RECYCLING OF LITHIUM-ION BATTERIES

2nd edition, 2023

PEM | RWTH AACHEN UNIVERSITY
VDMA
Waste Treatment and Recycling Technology

The Chair of Production Engineering of E-Mobility Components (PEM) of RWTH Aachen University has been researching lithium-ion battery production for many years. The team's range of topics extends from the automotive sector to stationary applications. Through its participation in numerous national and international industrial projects with companies at all stages of the value chain and thanks to key positions in renowned research projects, PEM offers extensive expertise.

The Battery LabFactory (BLB) is a research center of the TU Braunschweig and offers a platform for the development of circular production as well as diagnostic and simulation methods for current lithium-ion batteries as well as future technologies such as solid-state batteries and lithium-sulfur batteries. BLB unites 18 professorships from three universities as well as other battery experts and brings together broad expertise along the value chain for electrochemical battery storage.

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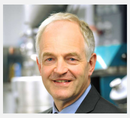

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Overall, VDMA represents more than 3,700 German and European mechanical and plant engineering companies. The Battery Production Department acts as a contact for all questions relating to battery machinery and plant engineering. The specialist department researches technology and market information, organizes customer events and roadshows, offers platforms for exchange within the industry and is in constant dialogue with research and science.

The VDMA Waste and Recycling Technology Association represents the interests of more than 100 member companies at national and European level. Its tasks also include a globally oriented trade fair policy. As a platform for informal meetings, the association promotes developments through technical and marketing-oriented working groups, particularly in the areas of recycling and processing technology as well as in the broad field of biological, mechanical and thermal processes.

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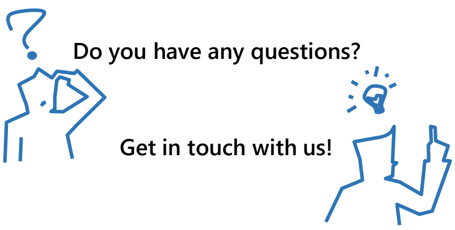

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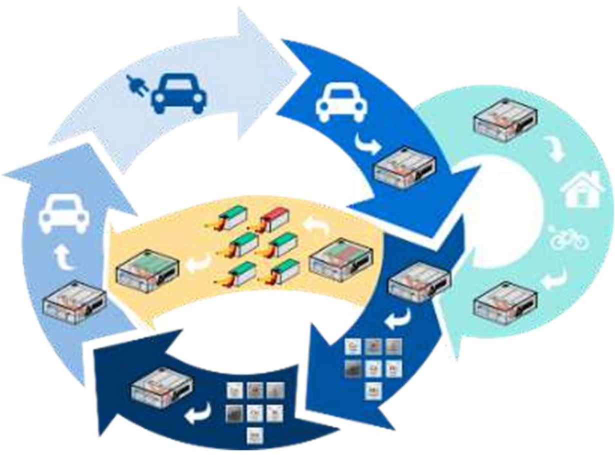
Market Development

Recycling of lithium-ion batteries (LIB)

- The **volume of lithium-ion batteries (LIB)** at their end of life (EoL) **will rise sharply in the coming years** due to the growing proportion of electric vehicles, which is why new concepts for recycling and raw material recovery must be developed.
- The **process rejects arising in battery production** will make higher recycling capacities necessary in the near future.
- In order to implement sustainable EoL concepts, all players along the value chain must address this issue – from material synthesis to battery cell, battery module and battery pack production to the utilization phase and recycling.
- **Recycling rates** for individual materials (up to 95%) are proposed by Circular Economy Initiative Deutschland, acatech (CEID)* and provided by the EU in its Battery Regulation.

Table of Content

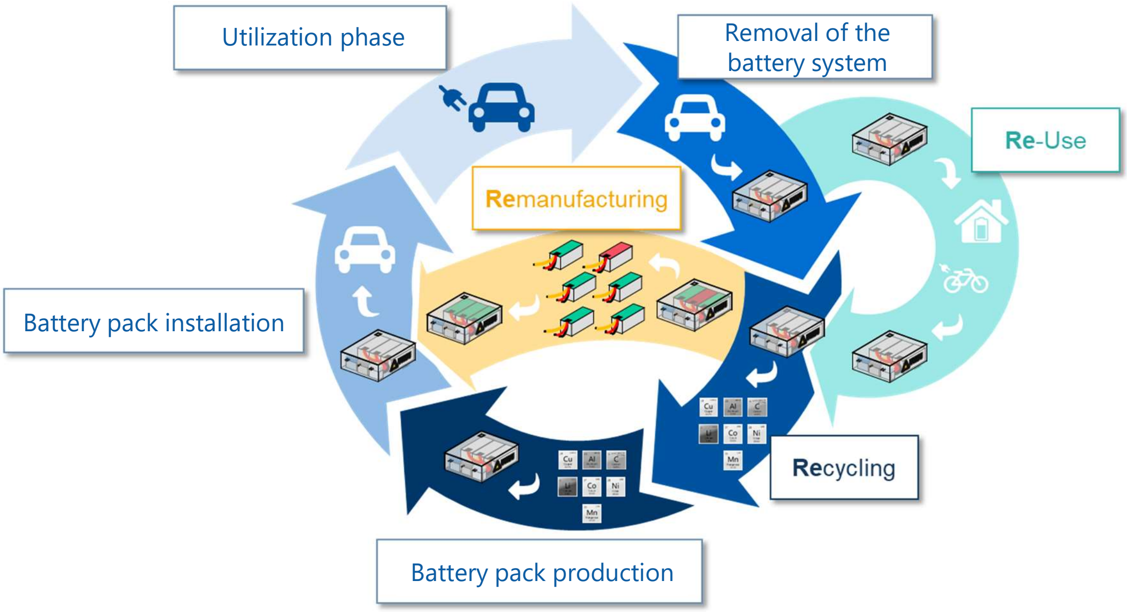
Recycling of lithium-ion batteries (LIB)



* Circular Economy Initiative Deutschland (Hrsg.): Ressourcenschonende Batteriekreisläufe – mit Circular Economy die Elektromobilität antreiben; Kwade, A., Hagelüken, C., et al., acatech/SYSTEMIQ, Munich/London 2020

End-of-Life Strategies for LIB

Fundamentals of battery recycling



Annotations

- At the end of the battery's service life, the battery pack is removed from the vehicle. Depending on the battery's condition, different end-of-life strategies are possible.
- The circular economy's main objective is to re-use materials as much as possible by closing the product life cycle and thus minimizing waste disposal and reducing pollutants as well as dependence on important primary materials.
- The classic value creation cycle can be extended through re-use (second use) and remanufacturing. The terms 're-use', 'remanufacturing' and 'recycling', which are relevant for a sustainable circular economy, can be summarized as 'Re-X'.
- Since twelve percent of Europe's total greenhouse gas emissions are caused by electric vehicle batteries in EoL status, special attention must be paid to this product life phase.
- The handling of EoL batteries by means of recycling is therefore explained below.

Re-Use

Lithium-ion batteries that still have sufficient properties (residual charging capacity, safety, internal resistance) at the end of their service life in battery electric vehicles (BEV) can still be used in stationary energy storage systems, for example.

Remanufacturing

Remanufacturing enables the re-use of batteries in BEVs by reconditioning spent batteries by exchanging or replacing damaged components.

Recycling

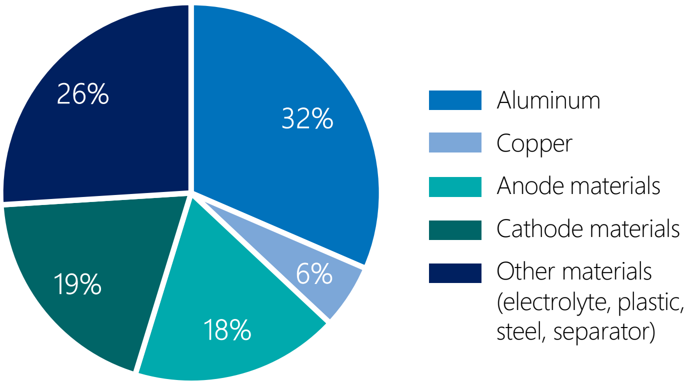
Recycling is necessary to recover raw materials from batteries through a safe method and return them to the battery production process.

Kampker et al.: International Conference on Engineering and Technology, 2018; Emilsson et al.: Lithium-Ion Vehicle Battery Production, 2019; Siret et al.: PEFCR for High Specific Energy Rechargeable Batteries, 2018; acatech et al.: Ressourcenschonende Batteriekreisläufe, 2020

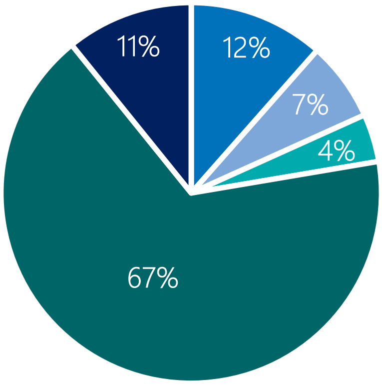
Material Composition

Fundamentals of battery recycling

Weight distribution (system level)[1]



Value distribution (system level)[1, 2]



[1] Based on NMC622 cathode chemistry

[2] Based on material prices in 2023

Annotations

- A battery system's **main components** are aluminum, copper, anode material, cathode material, and other components (electrolyte, plastic, steel, separator, etc.).
- In addition to being used as a current collector in the cathode, **aluminum** is mainly used for the housings of the cells, the modules and the pack. Graphite is used as the anode material in more than 90% of cells. The composition of the cathode material varies greatly (see next page) – however, it usually contains lithium, nickel, cobalt, and manganese.
- While **cathode materials** only account for just under 20% of the overall material weight, they are responsible for almost **70% of the entire material value** – in the case of NMC cathodes. Therefore, battery recycling focuses primarily on recovering the cathode materials.
- **Prices of individual battery materials** have been subject to strong price fluctuations for several years. This is a challenge for the predictability and profitability of recycling plants.
- As the **lithium price** has been significantly below the price for nickel and cobalt for a long time, recycling focused primarily on these two elements of the cathode and on easily recoverable materials for the housings, such as steel and aluminum.
- The **EU Battery Regulation**, which came into force in 2023, stipulates minimum recycling percentages per material, meaning that lithium, nickel, cobalt, and copper must be recycled from 2027.

