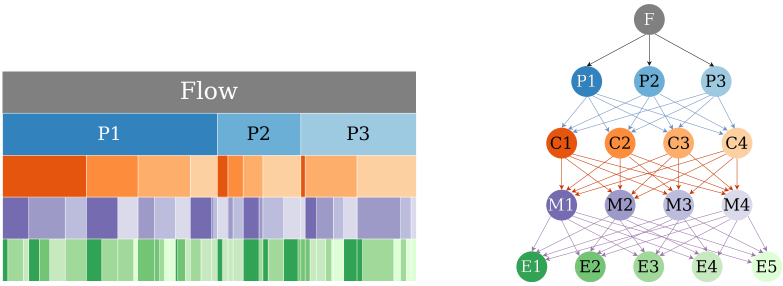


Figure 3.7. (left) example of a technology matrix with 4 regions (R_1, R_2, R_3 and R_4) and 2 sectors (S_1 and S_2) as conceptualised in MRIO tables; (right) technology matrix associated with figure 3.6.

Equation (3.1) can be expanded to encompass any number of layers without loss of generality. Similarly to Multi-Regional Input-Output tables, this is done by considering the combination (i.e., the Cartesian product) of every possible option for each layer. In this study, we focus on four layers: products, components, materials, and elements (figure 3.8).



(a) Treemap representation **(b)** Network representation

Figure 3.8. Multi-layer representation of a flow as a treemap (left) and a network (right). A network edge $u \rightarrow v$ represents the mass of v contained in u. For clarity, not every edge is represented. For example, there are actually 3 edges between $C1$ and $M1$ depending on if $M1$ is part of $P1$, $P2$ or $P3$.

3.2.2. Implementation

In practice, TCs can be understood as the product of process efficiency and market share. The market shares are directly derived from the previous literature review (figure 2.6) [190], with each plant assumed to process a portion of the EOL volume based on its capacity. It is further assumed that all batteries are collected, in line with the battery directive and consistent with practices for lead-acid batteries used in internal combustion engine vehicles. The following section details the data collection process for determining the process efficiencies. For a full overview of the model algorithm and its integration with the dMFA, we redirect the reader to appendix D.

Deactivation and dismantling

Quantifying the number of recycling plants that rely primarily on manual disassembly is challenging, but conducting the dMFA at the cell level helps mitigate this uncertainty. By focusing on the cell level, the analysis bypasses the need to account for pack and module components such as casings, wires, and coolant which are more likely to be targeted by manual dismantling. Additionally, mechanical pre-treatment can be seen as a form of rough and automated dismantling. Modeling deactivation is equally difficult since the boundary between deactivation and mechanical pretreatment can be blurry (e.g., see wet crushing). However, from a mass balance perspective, deactivation mainly results in the loss of electrolytes, a loss that also occurs during mechanical treatment. Consequently, the deactivation and dismantling steps are not explicitly modeled.

Mechanical pre-treatment

The literature reveals high recovery rates for cathode materials. Crushing and sieving alone achieve recovery rates of 80% to 90% for active materials, and 80% for aluminum and copper [140, 150, 154]. Incorporating additional separation techniques increases recovery efficiency, often reaching 96% to 99% for both the electrodes and collectors [100, 152, 153, 155, 160, 216]. Post-mechanical processing significantly enhances the purity of black mass. With a thermal or chemical dissolution treatment, over 99% of the binder can be removed which allows for the separation of cathode active materials from aluminum to exceed 97%, compared to only 83% to 85% without such processing [140, 150–152, 154, 155, 162, 174, 217]. Latini et al. [101] estimate recovery efficiencies of 75%-90% when binder removal steps are not employed. These estimates align with model data from Life Cycle Assessments (LCA) and techno-economic evaluations. Although the EverBatt model [209] and Ali et al.'s model [218] do not explicitly state the recovery rates for mechanical pre-treatment, recovery rates of 97.4% for both cathode and anode materials can be inferred. The two studies include a thermal treatment. In contrast, Woeste et al.[219], who omit this step, report a 96% recovery rate for cathode active material, a 94% recovery rate for copper, and only 85% for aluminum and 86% for steel, with the remaining metal fractions contaminating the black mass. They also report a 79% recovery rate for graphite against 96.6% for Dai et al. However, Dai and colleagues also assume that 6.9% of the binder is transferred to the black mass despite thermal treatment. While a few plants publicly disclose the use of thermal pre-treatment, most remain secretive about the specific processes they implement, leaving many details unknown. As a result, we included this aspect in the sensitivity analysis. The analysis considered both the current market scenario, with known market shares, and a progressive scenario in which all companies adopt some form of post-processing starting in 2022

Finally, battery chemistry and design significantly influence black mass yield and purity, as highlighted by Wilke et al. [220]. Their study reports recovery rates for cathode materials that are notably lower compared to other sources (40-60% on average for the cathode materials). But this

Table 3.1. Transfer coefficients for mechanical pre-treatment. LiPF6: lithium hexafluorophosphate. EC: ethylene carbonate. DMC: dimethyl carbonate. PVDF: Polyvinylidene fluoride. SBR: styrene-butadiene rubber. CMC: carboxymethyl cellulose. PE: polyethylene. PP: polypropylene

Inflow			Outflow			advanced	baseline
Cathode	in	EoL	Cathode	in	Black Mass	98	94
Anode	in	EoL	Anode	in	Black Mass	97	79
Collector (cathode)	in	EoL	Al alloy	in	Al & Cu scrap	96	85
Collector (cathode)	in	EoL	Al alloy	in	Black Mass	4	15
Collector (anode)	in	EoL	Cu alloy	in	Al & Cu scrap	97	90
Collector (anode)	in	EoL	Cu alloy	in	Black Mass	3	10
Binder	in	EoL	PVDF / SBR / CMC	in	Black Mass	5	15
Electrolyte	in	EoL	LiPF6 /EC /DMC	in	Black Mass	0	10
Separator	in	EoL	PE /PP	in	Black Mass	0	10
Additive	in	EoL	Carbon black	in	Black Mass	95	100
Casing (Al)	in	EoL	Al alloy	in	Al & Cu scrap	90	90
Casing (steel)	in	EoL	Steel alloy	in	Steel scrap	90	90
Casing (Al & steel)	in	EoL	Al /steel alloy	in	Black Mass	10	10
Casing (plastic)	in	EoL	Plastic	in	Plastics	50	50
Casing (plastic)	in	EoL	Plastic	in	Black Mass	0	10

lower recovery rate is likely due to their use of a simpler mechanical treatment, which does not accurately reflect industrial practices. Wilke et al. also mention that recovery rates can reach 95% with further processing steps. Given the limited data on how specific battery chemistry and design impact recovery rates, we simplified the model by adopting uniform recovery rates across all battery types. We also assume that separation techniques based on electromagnetic properties do not significantly break down the active material from LFP or NCA despite the presence of iron and aluminum. As a result, we use a recovery rate at the component level (e.g., cathode) rather than differentiating by specific chemical elements (e.g., lithium, nickel, etc.). A summary of the efficiency is provided in table 3.1. To ensure consistency, the model results were compared with the black mass composition reported in the literature (see table C.1 in appendix).

Pyrometallurgy

Pyrometallurgy achieves high recovery rates for heavier metals (typically 99% for nickel, 94% for cobalt, 93% for copper, and 64% for iron), while the remainder typically ends up in the slag [101, 208, 221]. The EverBatt model reports comparable efficiencies, with recovery rates of 98% for nickel and cobalt and 90% for copper [209]. However, it assumes a 90% recovery rate for iron, which is closer to the theoretical value of 97.8% reported by Rajaeifar et al [221]. Dobó et al. report nearly 99% recovery for cobalt and nickel and approximately 94% for copper, whereas Chen et al. report rates above 99% for all three metals [167]. Most studies, however, report final recovery rates that include the hydrometallurgical refining step. Generally, recovery rates range between 93% and 99% for nickel, 95% and 99% for cobalt, and 93% to 99% for copper [102, 103, 165, 167]. Bruno and Fiore [73] note more variability for cobalt, with a recovery rate of 86% ± 15%. However, they mention a recovery rate of 99% for aluminum and 88% for manganese, which contradicts the process description, as these elements usually end up in the slag. They also report no recovery of iron. The recovery rates for aluminum and manganese could be accurate if the slag is processed by hydrometallurgy. In that case, manganese recovery ranges between 79% and 99%, and similarly for lithium [92, 102, 103, 165, 167].

Table 3.2. Transfer coefficients for pyrometallurgy (in %). BM: Black Mass, S: Smelting, CTR: Carbothermic reduction, SAR: Salt-Assisted Roasting

Inflow			Outflow			S / CTR	SAR
Li	in	EoL / BM	Li	in	Alloy	0	0
Li	in	EoL / BM	Li	in	Flue dust	0	80
Li	in	EoL / BM	Li	in	Slag	100	20
Ni	in	EoL / BM	Ni	in	Alloy	93-99	93-99
Ni	in	EoL / BM	Ni	in	Slag	1-7	1-7
Co	in	EoL / BM	Co	in	Alloy	95-99	95-99
Co	in	EoL / BM	Co	in	Slag	1-5	1-5
Cu	in	EoL / BM	Cu	in	Alloy	93-99	93-99
Cu	in	EoL / BM	Cu	in	Slag	1-7	1-7
Mn	in	EoL / BM	Mn	in	Alloy	0	0
Mn	in	EoL / BM	Mn	in	Slag	100	100
Fe	in	EoL / BM	Fe	in	Alloy	64-90	64-90
Fe	in	EoL / BM	Fe	in	Slag	10-36	10-36

As mentioned in chapter 2, the slag is usually not treated for economic reasons. Consequently, manganese, aluminum, and lithium are considered lost after pyrometallurgy. The main exception is SAR. With this method, 60% to 70% of Lithium can be recovered in the flue dust, followed by an 80% to 90% recovery rate during the leaching step [169, 222]. Hu et al. also suggest that lithium recovery could reach as high as 91.4% with adjustments to certain parameters [222]. This could explain the 70% recovery rate claimed by Umicore, a leading global player in pyrometallurgy [210, 223, 224]. Accordingly, we assume the recovery of Lithium in the flue dust to be 80%, and the recovery of transition metals to be the same across technologies table 3.2. All other components are burnt for energy and are therefore lost.

Hydrometallurgy

The literature reports high efficiencies for both leaching and separation stages, with some nuances depending on the methods used [105, 163, 166, 167, 169, 172–174, 182, 225]. Leaching using inorganic acids typically yields 95-99% recovery for most metals (Nickel, Cobalt, Manganese, Copper), while organic acids can achieve 90-98% efficiency. Alkali methods can also achieve above 90% recovery for certain metals. Lithium recovery efficiencies typically range from 82% to 99%, while recovery rates for cobalt, nickel, and manganese are often slightly higher, generally between 95% and 99%. The subsequent precipitation or extraction steps also show high efficiencies, usually falling within the 90% to 99% range.

However, these overall efficiencies can be misleading, as the order of metal extraction is crucial. Lithium is usually recovered last due to its chemical properties, making it more susceptible to losses throughout the previous precipitation [95, 163, 169, 178, 181, 182]. This is why the recovery target for lithium specified in the EU Battery Regulation is lower than other metals. Currently, very few companies have achieved a recovery rate of Lithium exceeding 90% [226, 227], all of them leading companies from China or Korea, where the battery recycling industry has over a decade of experience ahead of European competitors.

Graphite, aluminum, and iron are often discarded during recycling due to limited economic incentives. However, the growing recognition of graphite's criticality, along with advancements in recovery methods and its relatively high mass fraction in black mass, could change this trend [186, 228–231]. Emerging techniques for the recovery and purification of graphite are improving the feasibility of recycling, and an increasing num-

Table 3.3. Transfer coefficients for hydrometallurgy (in %).

Inflow			*Outflow*			*Value*
Li	in	Black Mass	Li	in	Metal Salts	85
Li	in	Flue Dust	Li	in	Metal Salts	95
Ni	in	Black Mass	Ni	in	Metal Salts	95
Ni	in	Alloy	Ni	in	Metal Salts	99
Co	in	Black Mass	Co	in	Metal Salts	95
Co	in	Alloy	Co	in	Metal Salts	99
Cu	in	Black Mass	Cu	in	Metal Salts	95
Cu	in	Alloy	Cu	in	Metal Salts	99
Mn	in	Black Mass	Mn	in	Metal Salts	95
Graphite	in	Black Mass	Graphite	in	Slurry	95

ber of recyclers are now expressing their intention to recover this valuable material [190]. We assume a graphite recovery rate of 95% based on [43, 65, 79], but we only account for companies that explicitly declare their plans to recover it (from 20% currently to 30% by 2030). To account for uncertainty, we include this aspect in the sensitivity analysis by also considering recyclers with limited information about their processes, which could represent approximately 70% by 2030.

Finally, recovery rates of Nickel, Cobalt and Copper from the matté obtained by smelting and CTR are taken from [92, 166, 168, 232], and the recovery rate of Lithium contained in the flue dust from SAR is assumed based on the overall efficiency reported by Umicore [210, 223, 224].

Direct recycling

To maintain consistency across the various recycling routes and account for data limitations, we assume that lithium loss due to side reactions during the battery's life, as well as the dissolution of metals such as nickel, manganese, and cobalt into the electrolyte or their migration to the anode, is negligible. Additionally, we simplify the modeling of direct recycling by assuming a 90% recovery rate as assumed in [47, 70]. Lastly, we exclude upcycling possibilities from this analysis, as they remain in a more speculative stage than direct recycling.

4

Results

4.1. EOL EV batteries

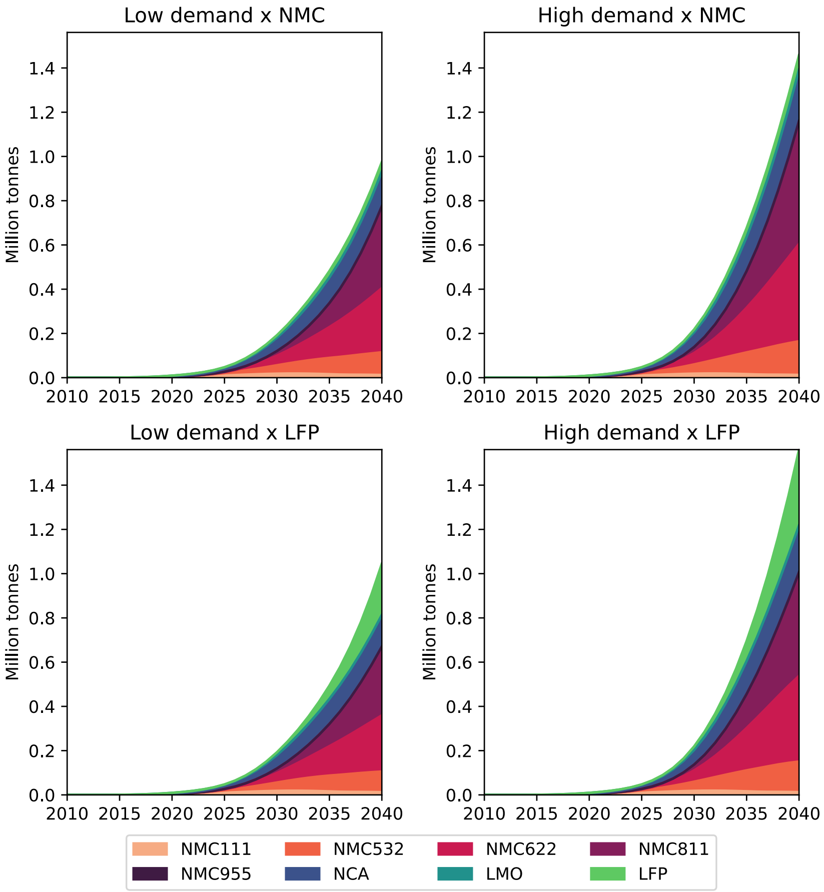


Figure 4.1. Batteries reaching End-of-Life, assuming an optimistic lifetime for batteries and EVs (in Mt).

The speed of EV adoption significantly impacts the volume of batteries reaching End-of-Life, particularly by the mid-2030s, where volumes could be 40-50% higher across all scenarios. Interestingly, battery lifetime has a comparable effect: a shorter battery lifetime typically increases volumes by over 40%, and in some cases, this rise exceeds 60% by the early 2030s. In contrast, vehicle lifetime has a negligible impact on battery volumes, with less than a 1% difference across scenarios, which is sensible given the modest increase in battery capacity assumed. The lower efficiency of LFP chemistries plays a secondary role, leading to an average 8-9% increase in volumes if they come to dominate the market. Therefore, obtaining more accurate data on actual battery lifetimes will be critical to accompany the scaling of the recycling industry, especially in the next decade.

4.2. European recycling capacity

Figure 4.2 provides an overview of the capacities for each recycling routes considered in the analysis, depending on the combination of processes used on each plant identified in the literature review [190]. One notable trend is the specialization and decentralization of recycling plants, which follow a ”hub-and-spoke” business model. The spokes represent mechanical pre-treatment plants, which are typically located near areas where EVs are disposed of, while the hubs are hydrometallurgical facilities, fewer in number and generally situated closer to cell manufacturing plants. Some facilities, however, choose to manage the entire recycling process, from the dismantling of EOL batteries to the production of precursors. Regarding pyrometallurgy, capacity remains relatively stable overall with a notable increase expected in 2026. This surge is due to a single plant operated by Umicore, which is set to begin operations that year and will significantly expand its capacity from 7,000 t/a to 157,000 t/a. Despite this increase, pyrometallurgy is anticipated to lose its current dominance, with mechanical and hydrometallurgical recycling routes expected to become more prevalent.

Europe is currently in overcapacity for recycling spent EV batteries (figure 4.3). Although this estimate does not account for batteries used in other energy storage systems or electronics, nor for production scrap, the margin remains large. In all scenarios, the volume of EOL batteries is projected to exceed 50% of recycling capacity only well after 2030. Addi-

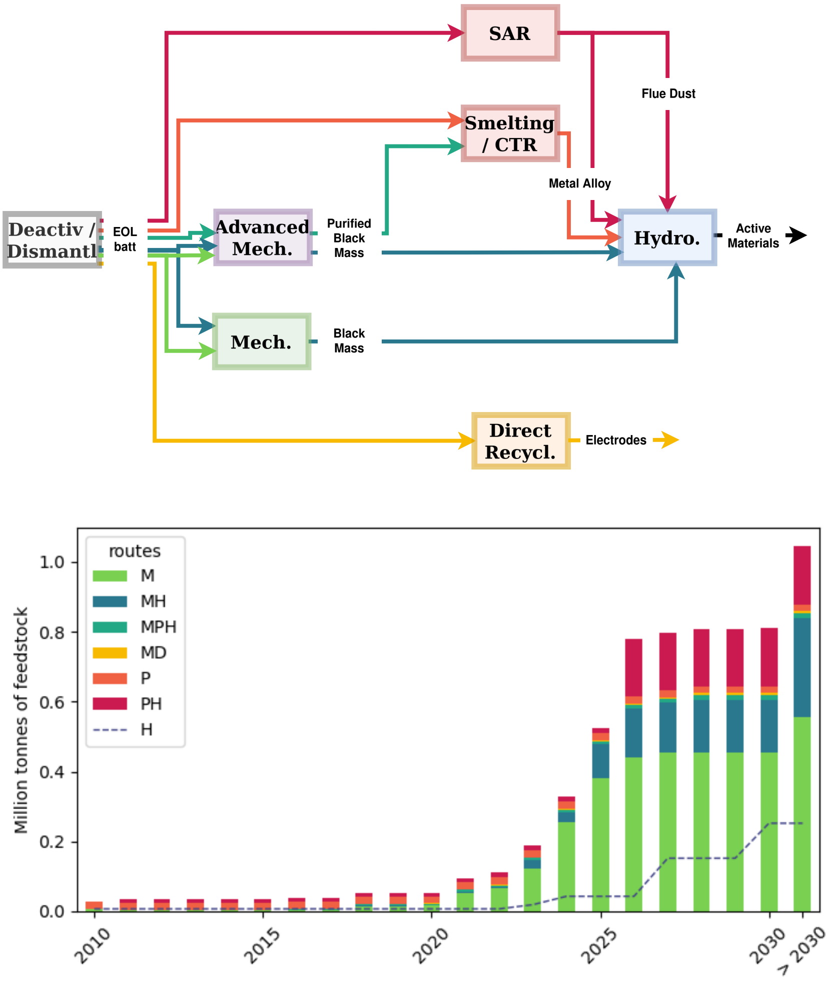


Figure 4.2. Overview of main processes in the European LIB recycling supply chain (top) and the associated capacities for each pathway (bottom). For clarity, only flows between processes are represented on the diagram. Plants that combine different treatment processes are grouped together on the graph to ensure consistency in terms of input/output. As such, single hydrometallurgical plants are shown separately since they process black mass as feedstock instead of spent batteries. (M = Mechanical pre-treatment; H = Hydrometallurgy; Pyrometallurgy; MD = Direct recycling).

tionally, the capacity is likely to increase, as the construction of recycling plants is typically announced only a few years in advance. However, the excess capacity until 2030 will lead to intense price competition, which may drive some companies out of the market. As a result, the total recycling capacity is also likely to change before 2030.

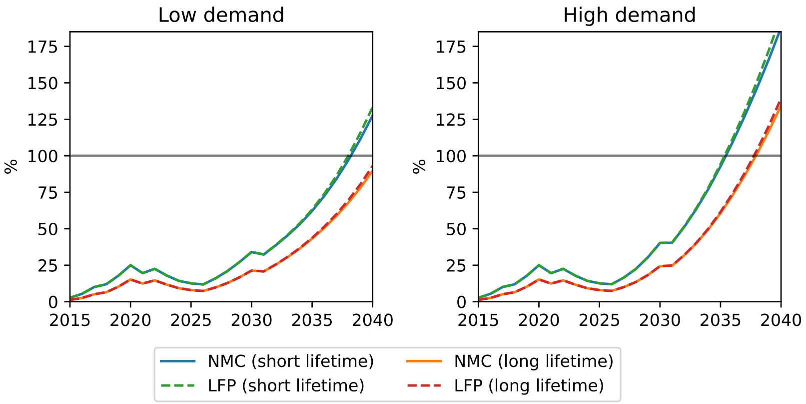


Figure 4.3. Utilization of recycling capacity resulting from spent EV batteries.

While market uncertainty is unlikely to affect the overall capacity to recycle spent batteries, bottlenecks are more likely to emerge downstream. Most recycling capacity resides in facilities that implement only mechanical pre-treatment, relying on external plants to process the resulting black mass. By tracking the whole range of constituents present in the black mass, the total production can be estimated precisely. Figure 4.4 reveals that the amount of black mass produced is substantial compared to the capacity of stand-alone hydrometallurgical facilities, with capacity being exceeded before 2040 in all scenarios. Although post-processing steps can be integrated into mechanical pre-treatment to remove impurities, this will not significantly reduce the overall volumes. In contrast, if pyrometallurgical processes remain the dominant recycling method despite the growing competition from mechanical treatment facilities, it could help alleviate concerns about black mass processing capacity and reduce the need to export black mass, as is currently done. This scenario is plausible, given that pyrometallurgy does not require battery sorting or extensive pre-processing and can be scaled more easily. In any case, the results highlight the importance of including spent batteries from energy storage systems, electronics, and production scrap in future analyses.

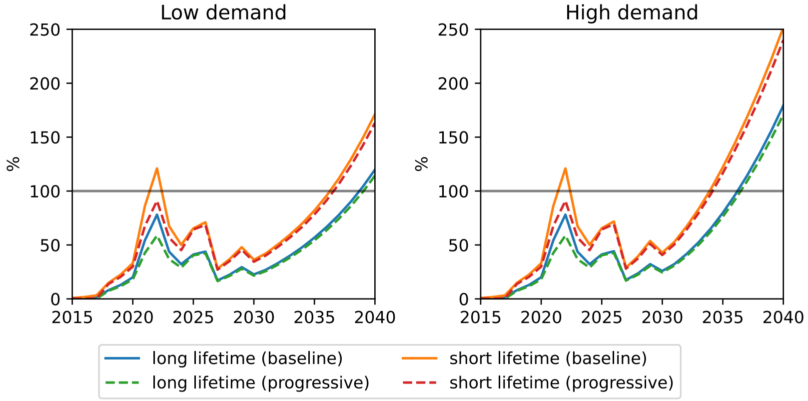


Figure 4.4. Capacity utilization of stand-alone hydrometallurgical plants (lifetime refers to the battery lifespan; baseline indicates the current post-processing practices, while progressive represents a scenario where all mechanical treatment facilities implement post-processing).

4.3. CRM recovery

The total mass of recovered materials, like the volumes of EOL batteries, will vary considerably depending on EV demand and battery lifetime. Recovery quantities range from 18–37 kt for lithium, 122–255 kt for nickel, 27–54 kt for cobalt, 19–38 kt for manganese, and 57–121 kt for copper (figure 4.5). These parameters also amplify uncertainties in nickel, cobalt, manganese, and copper recovery, as these are the metals most affected by the shift from NMC to LFP chemistries. In contrast, lithium recovery is less influenced by chemistry market share due to its relatively consistent mass fraction across different battery types. Similarly, the quantities of aluminum and iron recovered are not significantly impacted by the battery chemistry (e.g., NCA or LFP), as these metals primarily come from the battery casing rather than the cathode. The greatest uncertainty lies in graphite recovery, depending on how quickly and widely recyclers adapt their recycling processes. The balance between pyrometallurgical and hydrometallurgical routes will also be critical, as graphite is lost in the former, although this was not modeled.

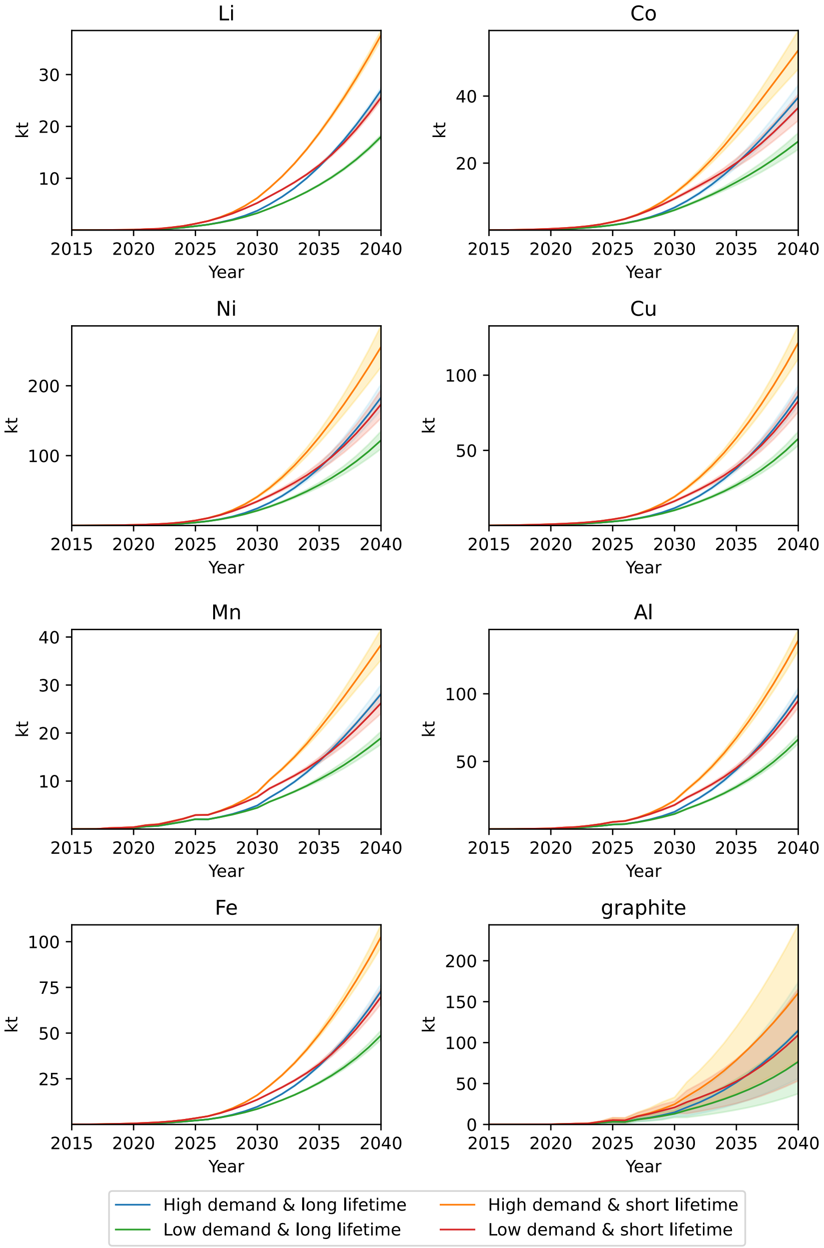


Figure 4.5. CRM recovery based on EV demand and battery lifetime.

4.4. Battery directive targets

4.4.1. Target 1: recycling efficiency

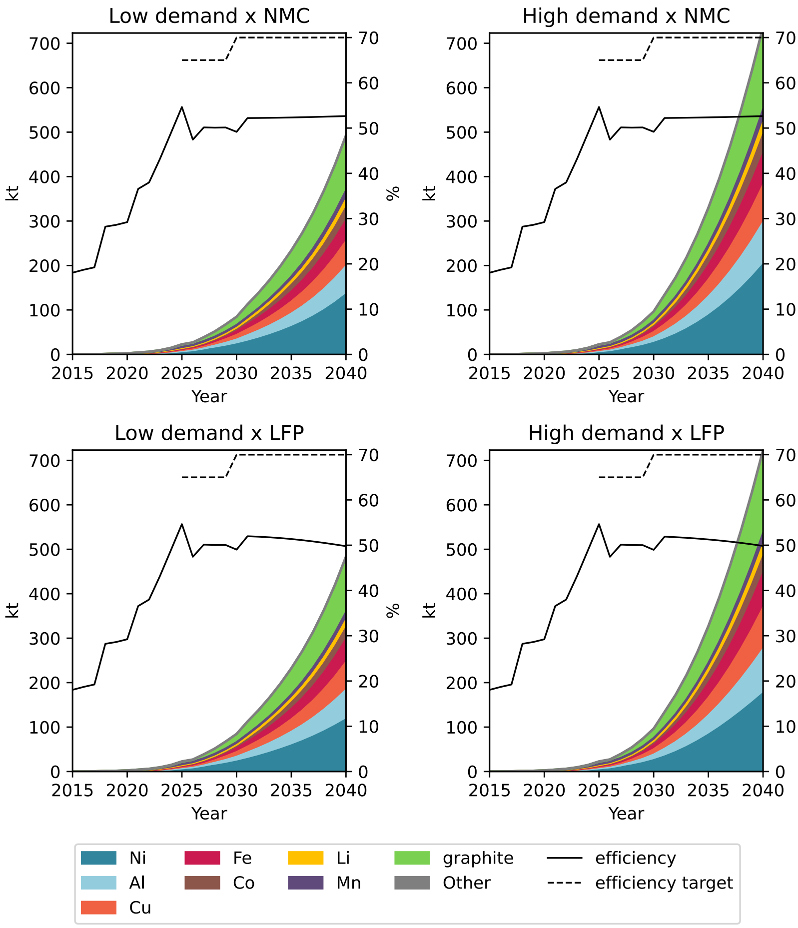


Figure 4.6. Recycling efficiency in case of long EV and battery lifetimes.