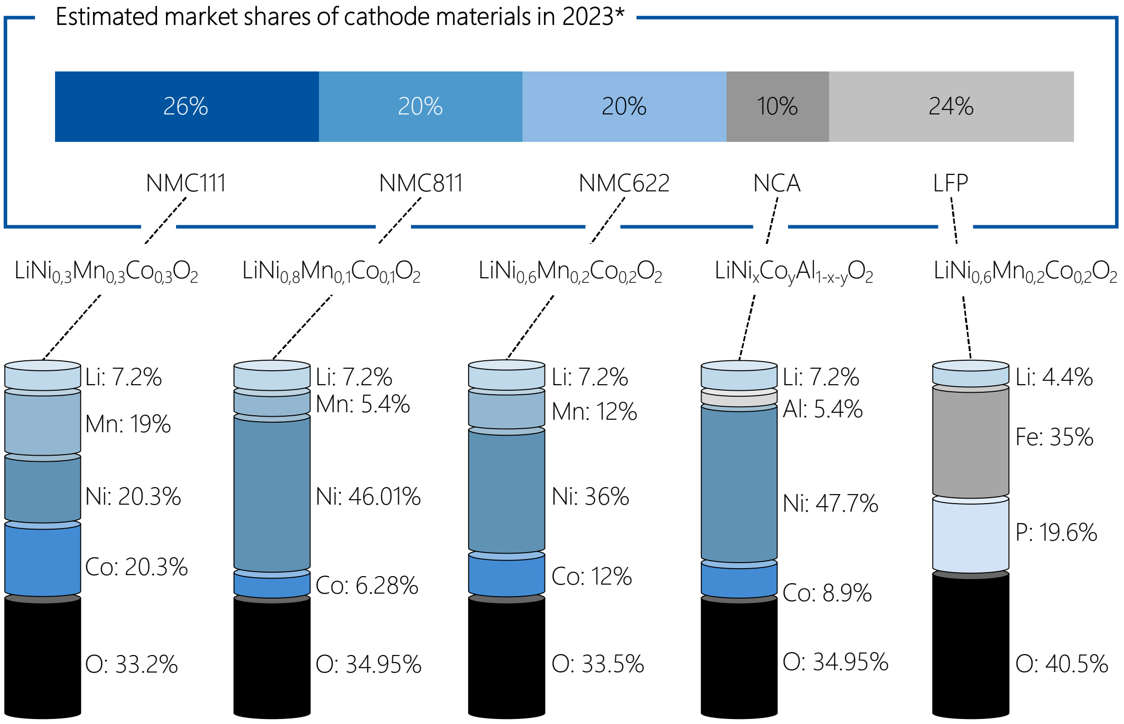
Key Takeaways

- In order to enable economically viable processes, recycling today focuses on the most valuable materials in batteries – usually the cathode materials. This is also being pushed politically, as the new EU Battery Regulation shows. Furthermore, recycling these raw materials reduces dependence on producing countries.
- The materials used in lithium iron phosphate (LFP) cell chemistry are cheaper and therefore pose a challenge to the economic viability of battery recycling.

Kampker et al.: Montageprozess eines Batteriemoduls und -packs, 2018;
VW: Batterie-Recycling: Alles zur Pilotanlage in Salzgitter, 2021

Cathode Composition

Fundamentals of battery recycling



Annotations

- The chart above shows the mass fractions of the five most important cathode active materials in 2023, which are made up of lithium, manganese, cobalt, nickel, aluminum, iron, and phosphate.
- The share of high-nickel cathode materials (NMC622 and NMC811) has increased significantly in recent years compared to NMC111.
- LFP plays an important role in China in particular, but is also gaining importance in other markets. There is also the LFMP variant in which low levels of manganese are integrated, thus increasing the cell material's energy density.
- NCA was favored by Tesla for a long time but is becoming less important as cathode material. However, due to the delayed effect on recycling, the material will have to be considered for a few more years.
- Highly fluctuating raw material prices and limited production capacities of the required elements are causing battery cell manufacturers to strive for raw material security, which can be achieved by re-cycling disused battery systems, for example.
- These fluctuations are also due to long supply chains, as raw material deposits are limited to certain countries.

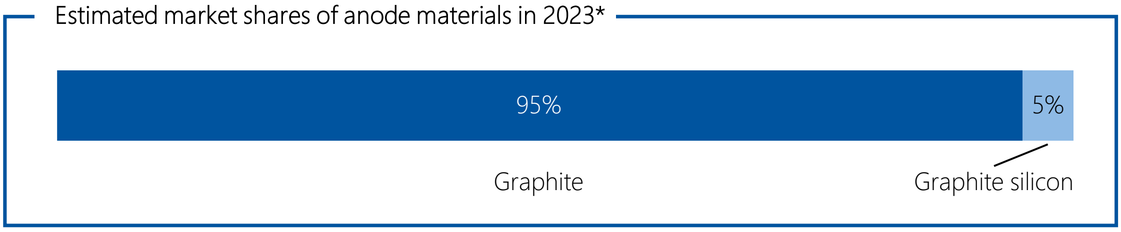
Key Takeaways

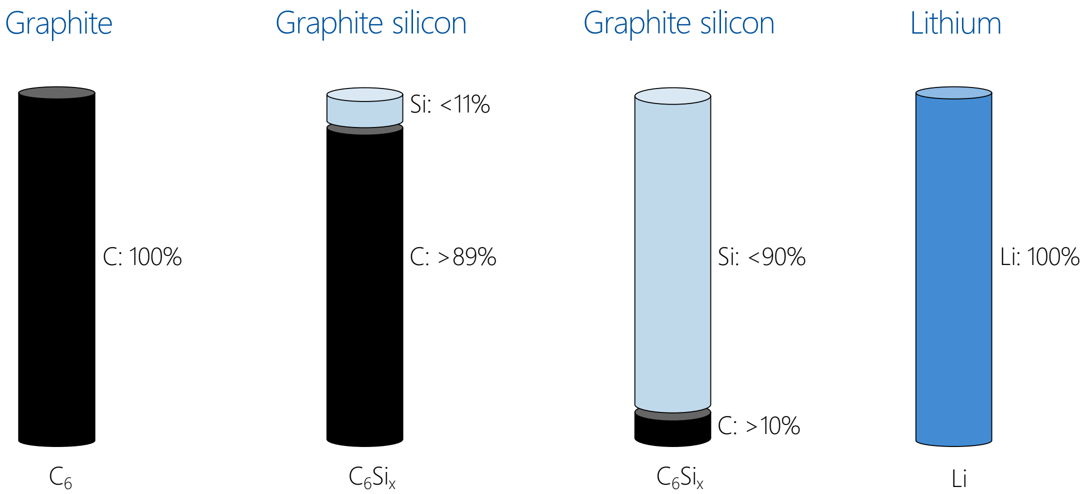
- There is currently a strong market concentration on NMC cathode materials, which are well suited for economical recycling processes due to their expensive materials.
- LFP is getting important due to its long service life and low material prices, which can pose an economic challenge for recycling.
- The trend towards high nickel content looks set to continue in the coming years.

* Internal market analysis PEM, 2023; Friedrich et al.: Recovery of Valuable Metals from E-Waste and Batteries by Smart Process Design, 2020; Mayyas et al.: The case for recycling: Overview and challenges in the material supply chain for automotive li-ion batteries, 2018

Anode Composition

Fundamentals of battery recycling





Annotations

- The chart above shows the **mass shares** of the four most important **anode active materials** in 2023. Graphite and graphite-silicon dominate the market. Silicon and lithium anodes are not currently produced on an industrial scale and are more likely to be alternatives in the future.
- **Silicon** is added to graphite anodes to increase energy density. Currently, shares of **less than 11%** are possible. However, this anode composition has not yet reached the mass market. Theoretically, anodes with significantly higher silicon content are also conceivable, which would then have even higher energy density. The biggest challenge in their implementation is the fact that the anode changes its volume considerably during the charging and discharging process – by up to 300%. This is often accompanied by an extremely short service life of the battery cell. There are currently several companies with promising approaches to meet this challenge so that anodes with high silicon content and a carbon matrix appear realistic in a few years' time.
- **Pure lithium metal anodes** theoretically enable even higher energy density than silicon and graphite anodes. Their biggest challenge at present is that they are not stable in combination with liquid electrolytes. The implementation of solid-state batteries (SSB) could be accompanied by an anode revolution, with the trend also moving towards lithium-free anodes. Another disadvantage of lithium anodes is that the raw materials are expensive and in short supply.

Key Takeaways

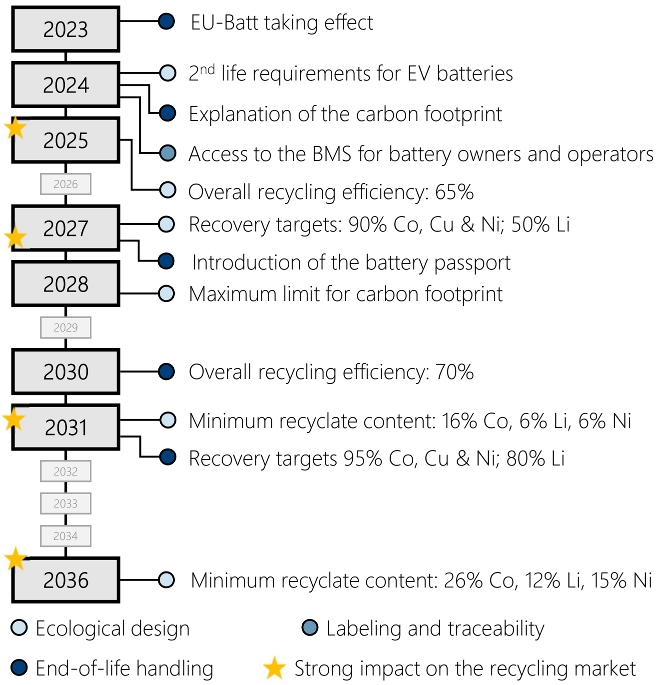
- Graphite currently dominates the market as anode material and is unlikely to be replaced in the coming decade.
- There are promising alternatives that could significantly increase batteries' energy density and performance in the future.