By 2030, approximately 50% of the total weight of batteries is projected to be recycled in the case where the recovery of graphite is actively being pursued, and just over 40% otherwise (figures 4.6, C.1 and C.2). In both cases, this falls short of the EU battery directive's targets of 65% by 2025 and 70% by 2030. The slight difference in efficiency between the LFP and NMC scenarios is due to the presence of iron in the black mass, which is discarded after hydrometallurgy. Given the mass fraction of the cathode active material in the overall battery cell, any improvement in its recovery rate will significantly impact the overall recycling efficiency. However, an analysis of material leakage by components shows that even with optimal recovery of cathode and casing materials, meeting the EU target is unlikely (see appendix C.2). To bridge the gap, optimal recovery of anodes is essential, which effectively rules out pyrometallurgical routes. Finally, while electrolyte recovery is actively being researched [178, 233–236], its potential contribution to overall recycling efficiency remains uncertain due to challenges posed by its reactivity and toxicity. Since it is unsuitable for reuse in batteries and energy recovery is not considered recycling under the battery directive, its impact seems limited.

4.4.2. Target 2: recovery rates

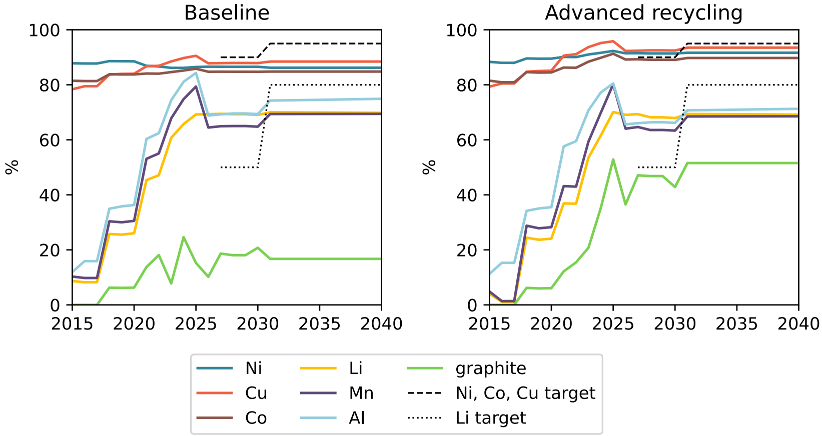


Figure 4.7. Recovery rates.

Figure 4.7 reveals that the recovery rate targets remain out of reach in the short term, particularly without widespread adoption of post-processing treatments. This challenge affects all critical raw materials covered by the directive: nickel, copper, cobalt, and lithium but lithium currently presents the greatest hurdle. While achieving a 70% recovery rate for lithium is feasible and sufficient to meet the 2027 target of 50%, it falls short of the 2031 target of 80%. As discussed in chapter 3 only a few companies can currently meet this requirement. In contrast, advanced recycling treatment improves recovery rates for Nickel (92% vs. 86%), Cobalt (90% vs.

85%), and Copper (93% vs. 88%), though these rates are still not entirely sufficient. The varying recovery rates among recycling industry players are likely to drive market consolidation, which is expected to result in overall improvements in average recovery efficiencies. Additionally, the introduction of the battery digital passport in 2027 is anticipated to enhance sorting processes and improve waste stream purity. Combined with ongoing technological advancements, this suggests that the recovery targets set by the directive are achievable within the specified timeframe.

4.4.3. Target 3: minimum recycling content

Even without factoring in production scrap — which currently drives the secondary material supply [5, 191] — the reserves in EOL batteries within Europe are projected to be sufficient to meet the targets across all scenarios (figure 4.8). These findings challenge previous results from the literature [55, 72, 74, 77]. For example, Barkhausen suggests that the recycling content targets are unlikely to be met by 2031 if the average battery lifetime exceeds 12 years, with projected material supply of 4% for lithium and 5% for nickel and cobalt [74]. While his outlook improves for 2036 for most metals, he forecasts that Cobalt could only reach 23%, falling short of the 26% mandated by the directive. He is also the only one to account for Copper and calculates a maximum material supply of 21%, which is aligned with the more conservative estimate of this study.

The discrepancy in results is more pronounced when compared to Boston Consulting Group's, which only predict a maximum material supply of 2% for both lithium and nickel in 2031, and 8-10% by 2036 — well below the targets of 6% and 12% for lithium, and 6% and 15% for nickel in those respective years [77]. The gap is smaller for Cobalt, and they equally estimate that the targets could be met.

Finally, Ginster et al. argue that the targets are unlikely to be achieved without incorporating production scrap. Out of their seven modeled scenarios, only two meet the recycling content targets based solely on EOL battery volumes, both of which assume a shorter battery lifetime, similar to the more conservative one used in this study.

While we have not accounted for imports and exports, this factor alone is unlikely to explain the discrepancies in findings as Ginster and colleagues do not account for them either, and Barkhausen restricts them to just 4%.

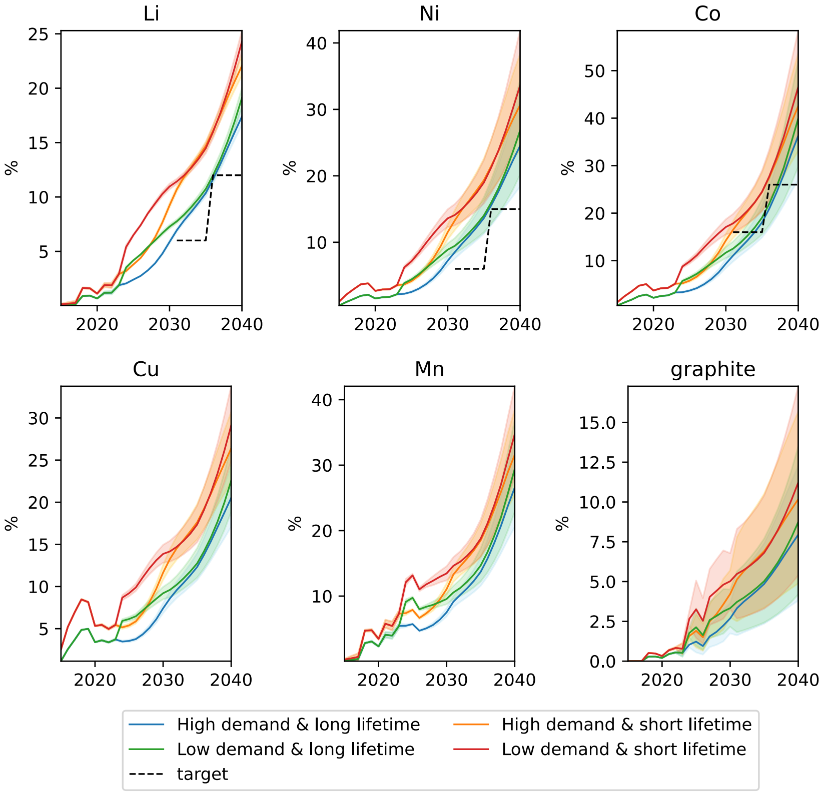


Figure 4.8. Potential recycling content (ratio of recoverable CRM from waste to CRM requirements induced by EV demand). Targets for 2031 are Li and Ni: 6%; Co: 16%. Targets for 2036 are Li: 12%; Ni: 15%; Co: 26%.

5

Conclusion & perspectives

5.1. Main findings

The volume of end-of-life EV batteries is projected to grow exponentially from the second half of the 2020s, reaching between 1 and 1.5 million tonnes by 2040. This surge will be primarily constituted of high-nickel NMC batteries, even if trends in LFP batteries market share penetration continues. Besides the rapid adoption of EVs, battery lifetime will be the most influential factor in determining the quantities entering waste streams. However, current literature appears to be conservative, forecasting battery lifetimes of 8-12 years, which contrasts with emerging field data suggesting longer operational lifespans.

European recycling capacities are expected to exceed the volumes of EOL EV batteries, likely intensifying competition, with resource recovery becoming a key differentiator. Hydrometallurgical plants are the most likely to face bottlenecks, potentially leading to increased black mass exports. In contrast, pyrometallurgical processes are anticipated to lose market share to mechanical treatment methods despite greater flexibility and higher cost effectiveness.

Regarding the circularity targets set by the EU Battery Directive, there is high confidence that recovery rates and minimum recycling content can be achieved within the specified timeframe. However, achieving the target of recycling 65-70% of the average battery weight remains a significant challenge, particularly without maximizing recovery from both cathodes and anodes. This goal will be especially difficult for pyrometallurgical industries and will necessitate substantial changes in hydrometallurgical practices, potentially leading to increased environmental impacts.

5.2. Limitations

While this study provides important insights into the circularity of critical raw materials, it is limited by the exclusion of production scrap and certain battery waste streams. These omissions extend beyond electronics and energy storage systems and include the mobility sector too with two-wheelers, electric bicycles, buses, and trucks, all likely to significantly affect overall EOL volumes [5, 200, 237]. Additionally, the exclusion of exports and informal losses, such as illegal battery exports or unregulated scrapping, could result in overestimations of the minimum recycling content. However, it is important to note that this specific requirement applies only to mobility and industrial batteries, excluding electronics. Since electronic devices are predominantly manufactured in Asia, critical raw materials from electronics could be redirected to the mobility sector, potentially mitigating the impact of export-related losses. Moreover, since China and the US lack equivalent minimum recycling content regulations, recovered materials could be allocated preferentially to the European automotive market without jeopardizing international competitiveness.

The study also did not account for future changes in battery composition, such as the integration of silicon in anodes or the adoption of emerging technologies like sodium or solid-state batteries [8–11]. These innovations will inevitably affect the recycling landscape but remain underexplored in the literature.

Finally, the analysis overlooked the reuse and repurposing of LIB, despite ongoing initiatives exploring this market opportunity [238]. Both could extend battery lifetimes and delay the availability of materials for new EV batteries but may also reduce the demand for virgin materials in other sectors. This highlights the importance of expanding the scope to other end-use applications. It also underscores the risk of focusing solely on circularity targets set by the battery directive and the importance of considering trade-offs with other circular economy practices.

5.3. Future research on CRM recoverability

The model excludes certain processes, such as sorting, and assumes that all battery chemistries are processed equally by each recycling route. This assumption is unrealistic. For instance, recycling Lithium Iron Phosphate

(LFP) batteries is less economically viable, as lithium is their only valuable component. If LFP's market share continues to rise, efficient sorting will become increasingly critical, particularly for pyrometallurgical processes, which are less effective at recovering lithium. It could also lead to maximizing the utilization of direct recycling capacities. This illustrates the need to simultaneously keep both data levels to determine the optimal route. Although time constraints limited the full exploration of this approach, the framework developed here lays the groundwork for future research, particularly when expanding the scope to include other battery types such as Nickel Cadmium (NiCd) and Nickel Metal Hybrid (NiMH). These types of batteries are still very present in electronics which has lead some recyclers to modify their processes to handle mixed feedstocks.

Incorporating intermediate layers can also help pinpoint where value is lost during recycling. For example it can aid in analyzing material leakage by tracking the original components and products throughout the system. Furthermore, and as discussed in the review of recycling processes, there is a growing trend to preserve chemical compounds, such as cathode active materials, rather than breaking them down into individual elements. A more flexible framework that allows for analysis at varying levels of material aggregation would enable more accurate calculations of stoichiometric reactions and the corresponding quantities of reagents required. This is particularly relevant given the widespread but often misguided assumptions regarding the environmental benefits of hydrometallurgical processes [207]. The spatial and temporal dimensions of the data collected on European recycling capacities could further enhance the precision of techno-economic-environmental analyses and multi-criteria decision-making. This approach will become increasingly important for analyzing complex waste streams generated by mixed inputs and for navigating the intricate trade-offs between economic, environmental, and regulatory dimensions [115, 239–241].

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Appendix

A

EOL batteries

A.1. Model assumptions

A.1.1. BEV / PHEV market shares

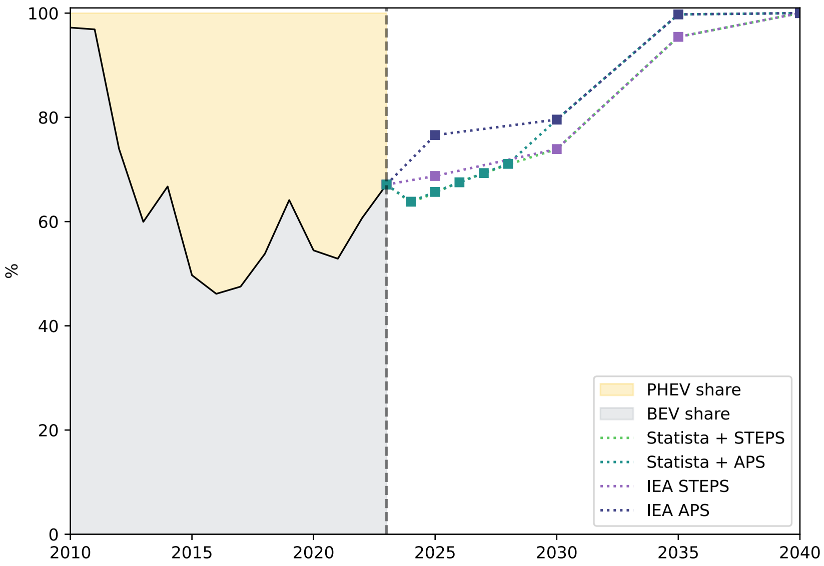


Figure A.1. BEV/PHEV market shares. Source: [191, 192]

A.1.2. Battery capacity

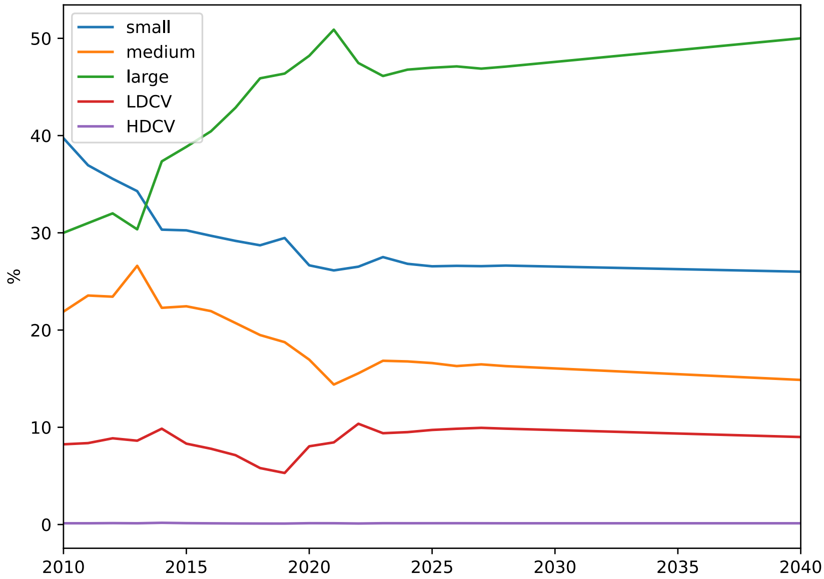


Figure A.2. Historical and projected car segmentation. LDCV = Light-duty commercial vehicles (pick-up + minivan). HDCV = Heavy-duty commercial vehicles (Full-Size Vans). Source: [192] and author's assumption after 2028.

A.1.3. Material intensity

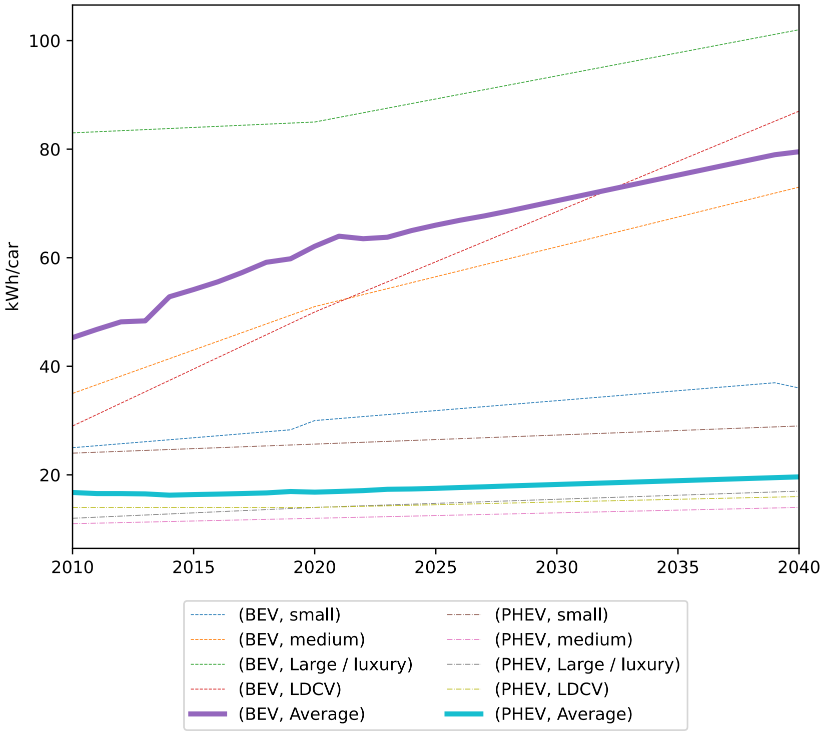


Figure A.3. Battery capacity by car segment. LDCV = Light-duty commercial vehicles (pick-up + minivan). HDCV = Heavy-duty commercial vehicles (Full-Size Vans). Source: [45, 55, 131]

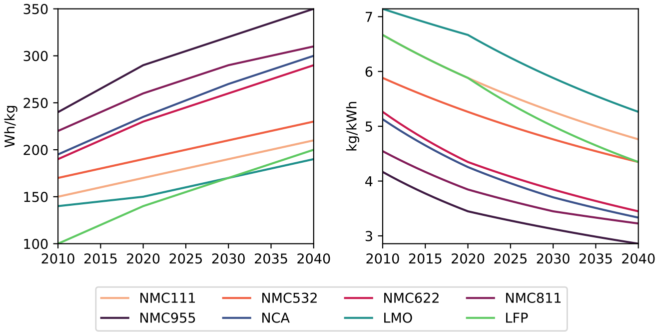


Figure A.4. Specific energy (left) and material intensities (right) of Lithium-ion batteriess (LIBs), based on [7, 37, 45, 141, 196, 242]

A.2. EV sales

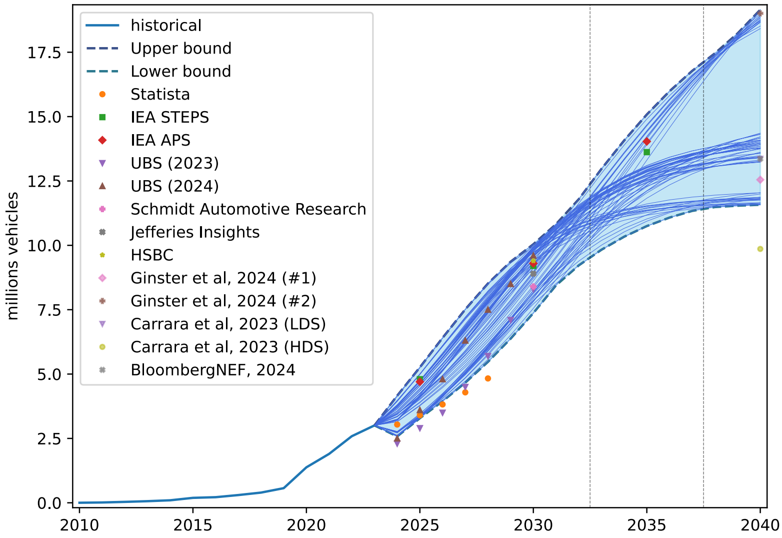


Figure A.5. Bass diffusion regression models used to forecast Electric Vehicle (EV) sales. EVs are expressed in vehicle units.

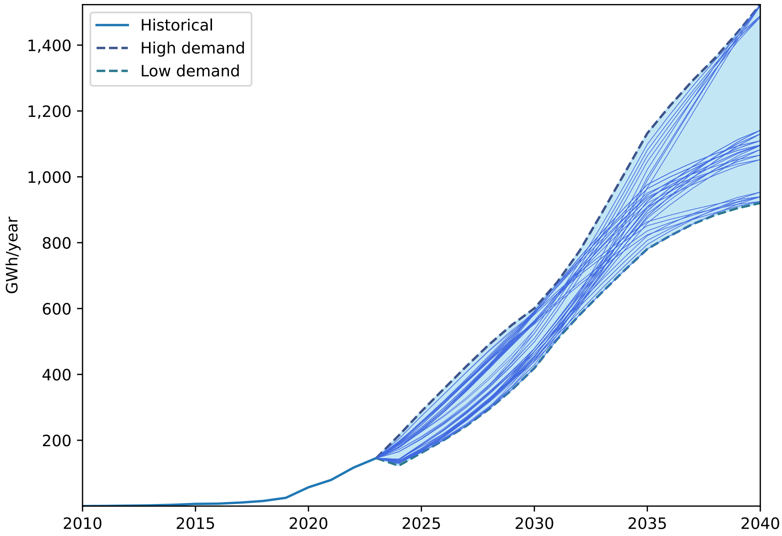


Figure A.6. Bass diffusion regression models used to forecast EV sales. EVs are expressed in energy units

A.3. Battery demand

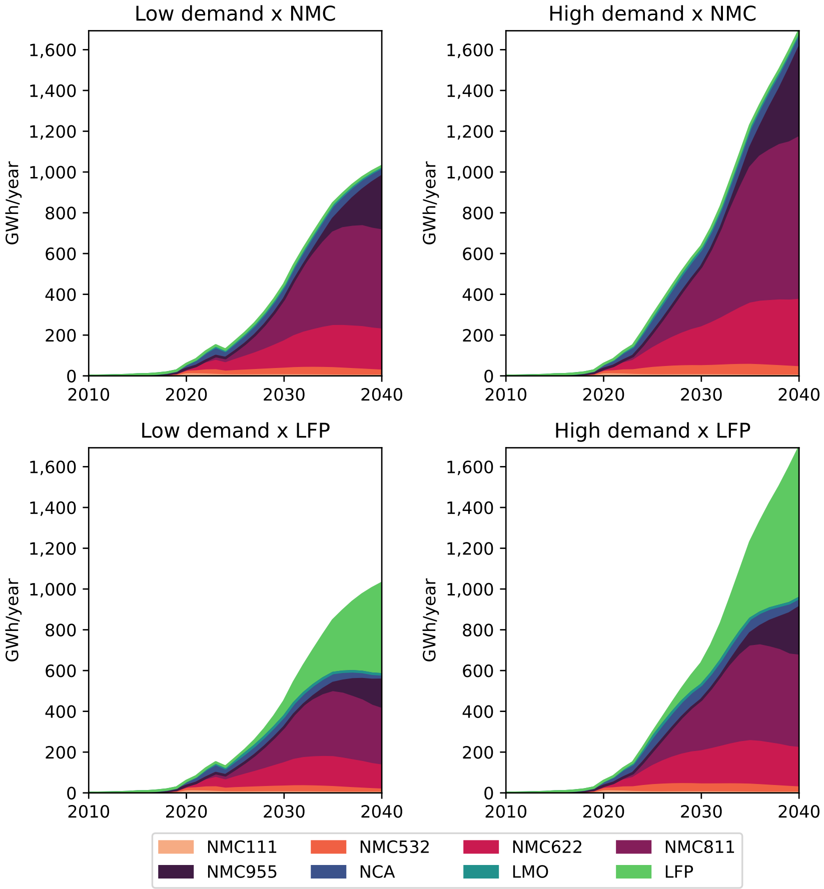


Figure A.7. Demand for EV batteries (GWh).

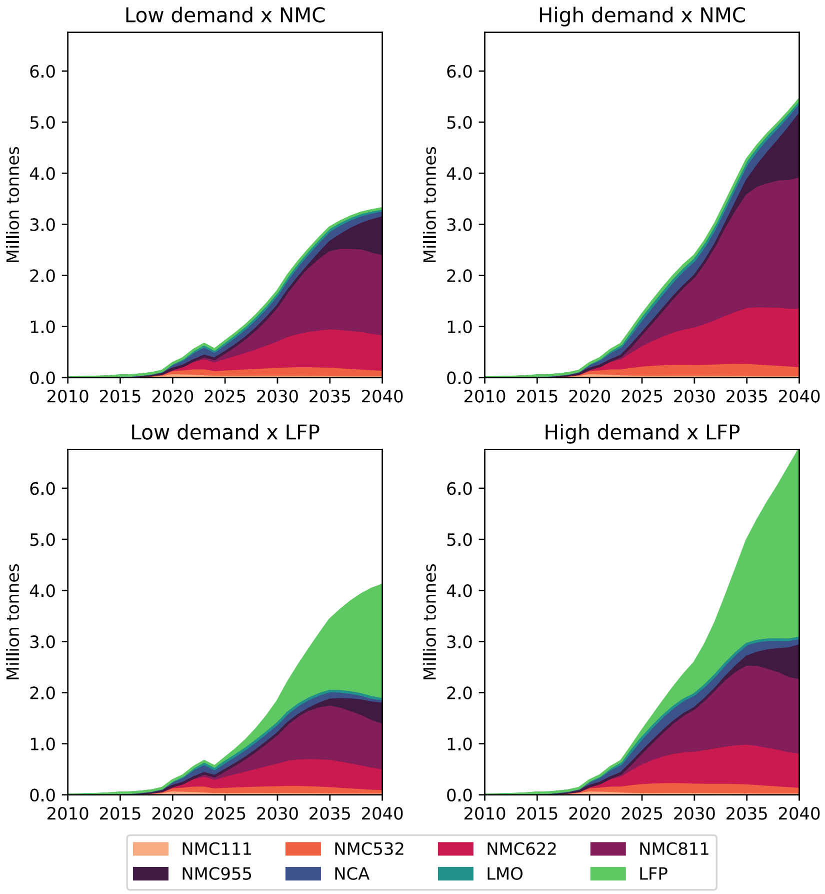


Figure A.8. Demand for EV batteries (Mt).

A.4. EOL Battery

A.4.1. Short battery lifetime

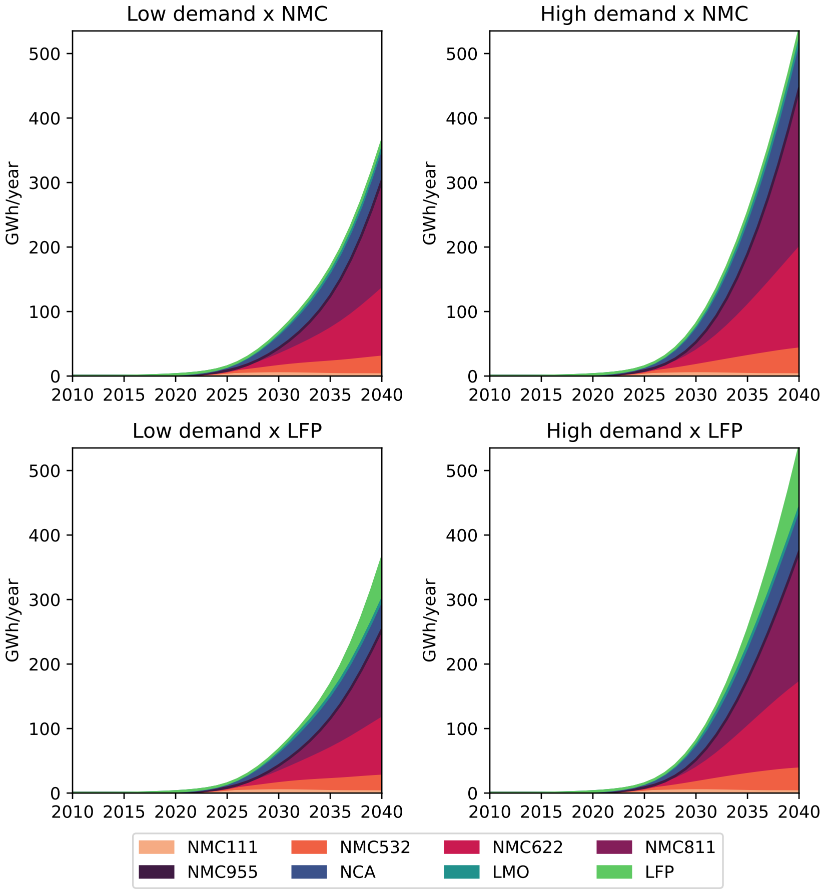


Figure A.9. Batteries reaching End-of-Life in case of short battery lifetime (GWh).

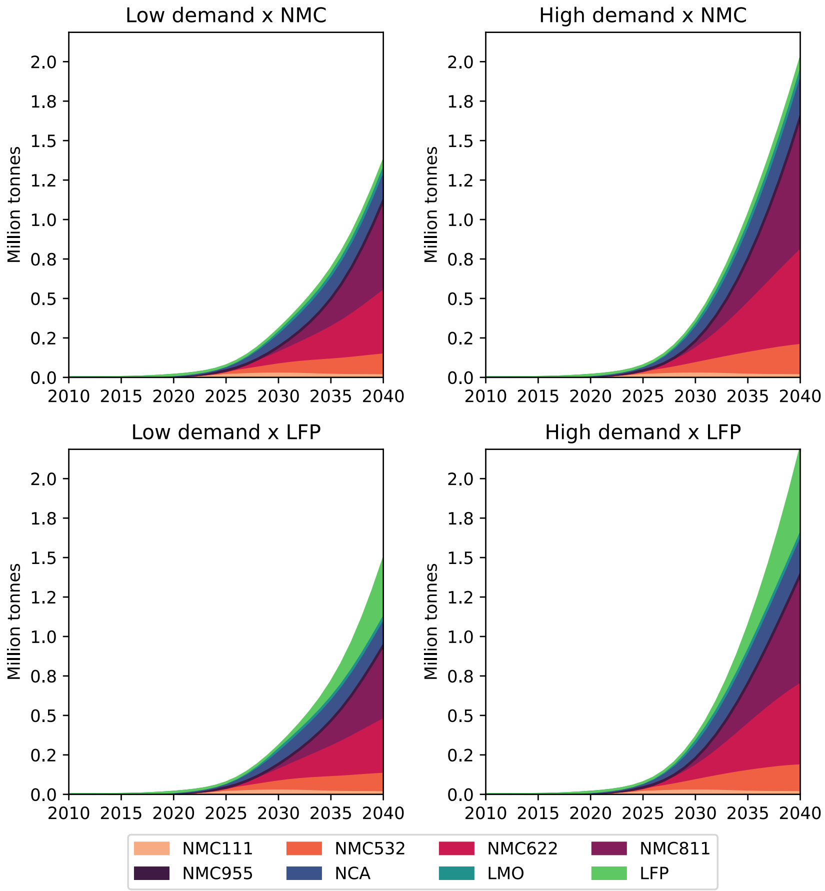


Figure A.10. Batteries reaching End-of-Life in case of short battery lifetime (Mt).