* Internal market analysis PEM, Chae et al.: Integration of Graphite and Silicon Anodes for Lithium-Ion Batteries, 2019; Asenbauer et al.: The success story of graphite as a lithium-ion anode material, 2020

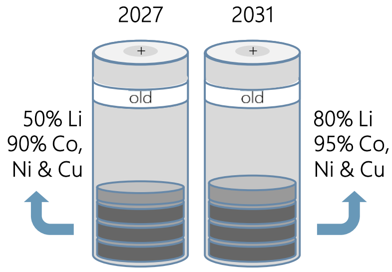
Legal Framework

Fundamentals of battery recycling

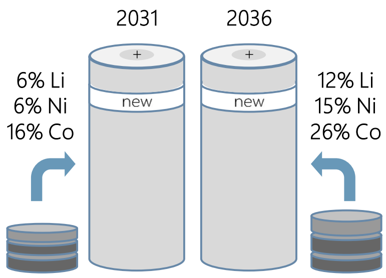
Main effects of the European Battery Regulation



Material recovery rates



Minimum recycled content



Annotations

- In order to achieve the climate policy goals with a view to the disproportionate growth in electrification, sustainability must be guaranteed in the form of minimum requirements. The entire life cycle of the batteries must be taken into account.
- The Battery Regulation adopted by the European Commission in 2023 replaces the German Battery Act 2006/66/EC. It ensures the sustainability and competitiveness of battery production in Europe.
- Key points are the collection of EoL batteries, introduction of the battery passport, maximum limits for the carbon footprint, overall recycling efficiency, recovery targets and minimum recycled content.
- From 2025, an overall recycling efficiency of 65% is required, rising to 70% in 2030. From 2027, all recycling processes must achieve a material recovery rate of 50% lithium, 90% cobalt and 90% nickel and copper each. The recovery rate requirement will increase by 2031. An 80% rate will be allocated to lithium, while copper, nickel and cobalt must each achieve a recovery rate of 95%.
- For the period from 2031, it is also required that newly produced LIBs contain a minimum recycled content of 16% cobalt, 6% lithium, and 6% nickel and that this is documented in the accompanying technical documentation. The minimum share of these raw materials is to be increased again in 2036. New LIBs are to contain 26% cobalt, 15% nickel, and 12% lithium recyclate.

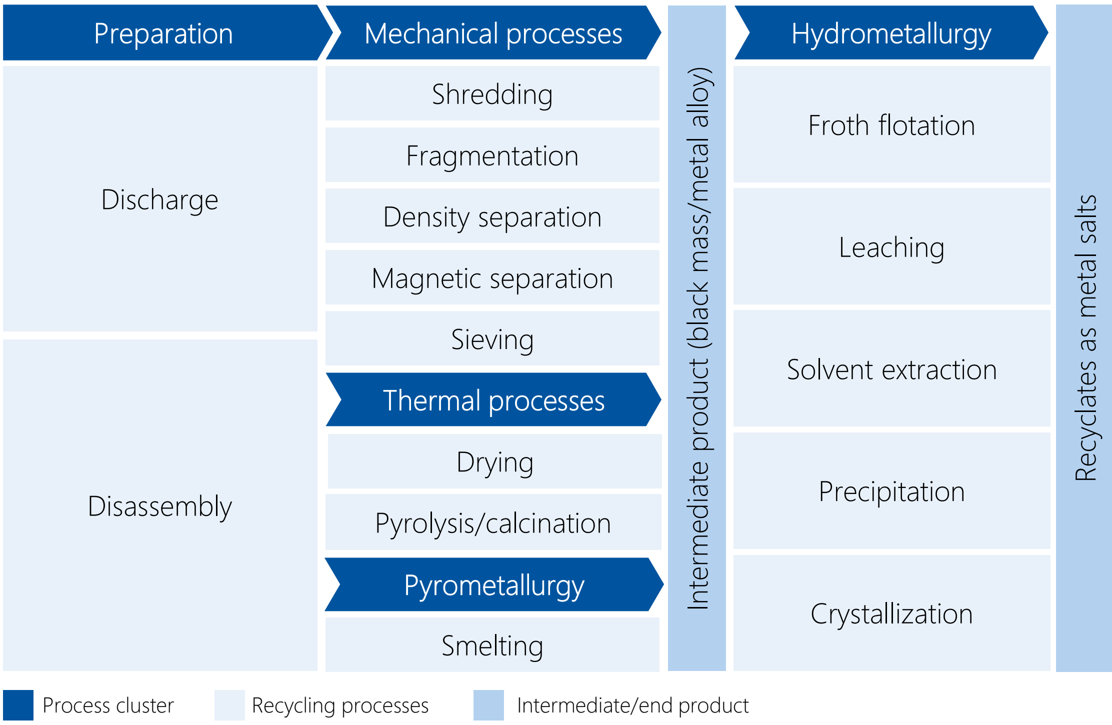
Key Takeaways

- High requirements for the recovery of materials and the use of recyclate volumes are strengthening the recycling market in Europe.
- The battery passport and the identification of the carbon footprint ensure greater transparency in the battery industry. Energy efficiency of recycling processes is also gaining in importance.

European Union: New EU regulatory framework for batteries, 2023

Process routes

Battery recycling overview



The recycling processes listed here can be combined with each other in different ways in recycling routes. Not all recycling processes have to be used.

Annotations

- The main objectives of recycling lithium-ion batteries are to reuse materials indefinitely by closing the product life cycle, thereby reducing waste disposal and dependence on important primary materials.
- The recycling process for lithium-ion batteries after discharge and disassembly consists of several process steps, each of which releases different products/recyclable materials.
- While the combination of the 'preparation', 'mechanical recycling' and 'thermal recycling' process clusters lead to the intermediate product (usually 'black mass'), pyrometallurgy can be used to recover the metals cobalt, nickel and copper while hydrometallurgy can also be used to recover lithium, aluminum and manganese as metal salts.
- The term 'black mass' defines a mixture of anode material and/or cathode material and/or electrolyte and/or other components that is produced during the recycling or recovery of battery materials after mechanical and possibly thermal processing and separation and that may contain impurities.
- One of the main challenges of the recycling process is the recyclable materials' recovery quality to ensure that they can be used again. However, current industrial technologies cannot recover every material.

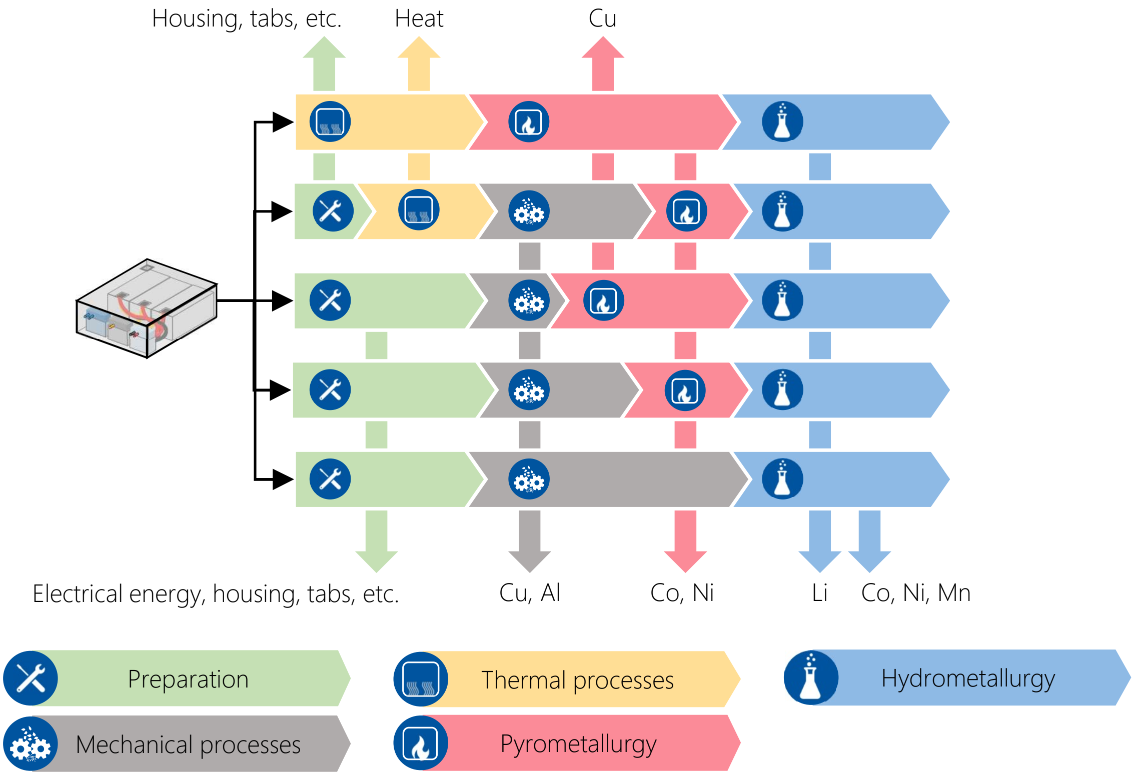
Key Takeaways

- Several recycling processes exist in research and industry. Many of those processes already exist on the market and have been adapted for the recycling of lithium-ion batteries.
- Many new recycling processes are being developed for the recycling of lithium-ion batteries.