A.4.2. Long battery lifetime

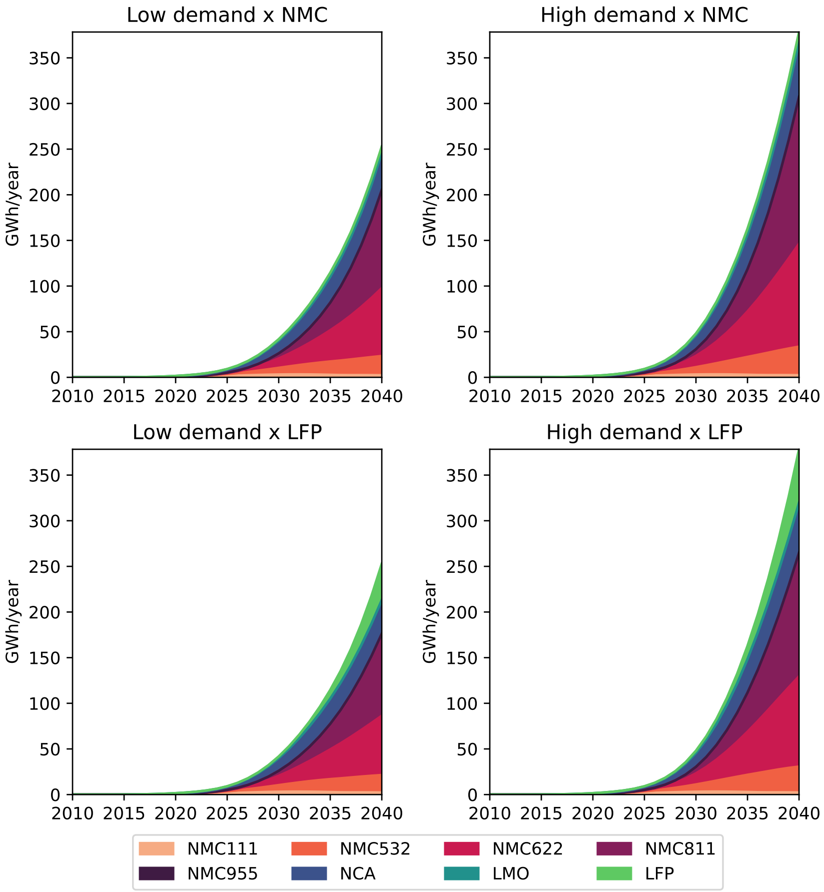


Figure A.11. Batteries reaching End-of-Life in case of long battery lifetime (GWh).

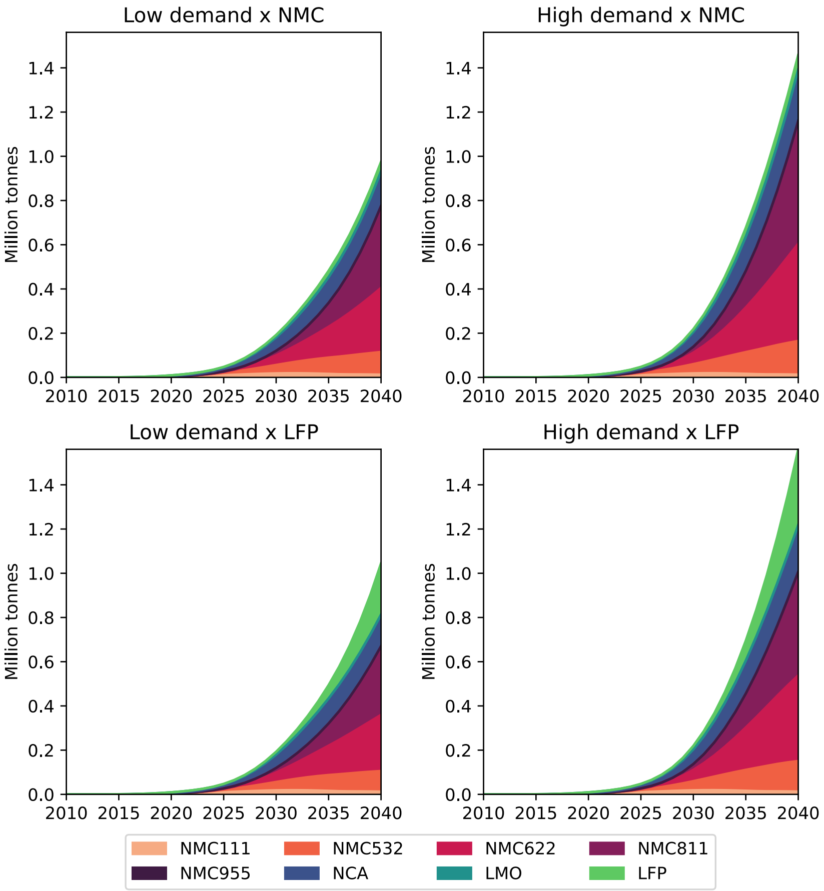


Figure A.12. Batteries reaching End-of-Life in case of long battery lifetime (Mt).

B

Recycling supply chain

B.1. EU recycling capacities

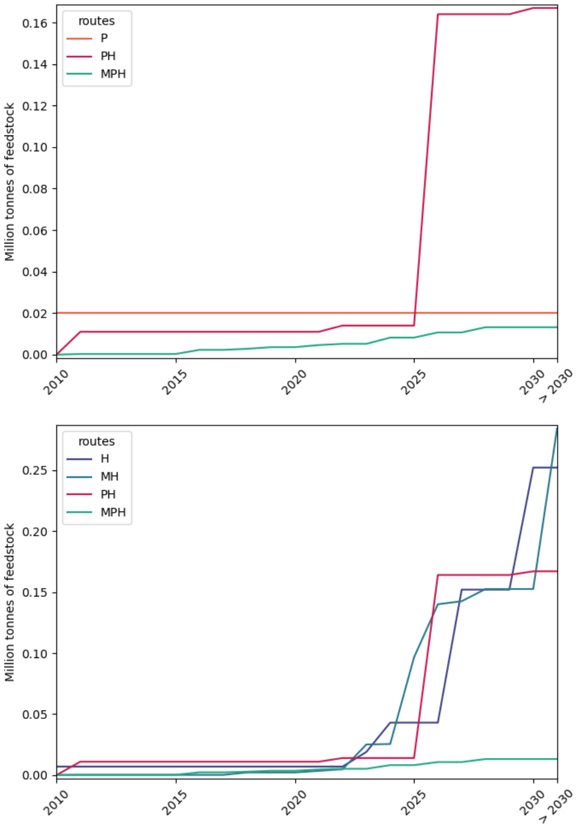
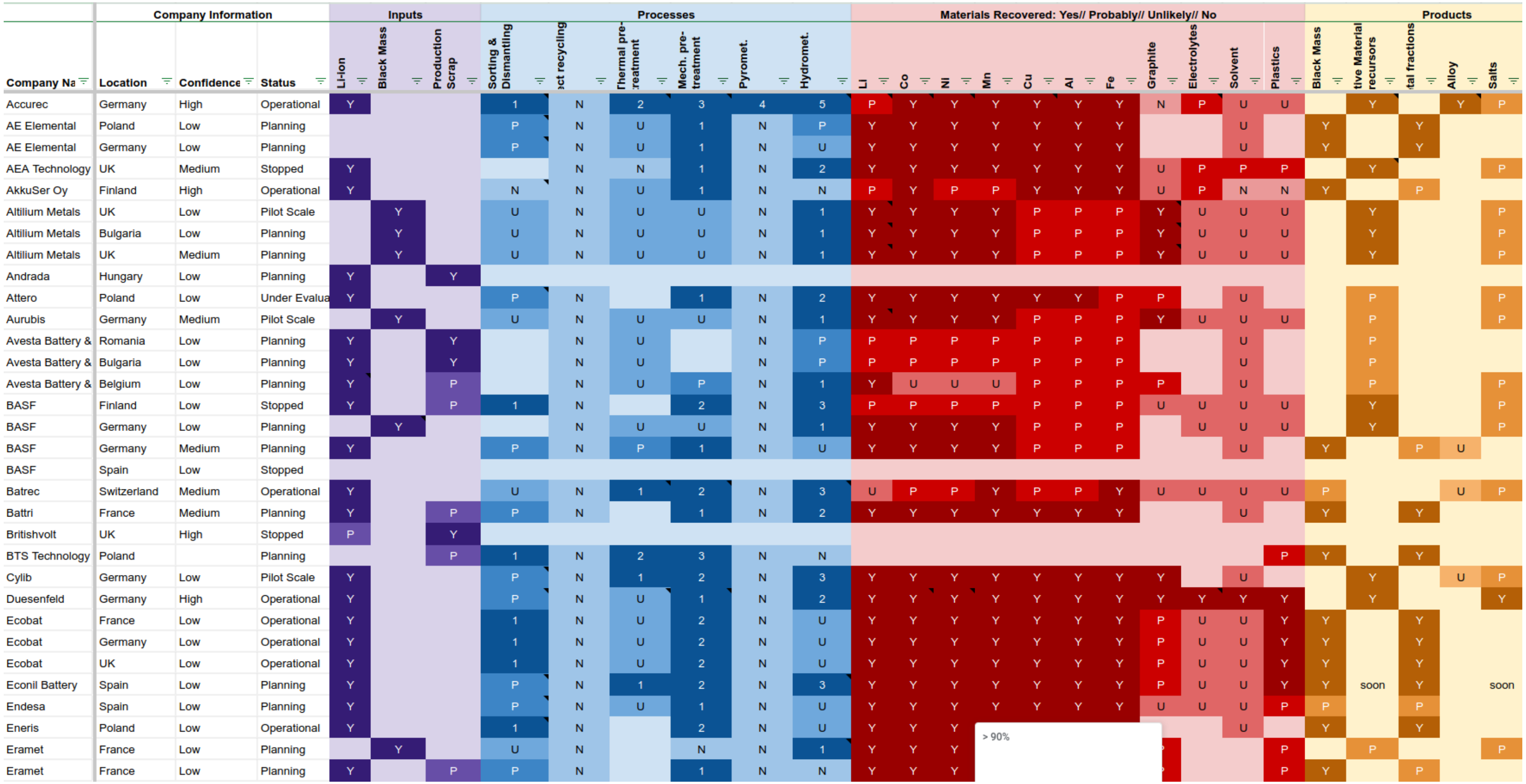


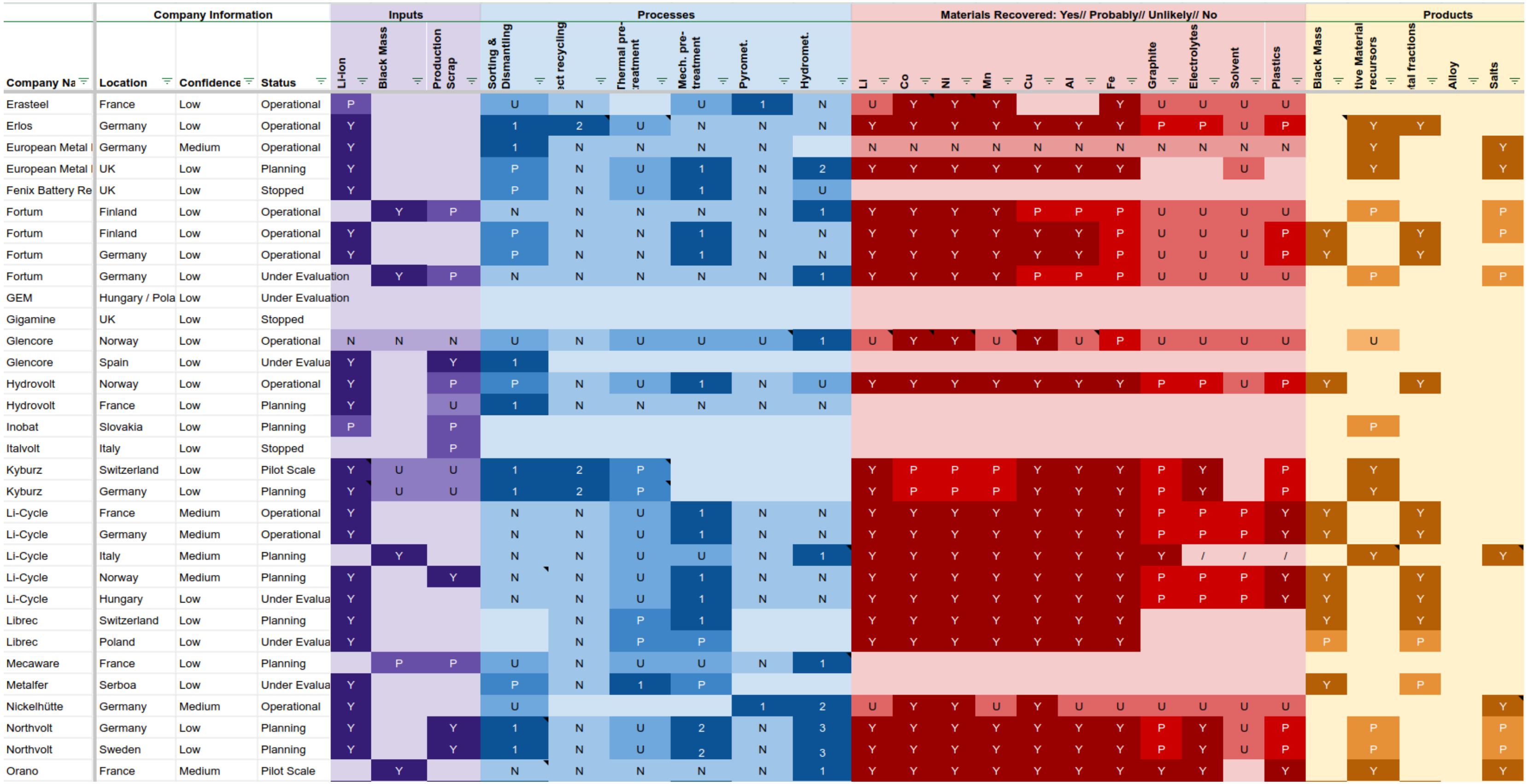
Figure B.1. EU capacity for pyrometallurgy (top) and hydrometallurgy (bottom). Capacities are in million tonnes of feedstock that the plants can process. Capacities can not be directly added since they do not not all accept the same feedstock, which can be either spent batteries or black mass (M = mechanical, P = pyrometallurgy, H = hydrometallurgy).



Company Information				Inputs			Processes						Materials Recovered: Yes// Probably// Unlikely// No											Products				
Company Na	Location	Confidence	Status	Li-Ion	Black Mass	Production Scrap	Sorting & Dismantling	ct recycling	Thermal pre- reatment	Mech. pre- treatment	Pyromet.	Hydromet.	Li	Co	Ni	Mn	Cu	Al	Fe	Graphite	Electrolytes	Solvent	Plastics	Black Mass	tive Material recursors	tal fractions	Alloy	Salts
Accurec	Germany	High	Operational	Y			1	N	2	3	4	5	P	Y	Y	Y	Y	Y	Y	N	P	U	U		Y		Y	P
AE Elemental	Poland	Low	Planning				P	N	U	1	N	P	Y	Y	Y	Y	Y	Y	Y			U		Y		Y		
AE Elemental	Germany	Low	Planning				P	N	U	1	N	U	Y	Y	Y	Y	Y	Y	Y			U		Y		Y		
AEA Technology	UK	Medium	Stopped	Y				N	N	1	N	2	Y	Y	Y	Y	Y	Y	Y	U	P	P	P		Y			P
AkkuSer Oy	Finland	High	Operational	Y			N	N	U	1	N	N	P	Y	P	P	Y	Y	Y	U	P	N	N	Y		P		
Altilium Metals	UK	Low	Pilot Scale		Y		U	N	U	U	N	1	Y	Y	Y	Y	P	P	P	Y	U	U	U		Y			P
Altilium Metals	Bulgaria	Low	Planning		Y		U	N	U	U	N	1	Y	Y	Y	Y	P	P	P	Y	U	U	U		Y			P
Altilium Metals	UK	Medium	Planning		Y		U	N	U	U	N	1	Y	Y	Y	Y	P	P	P	Y	U	U	U		Y			P
Andrada	Hungary	Low	Planning	Y		Y																						
Attero	Poland	Low	Under Evalua	Y			P	N		1	N	2	Y	Y	Y	Y	Y	Y	P	P		U			P			P
Aurubis	Germany	Medium	Pilot Scale		Y		U	N	U	U	N	1	Y	Y	Y	Y	P	P	P	Y	U	U	U		P			P
Avesta Battery &	Romania	Low	Planning	Y		Y		N	U		N	P	P	P	P	P	P	P	P			U			P			
Avesta Battery &	Bulgaria	Low	Planning	Y		Y		N	U		N	P	P	P	P	P	P	P	P			U			P			
Avesta Battery &	Belgium	Low	Planning	Y		P		N	U	P	N	1	Y	U	U	U	P	P	P	P		U			P			P
BASF	Finland	Low	Stopped	Y		P	1	N		2	N	3	P	P	P	P	P	P	P	U	U	U	U		Y			P
BASF	Germany	Low	Planning		Y			N	U	U	N	1	Y	Y	Y	Y	P	P	P		U	U	U		Y			P
BASF	Germany	Medium	Planning	Y			P	N	P	1	N	U	Y	Y	Y	Y	P	P	P			U		Y		P	U	
BASF	Spain	Low	Stopped																									
Batrec	Switzerland	Medium	Operational	Y			U	N	1	2	N	3	U	P	P	Y	P	P	Y	U	U	U	U	P			U	P
Battri	France	Medium	Planning	Y		P	P	N		1	N	2	Y	Y	Y	Y	Y	Y	Y			U		Y		Y		
Britishvolt	UK	High	Stopped	P		Y																						
BTS Technology	Poland		Planning			P	1	N	2	3	N	N											P	Y		Y		
Cylib	Germany	Low	Pilot Scale	Y			P	N	1	2	N	3	Y	Y	Y	Y	Y	Y	Y	Y		U			Y		U	P
Duesenfeld	Germany	High	Operational	Y			P	N	U	1	N	2	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y		Y			Y
Ecobat	France	Low	Operational	Y			1	N	U	2	N	U	Y	Y	Y	Y	Y	Y	Y	P	U	U	Y	Y		Y		
Ecobat	Germany	Low	Operational	Y			1	N	U	2	N	U	Y	Y	Y	Y	Y	Y	Y	P	U	U	Y	Y		Y		
Ecobat	UK	Low	Operational	Y			1	N	U	2	N	U	Y	Y	Y	Y	Y	Y	Y	P	U	U	Y	Y		Y		
Econil Battery	Spain	Low	Planning	Y			P	N	1	2	N	3	Y	Y	Y	Y	Y	Y	Y	P	U	U	Y	Y	soon	Y		soon
Endesa	Spain	Low	Planning	Y			P	N	U	1	N	U	Y	Y	Y	Y	Y	Y	Y	U	U	U	P	P		P		
Eneris	Poland	Low	Operational	Y			1	N		2	N	U	Y	Y	Y	[illegible]	[illegible]	[illegible]	[illegible]			U		Y		Y		
Eramet	France	Low	Planning		Y		U	N		N	N	1	Y	Y	Y	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]			P		P			P
Eramet	France	Low	Planning	Y		P	P	N		1	N	N	Y	Y	Y	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]			P	Y		P		

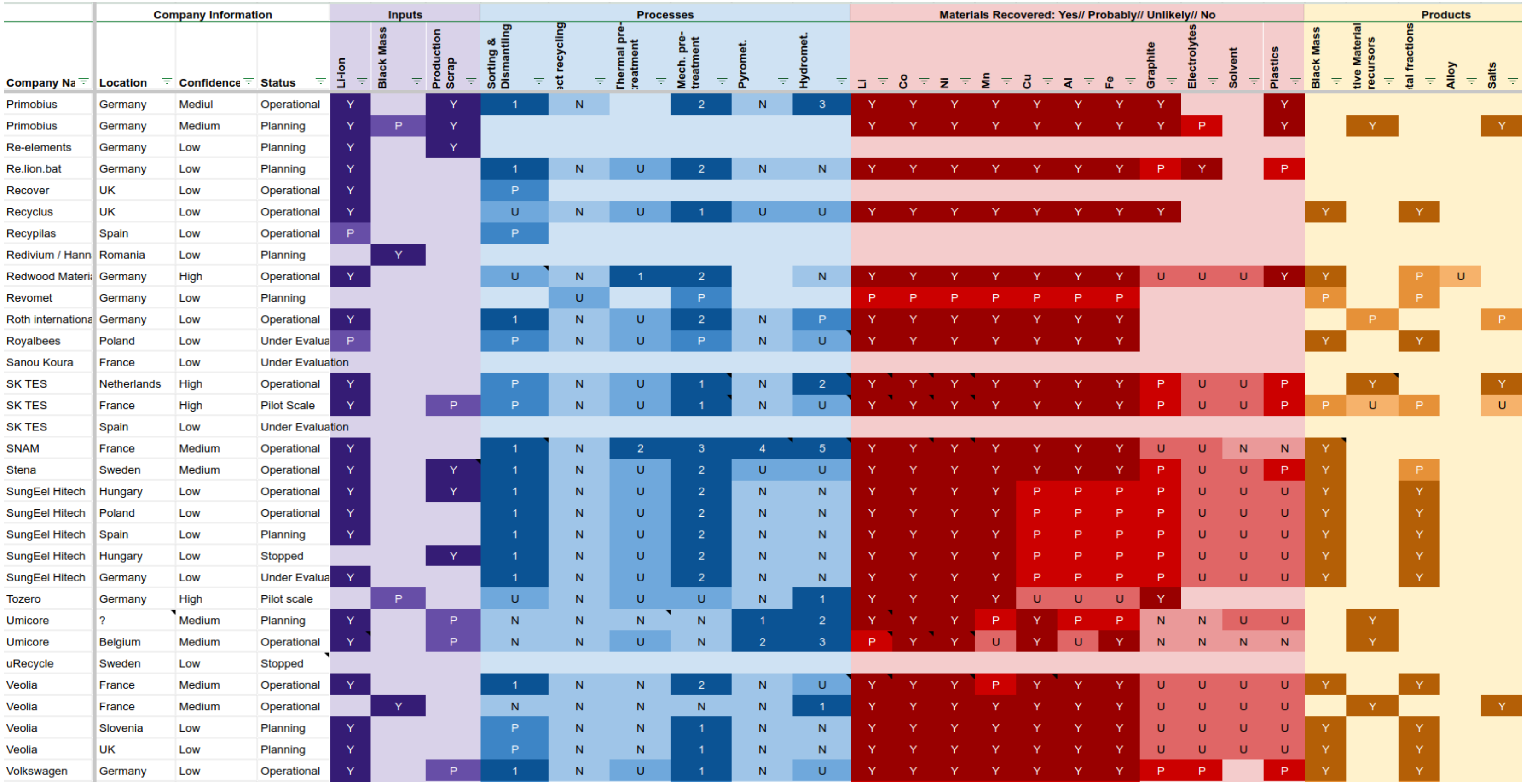
> 90%

Figure B.2. Dataset of European recycling capacities over 2010-2030 (Part 1).



	Company Information			Inputs			Processes						Materials Recovered: Yes// Probably// Unlikely// No											Products				
Company Na	Location	Confidence	Status	Li-Ion	Black Mass	Production Scrap	Sorting & Dismantling	ect recycling	Thermal pre-reatment	Mech. pre-treatment	Pyromet.	Hydromet.	Li	Co	Ni	Mn	Cu	Al	Fe	Graphite	Electrolytes	Solvent	Plastics	Black Mass	tive Material recursors	tal fractions	Alloy	Salts
Erasteel	France	Low	Operational	P			U	N		U	1	N	U	Y	Y	Y			Y	U	U	U	U					
Erlos	Germany	Low	Operational	Y			1	2	U	N	N	N	Y	Y	Y	Y	Y	Y	Y	P	P	U	P		Y	Y		
European Metal I	Germany	Medium	Operational	Y			1	N	N	N	N		N	N	N	N	N	N	N	N	N	N	N		Y			Y
European Metal I	UK	Low	Planning	Y			P	N	U	1	N	2	Y	Y	Y	Y	Y	Y	Y			U			Y			Y
Fenix Battery Re	UK	Low	Stopped	Y			P	N	U	1	N	U																
Fortum	Finland	Low	Operational		Y	P	N	N	N	N	N	1	Y	Y	Y	Y	P	P	P	U	U	U	U		P			P
Fortum	Finland	Low	Operational	Y			P	N	N	1	N	N	Y	Y	Y	Y	Y	Y	P	U	U	U	P	Y		Y		P
Fortum	Germany	Low	Operational	Y			P	N	N	1	N	N	Y	Y	Y	Y	Y	Y	P	U	U	U	P	Y		Y		
Fortum	Germany	Low	Under Evaluation		Y	P	N	N	N	N	N	1	Y	Y	Y	Y	P	P	P	U	U	U	U		P			P
GEM	Hungary / Pola	Low	Under Evaluation																									
Gigamine	UK	Low	Stopped																									
Glencore	Norway	Low	Operational	N	N	N	U	N	U	U	U	1	U	Y	Y	U	Y	U	P	U	U	U	U		U			
Glencore	Spain	Low	Under Evalua	Y		Y	1																					
Hydrovolt	Norway	Low	Operational	Y		P	P	N	U	1	N	U	Y	Y	Y	Y	Y	Y	Y	P	P	U	P	Y		Y		
Hydrovolt	France	Low	Planning	Y		U	1	N	N	N	N	N																
Inobat	Slovakia	Low	Planning	P		P																			P			
Italvolt	Italy	Low	Stopped			P																						
Kyburz	Switzerland	Low	Pilot Scale	Y	U	U	1	2	P				Y	P	P	P	Y	Y	Y	P	Y		P		Y			
Kyburz	Germany	Low	Planning	Y	U	U	1	2	P				Y	P	P	P	Y	Y	Y	P	Y		P		Y			
Li-Cycle	France	Medium	Operational	Y			N	N	U	1	N	N	Y	Y	Y	Y	Y	Y	Y	P	P	P	Y	Y		Y		
Li-Cycle	Germany	Medium	Operational	Y			N	N	U	1	N	N	Y	Y	Y	Y	Y	Y	Y	P	P	P	Y	Y		Y		
Li-Cycle	Italy	Medium	Planning		Y		N	N	U	U	N	1	Y	Y	Y	Y	Y	Y	Y	Y	/	/	/		Y			Y
Li-Cycle	Norway	Medium	Planning	Y		Y	N	N	U	1	N	N	Y	Y	Y	Y	Y	Y	Y	P	P	P	Y	Y		Y		
Li-Cycle	Hungary	Low	Under Evalua	Y			N	N	U	1	N	N	Y	Y	Y	Y	Y	Y	Y	P	P	P	Y	Y		Y		
Librec	Switzerland	Low	Planning	Y				N	P	1			Y	Y	Y	Y	Y	Y	Y					Y		Y		
Librec	Poland	Low	Under Evalua	Y				N	P	P			Y	Y	Y	Y	Y	Y	Y					P		P		
Mecaware	France	Low	Planning		P	P	U	N	U	U	N	1																
Metalfer	Serboa	Low	Under Evalua	Y			P	N	1	P														Y		P		
Nickelhütte	Germany	Medium	Operational	Y			U				1	2	U	Y	Y	U	Y	U	U	U	U	U	U					Y
Northvolt	Germany	Low	Planning	Y		Y	1	N	U	2	N	3	Y	Y	Y	Y	Y	Y	Y	P	Y	U	P		P			P
Northvolt	Sweden	Low	Planning	Y		Y	1	N	U	2	N	3	Y	Y	Y	Y	Y	Y	Y	P	Y	U	P		P			P
Orano	France	Medium	Pilot Scale		Y		N	N	N	N	N	1	Y	Y	Y	Y	Y	Y	Y	Y	Y		Y		Y			Y

Figure B.3. Dataset of European recycling capacities over 2010-2030 (Part 2).



Company Information				Inputs			Processes						Materials Recovered: Yes// Probably// Unlikely// No											Products				
Company Na	Location	Confidence	Status	Li-ion	Black Mass	Production Scrap	Sorting & Dismantling	ct recycling	Thermal pre- reatment	Mech. pre-treatment	Pyromet.	Hydromet.	Li	Co	Ni	Mn	Cu	Al	Fe	Graphite	Electrolytes	Solvent	Plastics	Black Mass	tive Material recursors	tal fractions	Alloy	Salts
Primobius	Germany	Mediul	Operational	Y		Y	1	N		2	N	3	Y	Y	Y	Y	Y	Y	Y	Y			Y					
Primobius	Germany	Medium	Planning	Y	P	Y							Y	Y	Y	Y	Y	Y	Y	Y	P		Y		Y			Y
Re-elements	Germany	Low	Planning	Y		Y																						
Re.lion.bat	Germany	Low	Planning	Y			1	N	U	2	N	N	Y	Y	Y	Y	Y	Y	Y	P	Y		P					
Recover	UK	Low	Operational	Y			P																					
Recyclus	UK	Low	Operational	Y			U	N	U	1	U	U	Y	Y	Y	Y	Y	Y	Y	Y				Y		Y		
Recypilas	Spain	Low	Operational	P			P																					
Redivium / Hann	Romania	Low	Planning		Y																							
Redwood Materi	Germany	High	Operational	Y			U	N	1	2		N	Y	Y	Y	Y	Y	Y	Y	U	U	U	Y	Y		P	U	
Revomet	Germany	Low	Planning					U		P			P	P	P	P	P	P	P					P		P		
Roth internationa	Germany	Low	Operational	Y			1	N	U	2	N	P	Y	Y	Y	Y	Y	Y	Y						P			P
Royalbees	Poland	Low	Under Evalua	P			P	N	U	P	N	U	Y	Y	Y	Y	Y	Y	Y					Y		Y		
Sanou Koura	France	Low	Under Evaluation																									
SK TES	Netherlands	High	Operational	Y			P	N	U	1	N	2	Y	Y	Y	Y	Y	Y	Y	P	U	U	P		Y			Y
SK TES	France	High	Pilot Scale	Y		P	P	N	U	1	N	U	Y	Y	Y	Y	Y	Y	Y	P	U	U	P	P	U	P		U
SK TES	Spain	Low	Under Evaluation																									
SNAM	France	Medium	Operational	Y			1	N	2	3	4	5	Y	Y	Y	Y	Y	Y	Y	U	U	N	N	Y				
Stena	Sweden	Medium	Operational	Y		Y	1	N	U	2	U	U	Y	Y	Y	Y	Y	Y	Y	P	U	U	P	Y		P		
SungEel Hitech	Hungary	Low	Operational	Y		Y	1	N	U	2	N	N	Y	Y	Y	Y	P	P	P	P	U	U	U	Y		Y		
SungEel Hitech	Poland	Low	Operational	Y			1	N	U	2	N	N	Y	Y	Y	Y	P	P	P	P	U	U	U	Y		Y		
SungEel Hitech	Spain	Low	Planning	Y			1	N	U	2	N	N	Y	Y	Y	Y	P	P	P	P	U	U	U	Y		Y		
SungEel Hitech	Hungary	Low	Stopped			Y	1	N	U	2	N	N	Y	Y	Y	Y	P	P	P	P	U	U	U	Y		Y		
SungEel Hitech	Germany	Low	Under Evalua	Y			1	N	U	2	N	N	Y	Y	Y	Y	P	P	P	P	U	U	U	Y		Y		
Tozero	Germany	High	Pilot scale		P		U	N	U	U	N	1	Y	Y	Y	Y	U	U	U	Y								
Umicore	?	Medium	Planning	Y		P	N	N	N	N	1	2	Y	Y	Y	P	Y	P	P	N	N	U	U		Y			
Umicore	Belgium	Medium	Operational	Y		P	N	N	U	N	2	3	P	Y	Y	U	Y	U	Y	N	N	N	N		Y			
uRecycle	Sweden	Low	Stopped																									
Veolia	France	Medium	Operational	Y			1	N	N	2	N	U	Y	Y	Y	P	Y	Y	Y	U	U	U	U	Y		Y		
Veolia	France	Medium	Operational		Y		N	N	N	N	N	1	Y	Y	Y	Y	Y	Y	Y	U	U	U	U		Y			Y
Veolia	Slovenia	Low	Planning	Y			P	N	N	1	N	N	Y	Y	Y	Y	Y	Y	Y	U	U	U	U	Y		Y		
Veolia	UK	Low	Planning	Y			P	N	N	1	N	N	Y	Y	Y	Y	Y	Y	Y	U	U	U	U	Y		Y		
Volkswagen	Germany	Low	Operational	Y		P	1	N	U	1	N	U	Y	Y	Y	Y	Y	Y	Y	P	P		P	Y		Y		

Figure B.4. Dataset of European recycling capacities over 2010-2030 (Part 3).