Harper et al.: Recycling lithium-ion batteries from electric vehicles, 2019;
Islam et al.: Lithium-ion battery recycling in the circular economy: A review, 2022

Recycling Process

Battery recycling overview



Annotations

- Various approaches to recycling lithium-ion batteries have been implemented in industry and science: mechanical treatment, thermal treatment, pyrometallurgy, and hydrometallurgy. The combination of these recycling approaches enables various process routes that vary in effectiveness and have advantages and disadvantages depending on the focus of material recovery (e.g. nickel vs. lithium).
- The efficiency with which individual recyclable materials are recovered from batteries depends largely on the selected combination of process steps. It is necessary to precisely coordinate these steps in order to ensure an optimum recycling rate and high quality of the recovered materials.
- The diversity of battery pack designs and the materials used in them requires robust and adaptable recycling processes. To ensure high recycling rates and product quality, the aim is to design the process routes in such a way that they function efficiently regardless of the material input.
- To ensure safety of the subsequent steps in the recycling process, measures against potential electrical, chemical and thermal risks of the battery system must be taken right at the beginning of the recycling chain.
- The recovered materials must have a purity that corresponds to 'battery grade' quality to ensure the high quality of newly produced batteries.

Key Takeaways

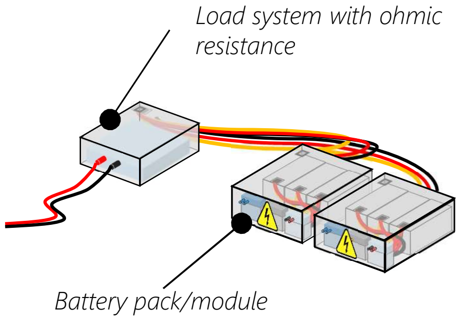
- There are numerous recycling processes for lithium-ion batteries, but no industry standard has yet been established.
- Although hydrometallurgy requires a higher initial investment, it is indispensable for obtaining metal salts of the quality required for batteries.

Doose, S.; Mayer, J.K.; Michalowski, P.; Kwade, A.: Challenges in Ecofriendly Battery Recycling and Closed Material Cycles: A Perspective on Future Lithium Battery Generations, in: Metals 2021, 11, 291; https://doi.org/10.3390/met11020291

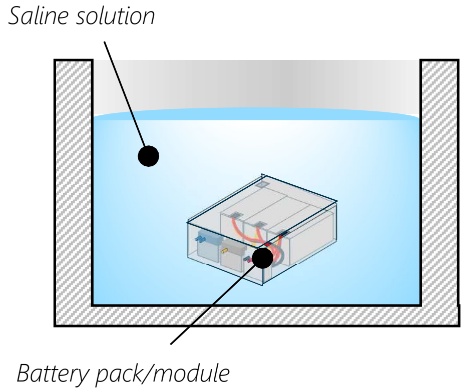
Discharge

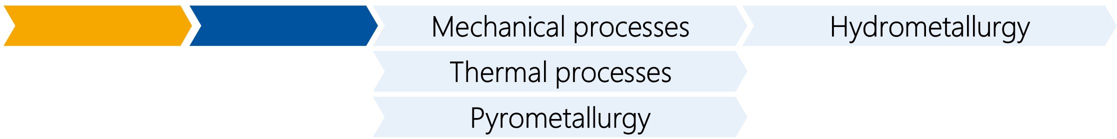
Preparation

<u>Technology 1:</u>
Discharge with energy recovery



<u>Technology 2:</u>
Saline solution





Annotations

- Batteries are deep-discharged and short-circuited as far as possible to reduce the risk for the further recycling process sequence. There is no industry standard for discharging electric vehicle batteries. Several procedures are proposed in the technical literature, but there is little detailed information on the methods used.
- Two possible technologies are discharge with optional energy recovery or by means of saline solution. With saline discharge, the battery cells of the module or battery pack are immersed in a saline solution for up to 24 hours. The dissolved salt acts like an electrolyte that undergoes electrolysis and conducts electrons between the poles in a slow short circuit.
- Deep discharge starts at 2.5 volts per cell. It has been determined that a cell with 2.5 volts (SOC of 0%) has too high a voltage for safe crushing, as the potential between the anode and cathode is high enough to cause thermal runaway or sparking during crushing. Inertization during shredding should therefore always be ensured.

Process advantages

- The discharge enables safe handling for further treatment and reduces the risk of thermal runaway.
- It is possible to recover the remaining energy in the battery system (at least for technology 1).
- Disassembly can be carried out in a stress-free state, whereby thermal runaway is no longer possible.

Challenges

- Discharging cells to 0 V (deep discharge) leads to the dissolution of Cu in the electrolytes.[3],[4]
- When the battery rests at 0 V after discharge, the voltage rises, Cu precipitates and causes its distribution throughout the cell, leading to contamination in downstream products.
- The condition and life cycle of an EoL battery is often unknown and can vary greatly.

Materials removed from main material flow stream

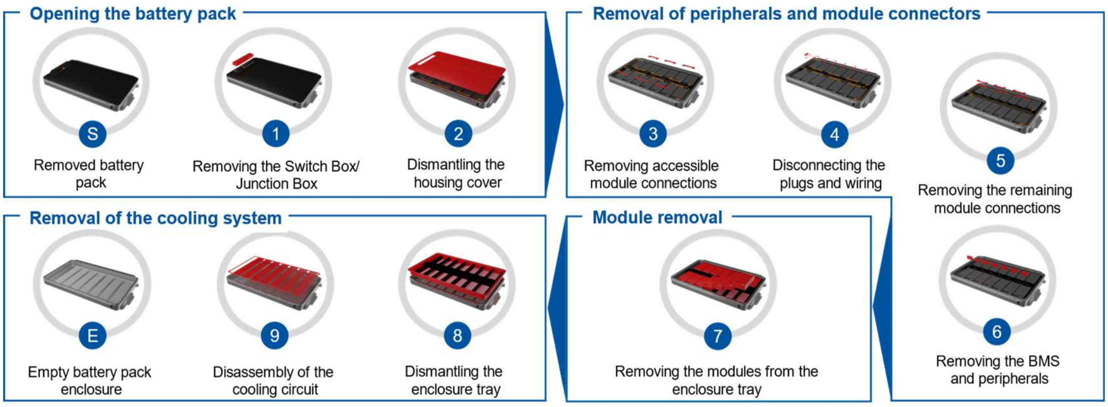
- None

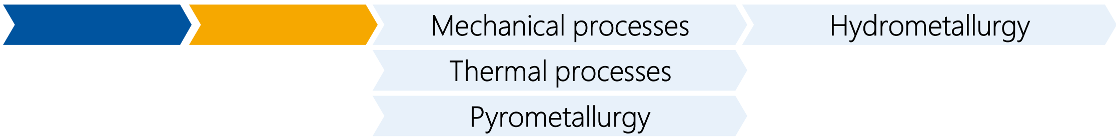
Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020;
Kim et al.: A comprehensive review on the pretreatment process in lithium-ion battery recycling, 2021

Disassembly

Preparation

Generic procedure for battery pack-to-module disassembly





Annotations

- The aim of disassembly is the early separation of component and material groups before further mechanical processing. By loosening the joints, individual material streams can be created that do not have to be separated later with complex recycling processes and the associated contamination.
- Disassembly not only is the basis for subsequent recycling processes but is also used as part of re-use and remanufacturing efforts.
- Currently, the disassembly process down to module level is usually carried out manually, as return quantities are still too low and there is a wide variety of battery system designs on the market.
- Dismantling is carried out by specialized workers (high-voltage training is required for a battery pack of 400 to 800 volts) and varies in process duration from half an hour to two hours, depending on the system to be disassemblied.
- Designs such as glued or sealed cell-to-pack approaches pose a challenge for the process.

Process advantages

- Compared to the shredding process, disassembly leads to easier separation and ultimately to improved material quality.
- Deep disassembly can generate cost savings for the entire recycling route, as it leads to high-purity product streams and a higher yield of products, but also to simpler flow-sheets.

Challenges

- The large number of battery systems variants is a hurdle for future automation efforts which are vital for cost reduction.
- Non-destructively detachable joints complicate process steps such as lid opening or module removal.
- Manual procedures involve potential hazards such as the high-voltage environment or contact with toxic electrolytes.

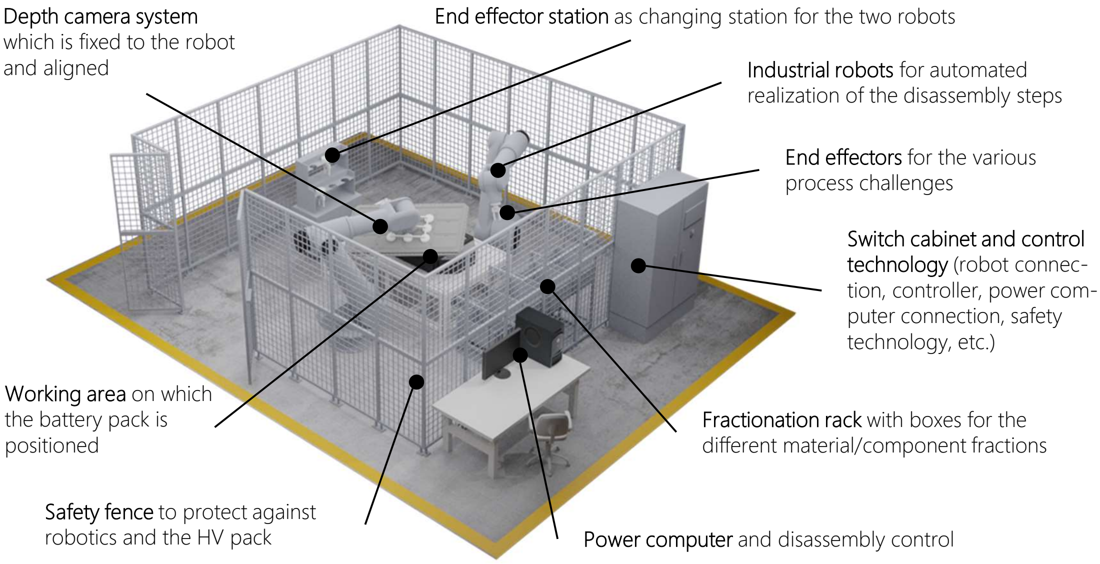
Materials removed from main material flow stream

- Housing components (e.g. aluminum), electronics, BMS, module connectors (e.g. copper)

Wu et al.: A systematic review on lithium-ion battery disassembly processes for efficient recycling, 2023; Klohs et al.: Automated battery disassembly – Examination of the product- and process-related challenges for automotive traction batteries, 2023

Automated Disassembly

Preparation



Concept illustration of a prototype cell from the DemoSens research project

Mechanical processes | Hydrometallurgy

Thermal processes

Pyrometallurgy

Annotations

- With steadily increasing sales figures for battery electric vehicles (BEV) and a currently estimated service life of ten to 15 years on average for battery systems until a state of health (SOH) of 70% to 80% is achieved, an economically scalable process for disassembly will be required in the future.
- Automating the disassembly process using robots reduces the risks to workers from the high-voltage environment and escaping electrolytes, but currently still poses many challenges due to the lack of market standards in the design of battery systems.
- In addition to fully automated approaches, collaborative concepts are also currently being developed to increase the level of automation.
- In addition to the different component designs, positions and quantities, accessibility to gripping and screwing points and the condition of the system play an important role in automation capability.

Process advantages

- Dealing with increasing return volumes
- Protection of workers from sources of danger such as the high-voltage environment and toxic electrolyte
- Reduction in the amount of time and therefore the costs of the disassembly steps

Challenges

- Due to the large number of battery system variants, a large amount of data is required for implementation
- Different conditions of the battery systems at the end of the first usage phase (e.g. dirt, rust, deformation)
- Dealing with accessibility and joints is becoming more complex.
- Combining process robustness with the greatest possible flexibility

Market situation

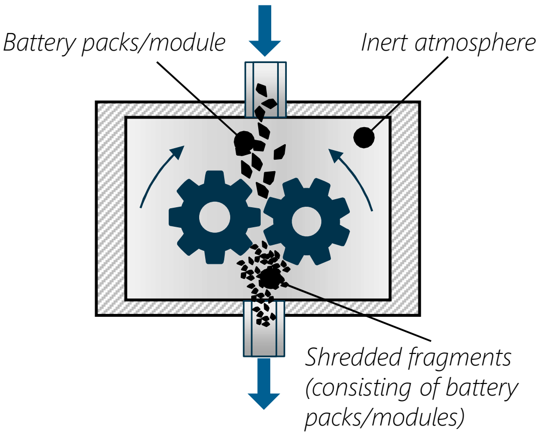
- First concepts/projects are currently available. Demand is increasing due to growing return volumes.

Wu et al.: A systematic review on lithium-ion battery disassembly processes for efficient recycling, 2023; Klohs et al.: Automated battery disassembly – Examination of the product- and process-related challenges for automotive traction batteries, 2023

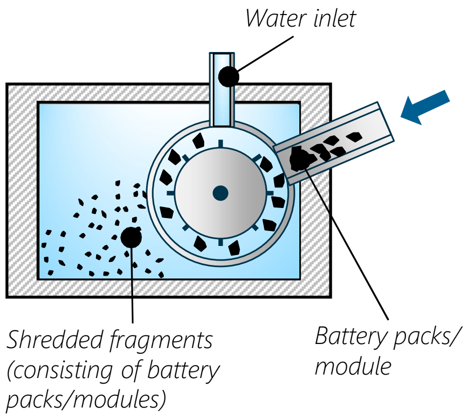
Shredding

Mechanical processes

Technology 1:
Dry shredding



Technology 2:
Wet shredding



Preparation | Hydrometallurgy

Thermal processes

Pyrometallurgy

Annotations

- Shredding is an essential process step for mechanical recycling, as it converts the battery cells or modules into a finer, conveyable bulk material. This process separates the active materials from their packaging and other surrounding components.
- Different shredding processes lead to different starting materials with different sizes and shapes, which influences the downstream separation processes.
- Dry shredding often takes place in an inert atmosphere. The use of inert gases reduces the risk of explosion and fire.
- Wet shredding can be carried out in a water medium that is enriched with the active materials, the electrolyte, and other fine particles. Shredding in water reduces the risk of fire and increases the detachment of the electrode layers from the current collector foils. This process requires complex water treatment.

Process advantages

- Shredding the batteries is essential in order to release and separate the active material in the following process steps.
- Shredded material is easy to transport and is the basis for further recycling processes.
- Shredding avoids difficulties when opening the products, and all products can be handled using the same method.

Challenges

- Compression and deformation during shredding have to be low to avoid the inclusion of valuable materials but must be high enough to produce the smallest possible parts.
- After shredding, a complex separation process is required.
- Product quality in terms of recycling efficiency and possible impurities are strongly influenced by the shredding process.

Materials removed from main material flow stream

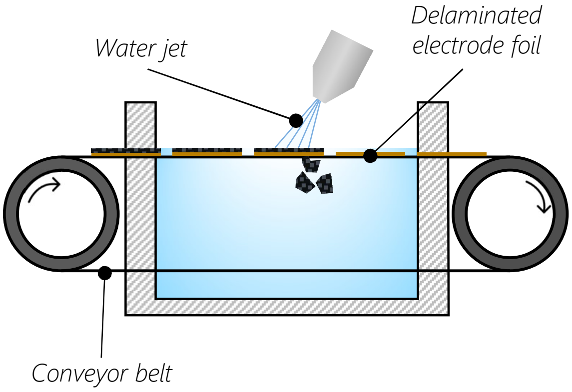
- Shredding does not remove materials but turns them into processable bulk material.

Yan et al.: Research on the high-efficiency crushing, sorting and recycling process of column-shaped waste lithium batteries, 2023;
Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020

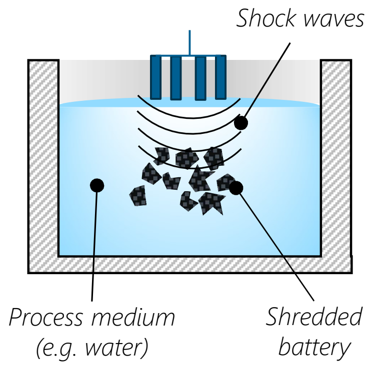
Fragmentation

Mechanical processes

<u>Technology 1:</u>
Water jet treatment



<u>Technology 2:</u>
Shock waves



Annotations

- The water jet treatment takes place on a conveyor belt on which the electrode tracks (anode and cathode separately) are exposed to a water jet, thus detaching the active materials from the electrode foils.
- The shock waves are caused by the electrohydraulic effect generated by a high-voltage discharge between electrodes in the process medium.
- Upstream mechanical shredding is required for the shock waves. This enables the shock waves to attack the interfaces of the multi-component system, such as a lithium-ion battery.
- Separation takes place at macroscopic interfaces during shock waves (bonding, screw connections, and interlocking) and at microscopic interfaces during shock waves and water jet treatment (grain and phase boundaries). The separation is based on the material-specific physical properties of the individual components.

Process advantages

- No need for hazardous and/or toxic chemicals
- No production of environmentally hazardous by-products
- Opens up recovery of high-quality materials through direct recycling

Challenges

- The scalability of the processes has yet to be proven.
- Drying required for subsequent processes

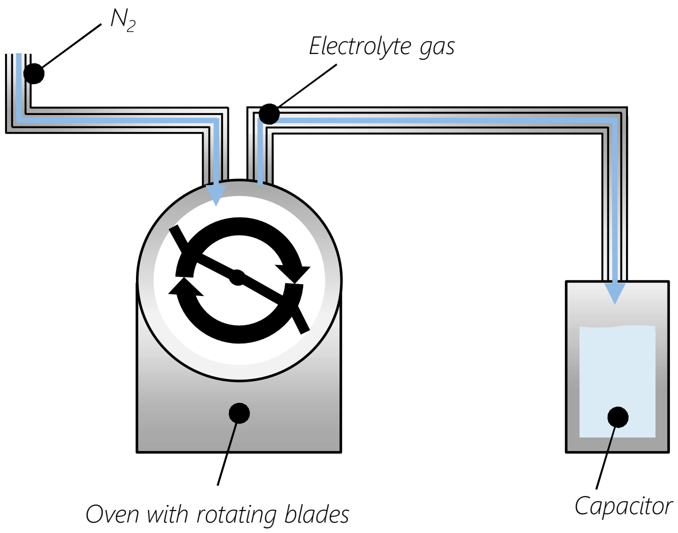
Materials removed from main material flow stream

- None

Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020;
Öhl et al.: Efficient Process for Li-Ion Battery Recycling via Electrohydraulic Fragmentation, 2019

Drying

Thermal processes



Annotations

- The main aim of the drying process is to extract the electrolyte from the crushed batteries by vaporizing it and condensing it elsewhere.
- The process can be carried out in an inert atmosphere, with nitrogen, or in a vacuum.
- Drying can be carried out by vacuum drying, contact drying, convection drying and/or infrared drying. It is advantageous if the drying is carried out while stirring and/or circulating the comminuted material. This leads to a more homogeneous temperature distribution and further exposure of the active materials.
- Vacuum drying is carried out at lower temperatures, preferably below 70°C, at a maximum pressure of 100 hectopascals, with the aim of removing at least one solvent from the conducting salt so that the formation of hydrogen fluoride is inhibited.

Process advantages

- Vacuum drying at low temperatures prevents the formation of hydrogen fluoride and therefore poses no potential danger to the battery processing plant or the environment.
- Vacuum drying at lower temperatures and a condensation system make it possible to recover the electrolyte.

Challenges

- Some electrolyte and binder components with high boiling points may not be able to be separated from the shredded battery material at temperatures of up to 120°C.