C

CRM recovery

C.1. Influence of Battery lifetime

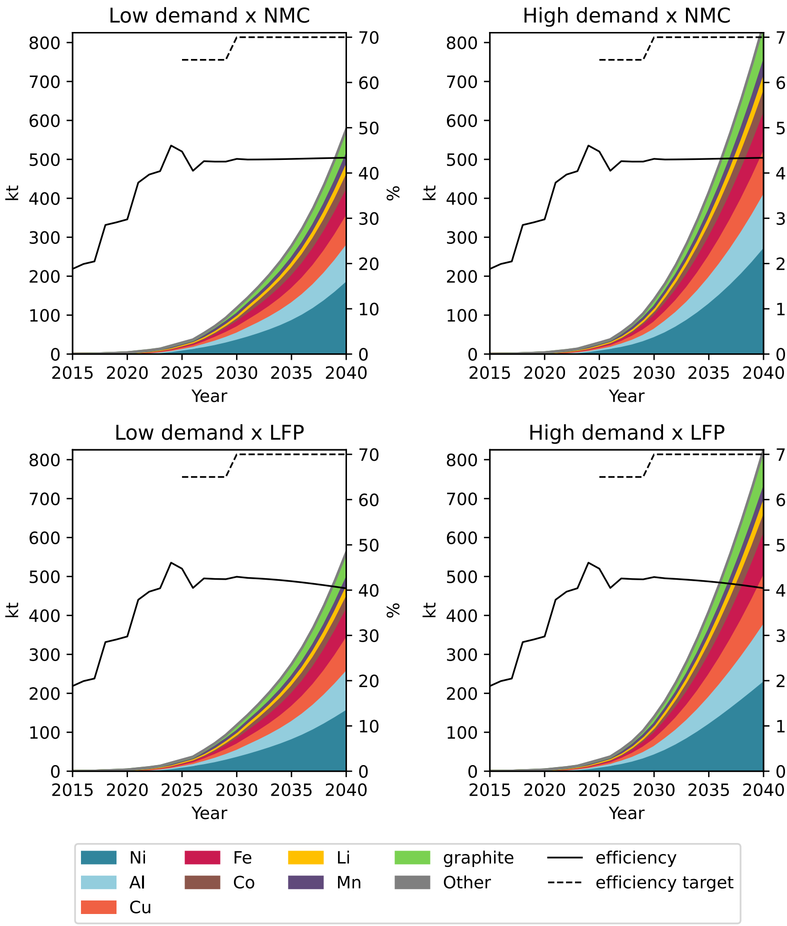


Figure C.1. Recycling efficiency (short battery lifetime, low graphite recovery).

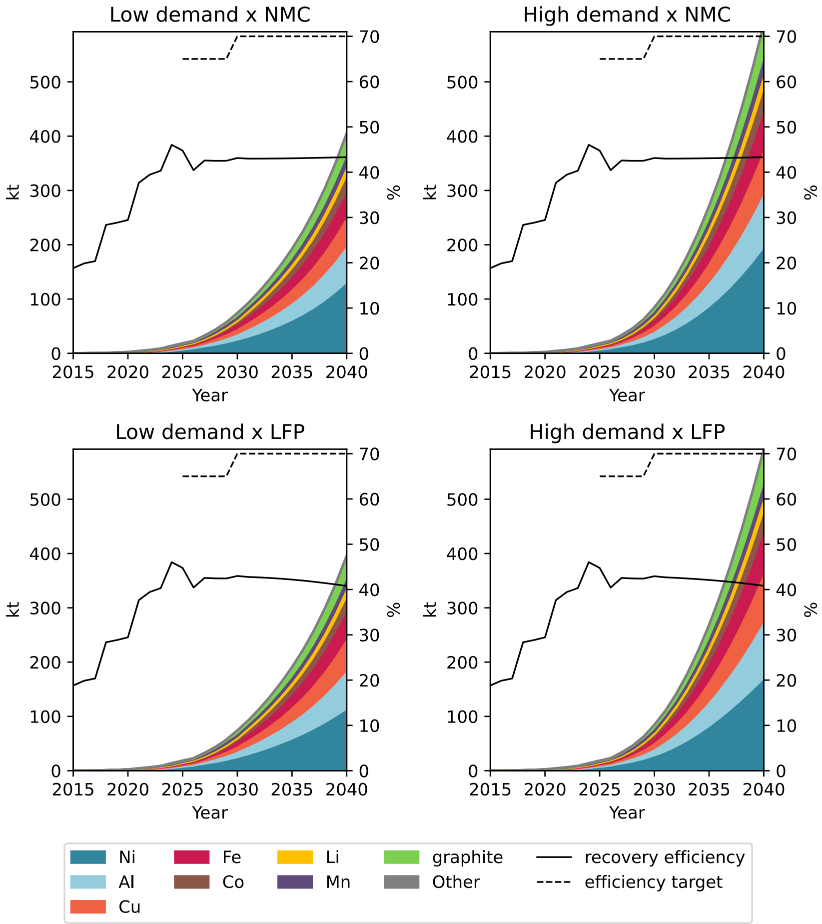


Figure C.2. Recycling efficiency (long battery lifetime, low graphite recovery).

C.2. Material leakage

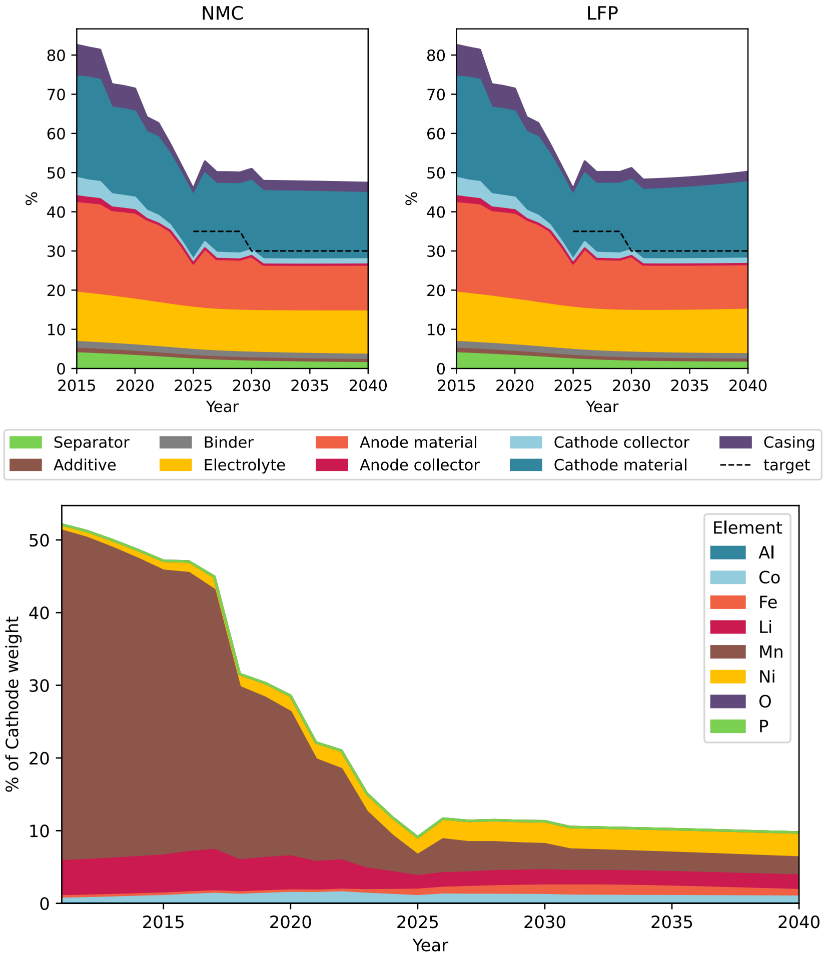


Figure C.3. Material losses at the component level (top) and at the cathode level (bottom). The efficiency rate represents an already advanced recovery of graphite. The battery directive mandates to pass below the threshold represented, which would require close to optimal recovery of both cathode and anode.

Table C.1. Black Mass Composition (%wt). Exp. = Experimental data; PMT = post-mechanical treatment (e.g. pyrolysis)

Input	Li	Ni	Co	Mn	Al	Cu	O	F	C	Fe	P	*Exp.*	*PMT*	*Source*
NMC111	4.91	11.9	11.92	11.11	1.14	0.93	22.45	0.78	34.86			×	✓	[218]
NMC111	4.54	12.8	12.7	11.98	1.92	1.3	20.94		29.26	4.56		×	×	[219]
NMC422	5.14	17.39	8.63	8.14	1.92	1.3	23.69		29.25	4.56		×	×	[219]
NMC532	4.98	18.1	7.27	10.16	1.16	0.94	21.16	0.8	35.44			×	✓	[218]
NMC532	4.53	19.17	7.61	10.77	1.92	1.3	20.9		29.24	4.56		×	×	[219]
NMC622	5.01	21.85	7.31	6.82	1.17	0.95	20.29	0.8	35.8			×	✓	[218]
NMC622	4.52	22.92	7.58	7.15	1.92	1.3	20.82		29.24	4.56		×	×	[219]
NMC811	5.10	29.61	3.72	3.46	1.2	0.97	18.61	0.82	36.52			×	✓	[218]
NMC811	4.50	30.43	3.77	3.56	1.92	1.3	20.74		29.23	4.56		×	×	[219]
NMC955	4.49	34.17	1.88	1.78	1.92	1.3	20.7		29.22	4.56		×	×	[219]
NMC	3.18	8.31	2.37	23.89	1.89	2.21	31.82		26.04	0.29		✓	×	[243]
NMC	3.90	5.10	17.50	3.00	1.60	3.90			36	1.60		✓	✓	[244]
NMC	2.60	12.5	5.4	10.9	4.8	3.1			42.1	2.30		✓	✓	[244]
NCA	2.39	20.2	20.04		11.21	1.3	11.01		29.29	4.57		×	×	[219]
LMO	2.42			38.29	1.92	1.29	22.3		29.22	4.56		×	×	[219]
(LCO ?)	4.99		38.99		2.82	2.19			25.12	0.02		-	-	[245]
-	3.69	11.5	11.7	8.91	2.1	0.88		4.1	33.9		0.44	✓	✓	[169]
mix (NMC rich)	3.95	9.61	9.4	9.12	3	4.25	31.42		29.13	0.12		✓	-	[246]
mix	3.5–4	11–26	3–33	3–11	1–5	1–3	0.5–1	2–4	35	0.1–0.3		-	-	[102]
mix (no LFP)	3.5–4	11–26	3–33	3–11	1–5	1–5	0.5–1	2–4	30	0.1–0.5	0.5–1	-	-	[247]

D

Multilayer MFA

D.1. System determinism

In the context of this work, we assume that the system is both consistent and uniquely determined, meaning it has exactly one solution, and we can assume the technology matrix $\mathbf{A}$ to be square. Various optimization techniques have been proposed to handle situations where there are either more equations than unknowns (over-determined systems) [211, 248–255] or more unknowns than equations (under-determined systems) [256–259]. However, adapting these methods to multi-layer MFA is beyond the scope of this study.

D.2. Model interface

To solve the system's equation (3.1), specifying initial conditions in addition to the Transfer Coefficients (TCs) is necessary. These initial conditions include (i) the total amount of products collected for waste processing and (ii) the mass composition of those products. Excel files are the simplest format for providing these inputs, and they can be easily structured to map to the technological matrix $\mathbf{A}$. Excel tables can also serve as an interface for the framework and enable users with limited programming or mathematical background to develop their own models. Consequently, the format of the tables should be generic and straightforward enough to not hinder the data collection process, while also being adaptable to the specific compositional structures of the system under study.

The figures D.1 to D.3 illustrate the format that was selected. To simplify the data collection process, the formats are different for each dataset. The inflow composition explicitly requires the user to specify each layer (product, component, material, element). In contrast, the data format for mass inflow (figure D.1) only requires the upper level (typically the product layer), and the data format for TCs (figure D.3) only needs the flow IDs and a single layer. This approach aims to simplify the data collection process.

Flow ID	Layer 1	Mass (kt)
F_{in}	P_1	2.5
F_{in}	P_2	1.7

Figure D.1. Data format for mass inflow.

Flow ID	Layer 1	Layer 2	Layer 3	Layer 4	Share
F_{in}	P_1	C_1			0.4
F_{in}	P_1	C_1	M_1		0.3
F_{in}	P_1	C_1	M_1	E_1	0.25

Figure D.2. Data format for inflow composition. The first row indicates that component $C1$ makes up 40% of the mass of product $P1$. The second row shows that material $M1$ represents 30% of the mass of component $C1$, which is embedded in product $P1$. Similarly, the last row states that element $E1$ represents 25% of the mass of material $M1$ within component $C1$ in product $P1$.

Process	Inflow		Outflow		TC
T_1	F_6	P_1	F_3	P_1	0.5
T_1	F_6	M_2	F_3	E_3	0.7
T_3	F_5	E_1	F_8	E_1	0.9

Figure D.3. Data format for the TCs. The first row indicates that 50% of product P_1 is recovered through process T_1. The second row illustrates a disassembly process where 70% of the element E_3 embedded in M_2 is recovered. The last row means that 90% of $E1$ present in F_5 is recovered via process T_3, regardless of what it was embedded in.

D.3. Algorithm

The algorithm consists of three steps. The first one is to restructure the Excel tables to ensure every layer is represented. This restructuring is straightforward for the mass inflow data, which is used to define the $\mathbf{y}$ vector in the equation $(\mathbf{I} - \mathbf{A})\mathbf{x} = \mathbf{y}$ (figure D.4), and for the TCs, where the corresponding rows and columns in the technology matrix can be easily derived (figure D.6). For the composition data, it can be noted that mathematically, expressing the mass of a sublayer (e.g., a component) based on the layer in which it is embedded (e.g., the product) and the corresponding composition fraction is equivalent to using the previously discussed TC formulation. This means that the technology matrix $\mathbf{A}$ can also incorporate these composition fractions (figure D.5).