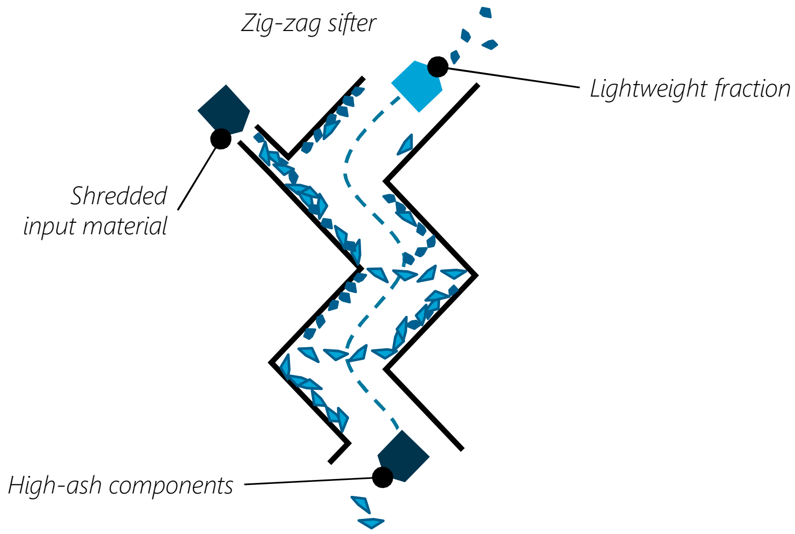
Materials removed from main material flow stream

- Low-boiling electrolyte components and water

Yan et al.: Research on the high-efficiency crushing, sorting and recycling process of column-shaped waste lithium batteries, 2023;
Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020

Density Separation

Mechanical processes



Annotations

- Density separation is used to separate low-density particles from high-density particles. In principle, there are two applications: the separation of the heavy housing parts from the remaining shredded material and the separation of the separator films from the heavier electrode foils made of aluminum with a density of 2.7 g/cm^{-3} or copper with a density of 8.96 g/cm^{-3}. Material for the separator in the battery cell – polyethylene and/or polypropylene – with a density of less than 1 g/cm^{-3} is separated via the light material.
- The size and shape of the particles have a significant influence on the separation performance. Sieving is often required before or after density separation.
- Density separation can be carried out using distributing jiggers, in a medium-density liquid, or by air separation (e.g. zig-zag sifters).
- Density and size separation using zig-zag sifters can be carried out twice: once for the separation of the heavy housing components and later of the separators.

Process advantages

- The degree of separation can be adjusted by the number of repetitions of the separation process depending on the required purity of the subsequent recycling steps.
- Under optimum process conditions, this is an efficient way of separating materials relatively homogeneously.

Challenges

- The geometry of the pieces and the degree of release of LIB fragments have a greater influence on the separation performance than the density differences alone.
- Pieces with high density and low thickness (platelet-shaped or needle-shaped) can have the same paths in the zig-zag sifter as pieces with low density and high wall thickness (cubes or spheres).
- The process reacts sensitively to changes in the input material.

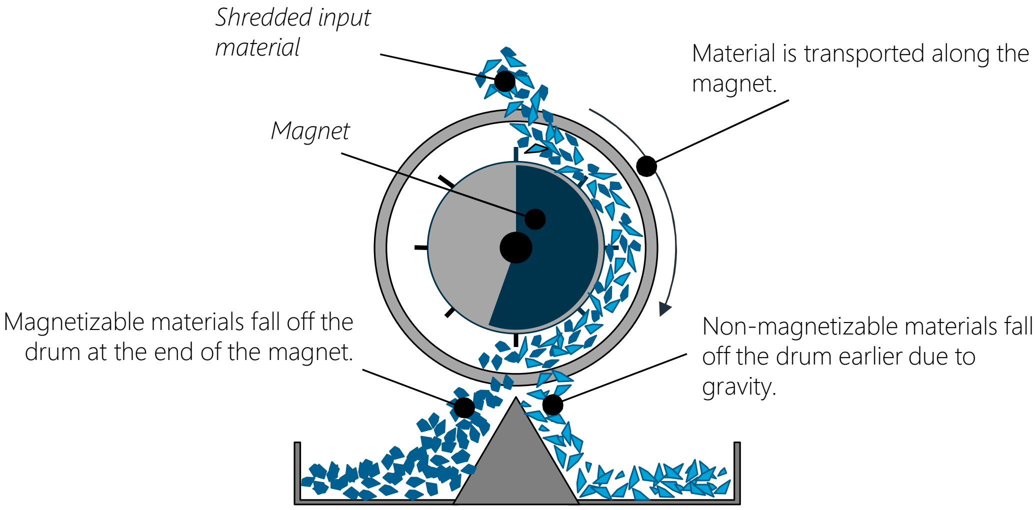
Materials removed from main material flow stream

- (Partially) aluminum, copper, separator material (polyethylene or polypropylene)

Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020;
Wang et al.: Effective separation and recovery of valuable metals from waste Ni-based batteries: A comprehensive review, 2022

Magnetic Separation

Mechanical processes



Annotations

- During magnetic separation, the particles are sorted into magnetic and non-magnetic material.
- The aim of magnetic separation is the removal of steel residues and other magnetic or magnetizable components from the material flow.

Process advantages

- Efficient recovery of coarse steel parts (housing parts, screws, peripheral devices) to protect the following process equipment from wear and tear

Challenges

- Additional steps are necessary to remove the other unwanted particles in the black mass.

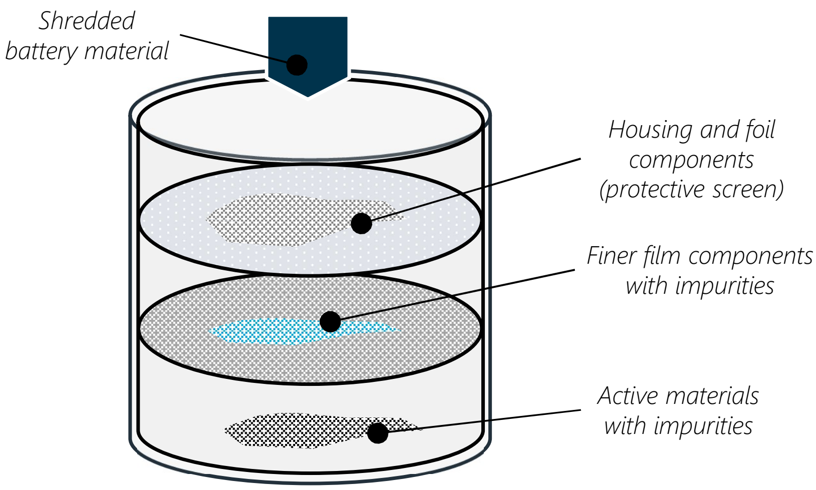
Materials removed from main material flow stream

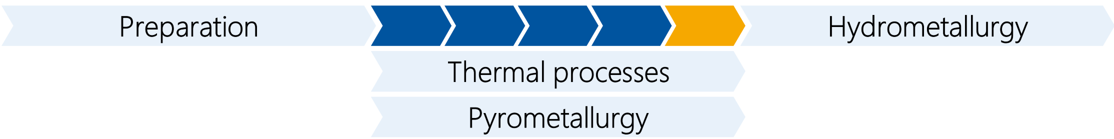
- Magnetic materials – primarily steel, but also copper and aluminum components

Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020;
Wang et al.: Effective separation and recovery of valuable metals from waste Ni-based batteries: A comprehensive review, 2022

Sieving

Mechanical processes





Annotations

- The recycling process step of sieving classifies the shredded battery material according to its particle size. The screening process retains the battery housing materials (Al/Fe/Mn alloy), plastics, and current collectors as coarse particles, while the fines fraction mainly consists of black mass (primarily active materials).
- This separation is possible because the battery housing, separator, and metal foils are elastically-plastically easily deformable and can only be meaningfully shredded by cutting, while the active electrode layers behave much more brittle and are originally present as fine powder.
- The sieves are stimulated by vibrations or tumbling movements to move the screened material in different directions depending on the drive. The process can take place in a sieving tower with different mesh sizes in order to separate the material in several stages and generate different output flows.
- The size fraction of the battery types is quite different in terms of material concentration.

Process advantages

- The homogeneity of the materials separated by sieving can simplify the use of subsequent recycling steps and minimize the loss of valuable resources in waste streams.
- Active materials can be efficiently separated from other cell components due to their particulate nature.

Challenges

- Loss of active material in the coarser fractions that adhere to metals (Cu, Al, Fe) from the battery housing and the current collectors

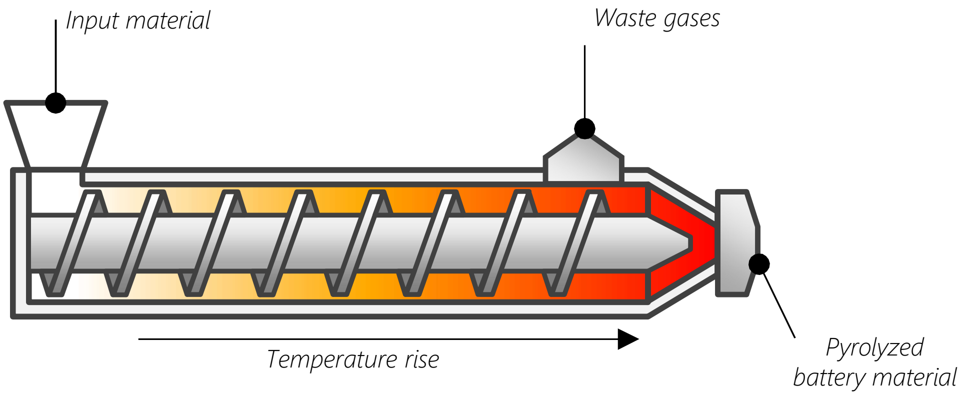
Materials removed from main material flow stream

- Coarse particles of Al/Fe/Mg alloy, plastics, and current collectors

Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020;
Wang et al.: Effective separation and recovery of valuable metals from waste Ni-based batteries: A comprehensive review, 2022

Pyrolysis/Calcination

Thermal processes



This visualization is one way of implementing the technology.

Annotations

- Pyrolysis or calcination can be used before or within the mechanical recycling chain (e.g. for shredded material) to improve the recovery of cathode and anode materials from spent lithium-ion batteries.
- During pyrolysis, organic compounds are decomposed at high temperatures (600 °C) in the absence of oxygen. Calcination takes place in an oxygen atmosphere. The main aim of these thermal processes is to remove high-boiling solvents and salts from the electrolyte as well as the binders so that further recycling steps can be carried out with a low fire risk and lower requirements for hydrogen fluoride formation.
- By decomposing the binders, such as polyvinylidene fluoride (PVDF), the active materials of the anode and cathode are better detached from the copper or aluminum foil, which increases the recycling yield and safety in material handling.
- In addition to whole batteries, pyrolysis is also used for black mass to improve flotation and in hydrometallurgical processing. After pyrolysis, the material must be cooled in cooling pipes or cooling coils.