Flow ID	Layer 1	Mass (kt)
F_{in}	P_1	2.5
F_{in}	P_2	1.7

$\Downarrow$

Row index					Mass (kt)
F_{in}	P_1	$\varnothing$	$\varnothing$	$\varnothing$	2.5
F_{in}	P_1	$\varnothing$	$\varnothing$	$\varnothing$	1.7

Figure D.4. Restructuring the Excel data on mass inflows to ensure all layers are represented. Row indexes refers to the indexes of the $\mathbf{y}$ vector.

Flow ID	Layer 1	Layer 2	Layer 3	Layer 4	Share
F_{in}	P_1	C_1			0.4
F_{in}	P_1	C_1	M_1		0.3
F_{in}	P_1	C_1	M_1	E_1	0.25

$\Downarrow$

Column index					Row index					share
F_{in}	P_1	$\varnothing$	$\varnothing$	$\varnothing$	F_{in}	P_1	C_1	$\varnothing$	$\varnothing$	0.4
F_{in}	P_1	C_1	$\varnothing$	$\varnothing$	F_{in}	P_1	C_1	M_1	$\varnothing$	0.3
F_{in}	P_1	C_1	M_1	$\varnothing$	F_{in}	P_1	C_1	M_1	E_1	0.2

Figure D.5. Restructuring Excel data on compositions. Restructuring follows from the observation that mass$([F_{in}, P_1, C_1]) = 0.4 \cdot$ mass$([F_{in}, P_1])$ and similarly for other rows. Row and column indexes refer to the indexes of the $\mathbf{A}$ matrix.

The algorithm's second step involves inferring all possible combinations of products, components, materials, and elements that a row from the TC table should apply to. The underlying assumption is that users will specify TCs for the most relevant layer rather than for every possible combination of embedded sub-layers. For instance, if a component has a recovery rate of 50%, it is assumed that all materials and elements within that component are recovered at the same rate. Similarly, users might specify a default recovery value for a chemical element without knowing exactly which component or product it will be part of when it reaches this particular process. To accommodate such cases, the empty symbol is used,

Inflow		Outflow		TC
F_4	C_2	F_7	M_1	0.18

$\Downarrow$

Column index					Row index					TC
F_4		C_2			F_7			M_1		0.18

$\Downarrow$

Column index					Row index					TC
F_4		C_2	M_1		F_7		C_2	M_1		0.18

Figure D.6. Restructuring Excel data on TCs to ensure all layers are represented, with placeholders maintained in anticipation of step 2. Row and column indexes refer to the indexes of the $\mathbf{A}$ matrix.

ensuring that every combination of flow, product, component, material, and element can be represented as an ordered sequence of length 5, even when dealing with aggregated entities or when an upper layer becomes irrelevant (see figure D.7). This inference process is achieved by generating the Cartesian product, as shown in figure D.8. Unlike for TCs, this step is not required for composition data, as it is assumed that users will provide complete composition information for the system, which cannot be inferred otherwise.

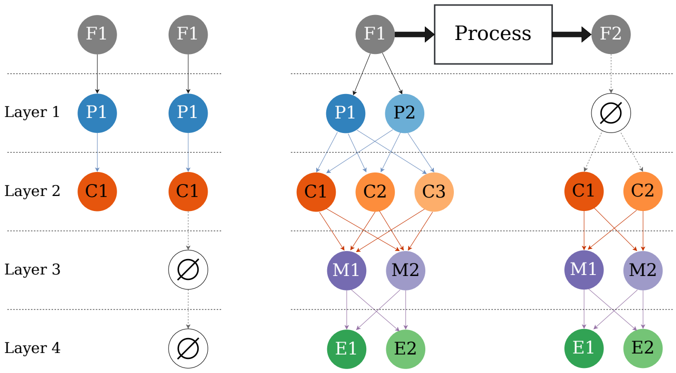


Figure D.7. The symbol $\varnothing$ is used to maintain a fixed length ordered sequence (F, P, C, M, E) when considering an aggregated entity (left) or when an upper level is no longer present (right).

The third step involves populating the vector $\mathbf{y}$ and the technology matrix $\mathbf{A}$ with the newly obtained data. However, for the technology matrix,

Column index					Row index					TC
F_4		C_2	M_1		F_7		C_2	M_1		0.18

$$\Downarrow$$

Column index					Row index					TC
F_4	P_1	C_2	M_1	E_1	F_7	P_1	C_2	M_1	E_1	0.18
F_4	P_1	C_2	M_1	E_2	F_7	P_1	C_2	M_1	E_2	0.18
F_4	P_1	C_2	M_1	E_3	F_7	P_1	C_2	M_1	E_3	0.18
...					...					...
F_4	P_2	C_2	M_1	E_1	F_7	P_2	C_2	M_1	E_1	0.18
F_4	P_2	C_2	M_1	E_2	F_7	P_2	C_2	M_1	E_2	0.18
F_4	P_2	C_2	M_1	E_3	F_7	P_2	C_2	M_1	E_3	0.18
...					...					...
F_4	$\varnothing$	C_2	M_1	E_1	F_7	$\varnothing$	C_2	M_1	E_1	0.18
F_4	$\varnothing$	C_2	M_1	E_2	F_7	$\varnothing$	C_2	M_1	E_2	0.18
F_4	$\varnothing$	C_2	M_1	E_3	F_7	$\varnothing$	C_2	M_1	E_3	0.18
...					...					...

Figure D.8. The second step of the algorithm implementation is to infer the TC for the missing combinations of products, components, materials, and elements.

this process may introduce potential conflicts that could produce incorrect outputs and be difficult to detect. Indeed, the expansion from the second step can lead to a sequence being assigned multiple TCs. For example if for the same process the user specify a TC α for a component C_1, and at the same time assume a TC β for material M_1, then the sequence $[C_1, M_1]$ will be associated to both α and β. This is incompatible with the assumption of a determined system, as only a single TC should be used to ensure that the technology matrix $\mathbf{A}$ is square. By default, TCs specified closer to the chemical element are considered more reliable and are used to populate the matrix. However, any conflicts can be visualised to allow the user to review and verify the matrix entries. It would also be trivial to add a column to the TCs Excel file to specify the priority (i.e., the certainty) of each coefficient

Figure D.9 illustrates the integration of mass composition and TCs within the technology matrix $\mathbf{A}$ following the three steps described above. Finally, the equation $(\mathbf{I}-\mathbf{A})\mathbf{x} = \mathbf{y}$ can be solved with a simple linear algebra solver such as the one provided by the Python's Scipy library [260].

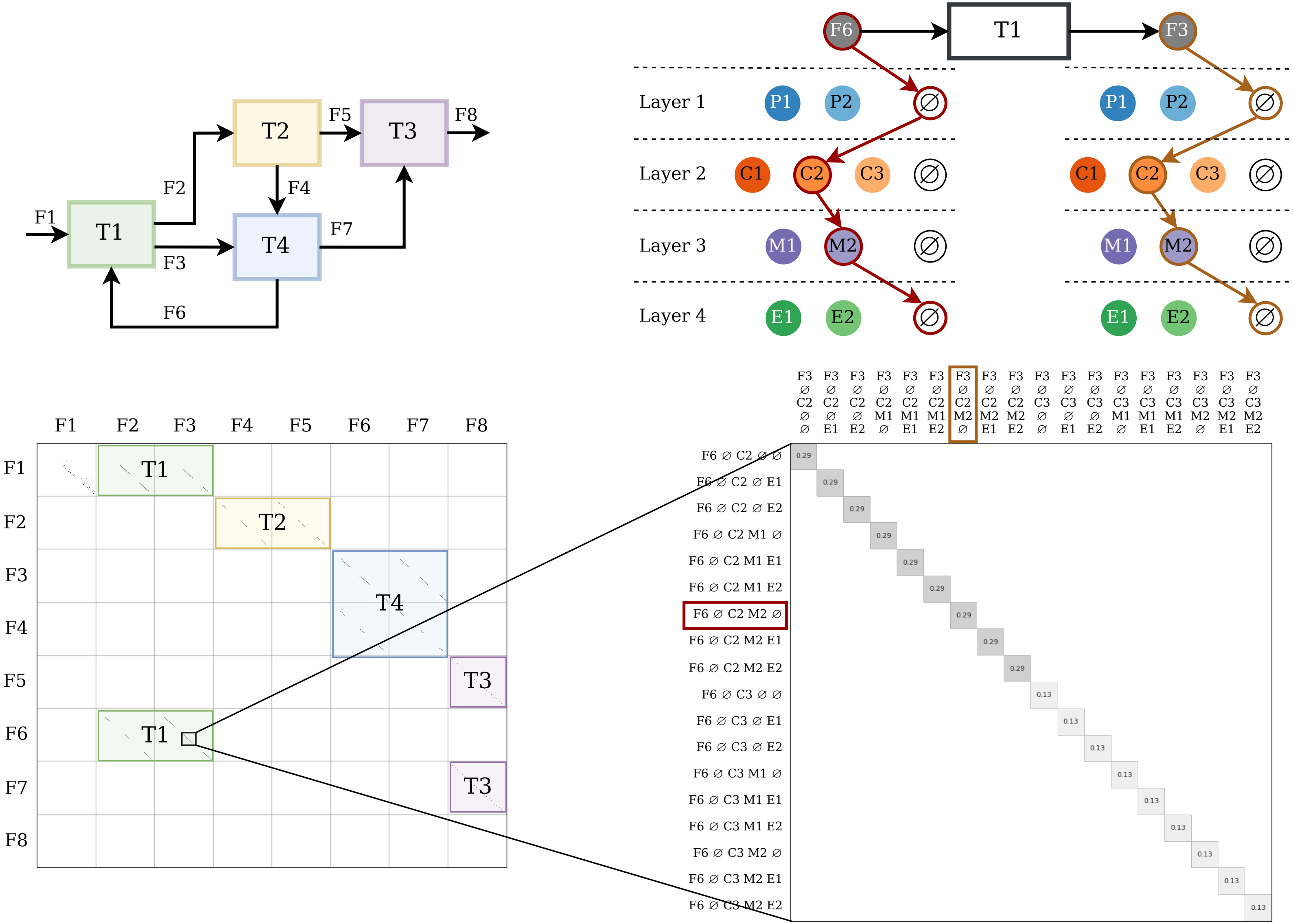


Figure D.9. A multi-layer MFA system (top) and its transposed technology matrix (bottom). The cell $(F1, F1)$ in the technology matrix represents the mass composition of the inflow $F1$.

Table D.1. Flow equations derived from the system depicted in figure D.9. By contention, inflows are positive and outflows are negative. The table can then be used to verify the mass balance of the system.

Process	F_1	F_2	F_3	F_4	F_5	F_6	F_7	F_8
T_1	1	-1	-1			1		
T_2		1		-1	-1			
T_3					1		1	-1
T_4			1	1		-1	-1	

D.4. Model output and mass balance

The output of the static multi-layer MFA reproduces the same format as the inflow composition (figure D.1). Additionally, a table with the flow equations is generated along with the solution (see table D.1 for more details). This table is then used to verify the mass balance of the system. However, strict enforcement of mass balance is not applied for three reasons: i) mass balance only applies at the chemical element layer, therefore attention to edge cases should be paid when applying mass balance to more aggregated levels (e.g., a product no longer exist after disassembly, and only the components remain); ii) some chemical elements may not be of interest to the modeler, in which case TCs might not have been provided (e.g. the amount of carbon or oxygen released by air emissions during incineration); iii) uncertainty and sensitivity analysis may be conducted to identify hotspots, and the value ranges may not necessarily add up to 100%.

D.5. Computational complexity

The algorithm's simplicity and maintainability come with a trade-off: exponential complexity in both space and time. As shown in figure D.9, most of the technology matrix is empty, making it inefficient to store it as a full square matrix. To address space complexity, the matrix is stored as a Compressed Sparse Row (CSR) matrix [261] and equation (3.1) is processed using SciPy's wrapper of the UMFPACK sparse solver [262]. Converting the square technology matrix $\mathbf{A}$ into a CSR format requires first transforming the ordered sequence $[F_m, P_n, C_o, M_p, E_q]$, which is used for indexing and retrieving TCs within the technology matrix, into a unique integer. To achieve this, we assign a unique integer to each product, component, material, and element: $[m, n, o, p, q]$. We then define a bijective mapping to establish a correspondence between such a sequence and a unique integer. Let s be the vector representing the number of variables for each layer, with d layers:

$$\mathbf{s} = [s_0, s_1, \ldots, s_{d-1}]$$

and the linear indexing coefficients $\mathbf{c} = [c_0, c_1, \ldots, c_{d-1}]$, where each coefficient c_i is given by:

$$c_i = \begin{cases} \prod_{j=i+1}^{d-1} s_j & \text{for } i < d-1 \\ 1 & \text{for } i = d-1 \end{cases}$$

Let $\alpha = [\alpha_0, \alpha_1, \ldots, \alpha_{d-1}]$ be the vector that represent the sequence $[F_m, P_n, C_o, M_p, E_q]$. Its unique scalar index k is computed as:

$$k = \sum_{i=0}^{d-1} \alpha_i \cdot c_i$$

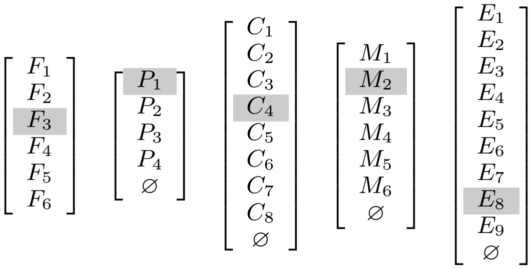


Figure D.10. A bijective mapping ensures a correspondence between a sequence and a unique integer. Here, the index of $[F_3, P_1, C_4, M_2, E_8]$ is $10\,388$. This index is then used to convert the technology matrix $\mathbf{A}$ into a CSR format.

Further optimization is necessary due to the RAM usage of the sparse solver, which often becomes the limiting factor. To mitigate this, two non-exclusive approaches can be employed, though they increase time complexity: i) Matrix Reduction: The size of the technology matrix can be reduced by recognizing that, due to the mass conservation principle, the element layers are independent of each other. This allows for the generation of a smaller technology matrix for each chemical element. The solution for each sub-system can then be computed independently and aggregated afterward. ii) Graph Partitioning: An MFA system can be viewed as a graph network that may contain cycles. By identifying these cycles and aggregating the nodes within each loop into a ”virtual” node (i.e., partitioning the graph into strongly connected components, see figure D.11), the graph can be converted into a ”virtual” directed acyclic graph (DAG). Once this DAG is obtained, a topological sort can order these ”virtual” nodes, effectively dividing the overall MFA system into sub-systems that can be processed sequentially.

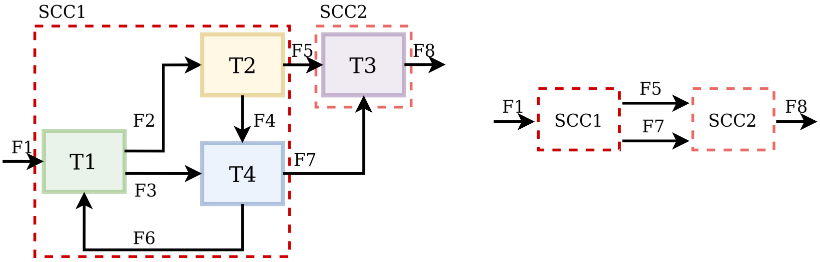


Figure D.11. Example of sub-systems division.