Process advantages

- Batteries can be safely stored after pyrolysis without the risk of explosion as the battery is deactivated and the electrolyte removed.
- Recovery rate and recovery quality both increase as the organic binders are decomposed and the decoating of the electrode foils and thus the recovery of the active materials is facilitated.

Challenges

- PVDF decomposes during treatment, releasing gaseous hydrogen fluoride and a fluorine-containing organic by-product. Hydrofluoric acid is harmful and can attack glass, concrete, and numerous metals.
- Waste gas treatment is necessary and increases investment and maintenance costs of the recycling plant.
- Electrolyte and binder components are lost and cannot be recycled.

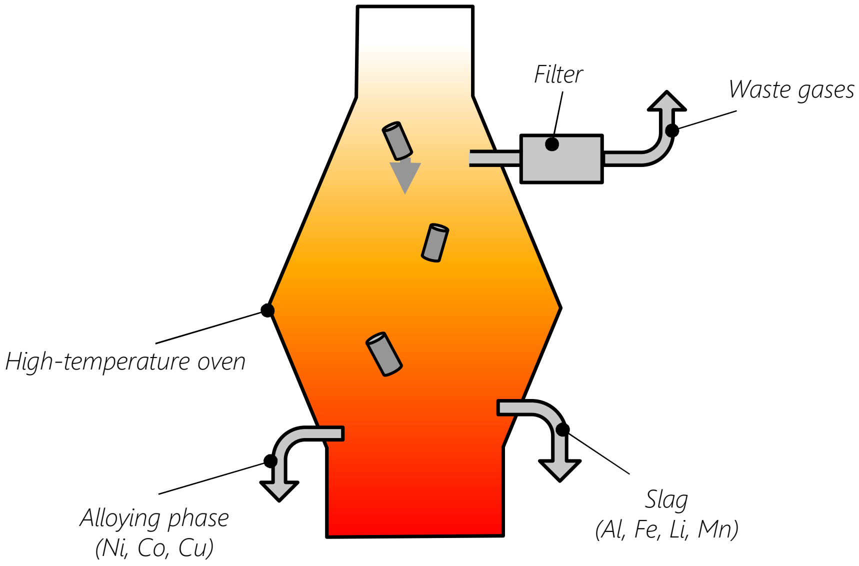
Materials removed from main material flow stream

- Binding agents such as polyvinylidene fluoride (PVDF) and electrolyte components, partly conductive salt

Kim et al.: A comprehensive review on the pretreatment process in lithium-ion battery recycling, 2021

Smelting

Pyrometallurgy



Preparation | Mechanical processes | Hydrometallurgy

Thermal processes

Annotations

In the course of pyrometallurgy, battery materials are melted at high temperatures, resulting in molten metals (Ni, Co, Cu), slag, and dust. The process takes place in a furnace in which the material is heated to between 1,200°C and 1,450°C. This heating burns off organic components (such as plastic, electrolyte, graphite) and utilizes them for energy.

- As the melting temperature of the metals increases, nickel (Ni), cobalt (Co), and copper (Cu) pass into the alloy phase, while aluminum (Al), iron (Fe), lithium (Li), and manganese (Mn) pass into the slag phase.
- In addition to the metal alloy, slag and furnace dust are also produced. These by-products contain metals such as lithium and manganese and must be processed in further recycling steps, in particular hydrometallurgy. A separation of lithium and other metals upstream of pyrometallurgy for their recovery is currently being investigated but not yet in commercial use.

Process advantages

- The battery cells and smaller battery modules can be inserted directly into a pyrometal-lurgical process, and the molten metal (alloy phase) can be fed directly into the leaching process, drastically reducing the total number of processes.
- Robustness against different battery designs and battery chemistries

Challenges

- Graphite cannot be recovered as it is used for energy and as a reducing agent.
- As the slag is chemically very stable, an ex-tremely high mechanical and chemical effort is required to separate the materials.
- Achievement of EU recycling rates, especially for lithium, which is strongly bound in the slag

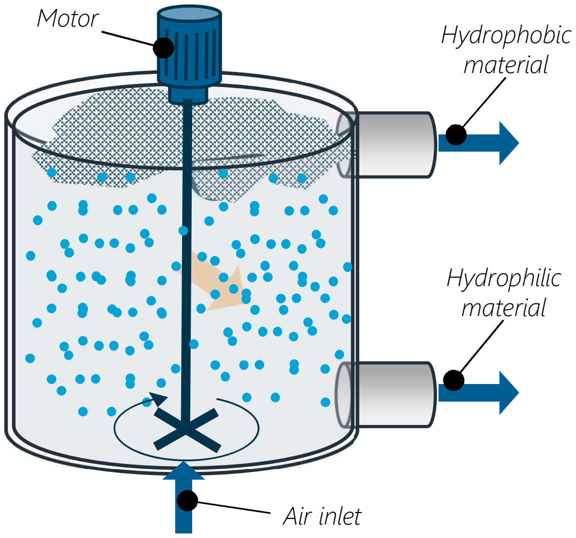
Materials removed from main material flow stream

- Slag (aluminum, iron, lithium, manganese)

Makuza et al.: Pyrometallurgical options for recycling spent lithium-ion batteries: A comprehensive review, 2021

Froth Flotation

Hydrometallurgy



Preparation | Mechanical processes | Thermal processes | Pyrometallurgy

Annotations

- The treatment of black mass using **froth flotation** has recently generated interest as a method for separating valuable cathode materials and graphite particles.
- Foam flotation uses flotation additives to exploit the difference in **hydrophobicity** between the materials in order to separate the active anode and cathode materials of the lithium-ion batteries.
- Fine air bubbles are blown into an agitated tank at the bottom, in which the material to be separated is kept in suspension together with chemical additives. The air bubbles attach themselves to the **hydrophobic materials (graphite)** and transport them to the surface where they are separated. **Hydrophilic particles** remain in the suspension and are removed with the underflow of the separation unit.
- It is advisable to use surface modification techniques – such as **anaerobic pyrolysis** and/or **mechanical pulverization** – to improve the efficiency of the froth flotation process.

Process advantages

- By removing the graphite particles (as a product) from the black mass, the material flow becomes more homogeneous and less voluminous, which is advantageous for the downstream hydrometallurgical processes.

Challenges

- If the binder is not sufficiently removed, a considerable amount of cathode material passes into the foam phase together with the graphite, resulting in losses of valuable metals and inferior graphite at the same time.

Materials removed from main material flow stream

- Graphite particles

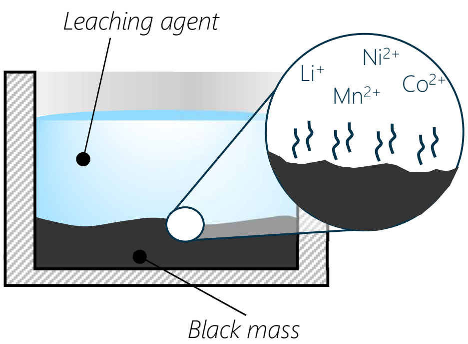
Sommerville et al.: A qualitative assessment of lithium-ion battery recycling processes, 2020

Wang et al.: Effective separation and recovery of valuable metals from waste Ni-based batteries: A comprehensive review, 2022

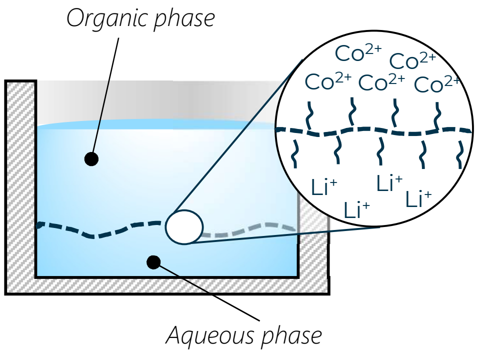
Leaching and Extraction

Hydrometallurgy

Leaching



Solvent extraction



Preparation | Mechanical processes | Thermal processes | Pyrometallurgy

Annotations

- Leaching is used to convert metals from the cathode material into ions in aqueous solution using acid so that the metals can be recovered in subsequent processes using a range of chemical methods (e.g. precipitation, solvent extraction).
- The leaching reaction takes place at the interface between the solid and liquid phases.
- There are several acids that can be used for the leaching process, including organic and inorganic acids.
- Leaching processes can be influenced by various parameters, such as leaching time, temperature (60°C to 90°C) of the solution, ratio of solid to liquid, the addition of an oxidizing agent and the concentration of the leaching acid.
- Solvent extraction is a separation method in which the different solubility of substances in two immiscible solvents is utilized. It follows leaching to selectively extract the metals from the leachate into an organic phase, which has a lower density and floats.

Process advantages

- The leaching process can be used with most cathode chemistries (only LFP requires pre-treatment).
- Leaching enables high purity and high recovery rates with low energy consumption and is suitable for large-scale applications.
- Extraction achieves high levels of efficiency with very low energy consumption.

Challenges

- Low process flexibility regarding the material composition of the cathode material
- Inorganic acids release toxic gases (Cl_2, SO_3, NO_x) as a by-product.

Materials removed from main material flow stream

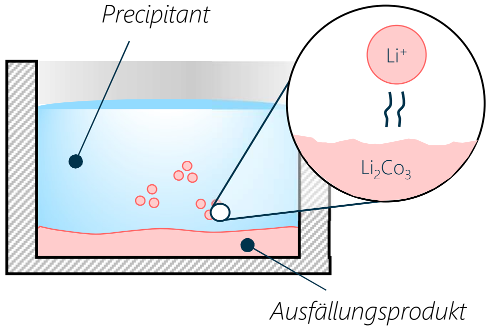
- None

Jung et al.: A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments, 2021

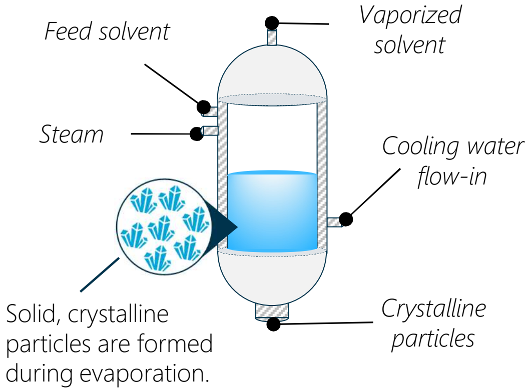
Precipitation and Crystallization

Hydrometallurgy

Precipitation



Crystallization



Preparation | Mechanical processes | Thermal processes | Pyrometallurgy

Annotations

- **Precipitation** is used to recover salt from a solution by adding precipitants – for example after leaching or solvent extraction – in order to obtain valuable substances such as Li, Ni, Mn, or Co.
- The **separation effect** occurs when the solubility of the salt is exceeded due to changes in the ambient conditions (e.g. adjustment of pressure, temperature, or pH value).
- A well-known process sequence is **selective precipitation**, in which the pH value of the leachate is successively increased in order to extract the valuable substances from the liquid step by step.
- The most common process for recovering metal salts from lithium-ion batteries is **evaporation crystallization**. It is generally used to produce Ni ($NiSO_4$), Co ($CoSO_4$) and Mn ($MnSO_4$) sulphates after solvent extraction and achieves relatively high crystal growth rates due to high temperatures (approx. 350°C).

Process advantages

- Precipitation is the safest, most economical and most efficient process (recovery efficiency) of the hydrometallurgical methods.
- Precipitation is generally used to produce Li_2CO_3, as it is practically insoluble in neutral or basic aqueous solutions.
- Low operating costs make crystallization an attractive process step.

Challenges

- Precipitation methods are best researched. However, precise control of the operating conditions (e.g. pH value and temperature) is necessary to achieve the desired property profile.
- Evaporation crystallization is a very time-consuming and energy-intensive process step, as the aqueous solution must be completely evaporated.
- Processes react sensitively to variations in the input material.

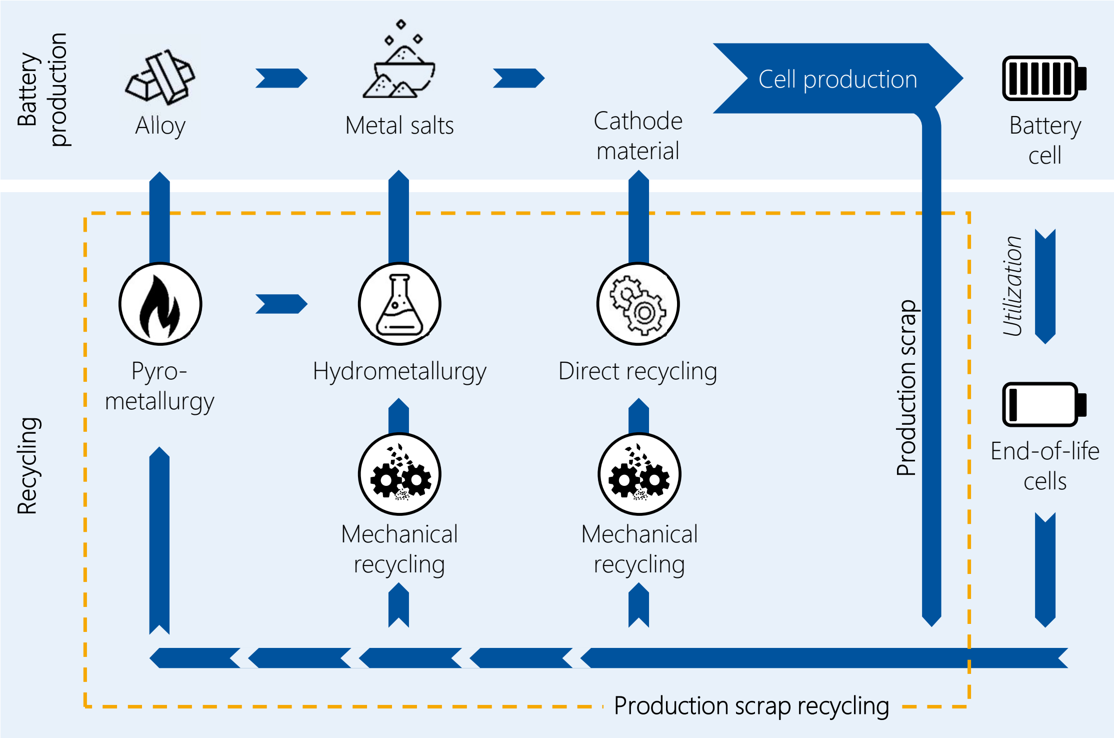
Materials removed from main material flow stream

- None

Jung et al.: A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments, 2021

Production Scrap Recycling

Special forms of recycling



Annotations

- Production scrap recycling refers to the recycling of production waste from battery manufacturing. Due to the exponentially increasing volume of batteries produced and the time it takes to reach the end of their life, production scrap recycling will be more relevant in terms of volume than end-of-life recycling in the coming years.
- The variety of materials at cell level is lower than at pack level. This simplifies separation and leads to a potential reduction in individual process steps compared to the recycling of EoL batteries. The direct separate processing of anodes and cathodes is also interesting.
- By using production waste, individual recycling process steps can be omitted. This includes disassembly, discharge, or pyrolysis, depending on the process step in which the production waste is generated.
- Depending on when the production scrap is generated, the recycling process can be simplified. Overall, the recycling of production scrap is less dangerous as fewer critical components or systems need to be handled.

Battery production scrap

- Assuming a reject rate of 10%, the amount of rejects in worldwide LIB production is forecast to reach 905,000 tons in 2030.
- The source of the battery scrap can be found in the battery production facilities. Separate collection of anodes and cathodes is possible until cell assembly.
- Mechanical, thermal and wet-chemical processes are used.

EoL batteries

- End-of-life (EoL) batteries are forecast to reach 820,000 tons worldwide in 2030.
- In addition to exceeding the expected service life, the sources of EoL batteries also include accident scenarios or recalls before delivery to customers due to defects.

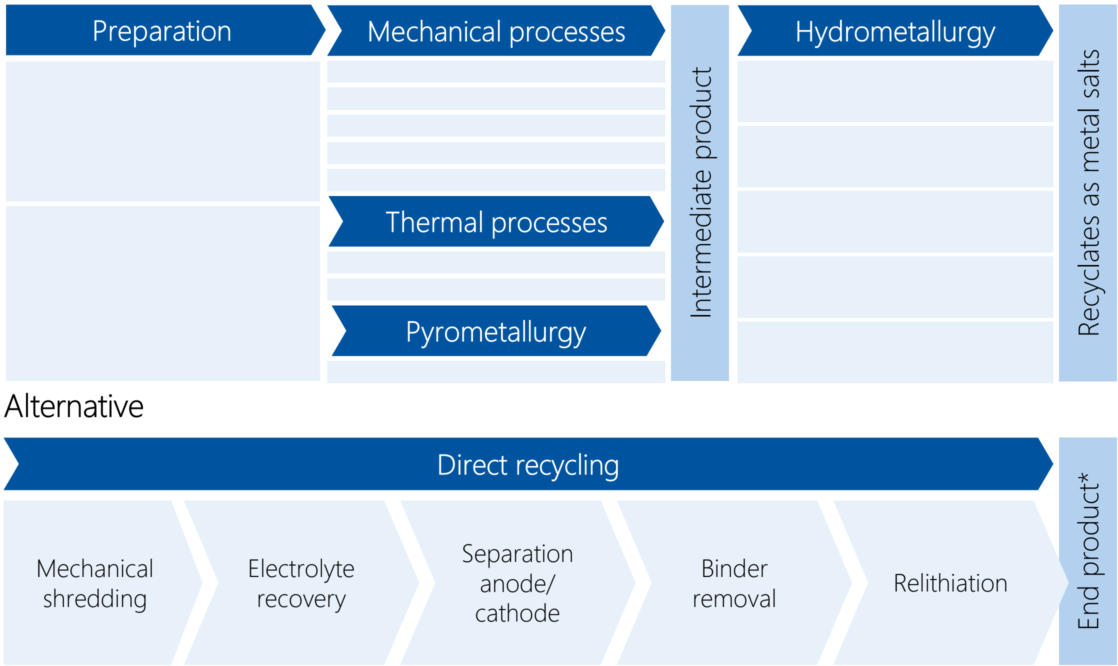
By 2030, the global amount of waste from battery production will exceed the amount of end-of-life batteries.

Gaines et al.: Tracking Flows of End-of-Life Battery Materials and Manufacturing Scrap, 2023;
Gaines et al.: Direct Recycling R&D at the ReCell Center, 2021

Direct Recycling

Special forms of recycling

Conventional recycling processes



* Here, the end product is directly cathode material.

Annotations

- 'Direct recycling' describes the approach of not breaking down the material to be recycled into its basic elements but recovering cathode material directly. This process belongs to a narrower circle of the battery cycle and is potentially associated with lower costs and fewer emissions than state-of-the-art recycling processes such as pyrometallurgy and hydrometallurgy. For this reason, direct recycling has received some attention in recent years and is being intensively researched by scientists and industry.
- Direct recycling usually involves mechanical shredding, recovery of the electrolyte, separation of the anode and cathode material, removal of the binder, and final relithiation. Processes such as shock wave shredding also have great potential for direct recycling, as this enables single-variety shredding.
- The process of relithiation serves to repair the cathode material at a molecular level. During service life, lithium ions are chemically bound, which can then no longer contribute to the charge transfer and thus lead to a reduced battery capacity. These losses can be partially reversed through various relithiation processes.
- The greatest challenge currently is the still inadequate quality of the cathode material and the low level of technological maturity of the processes involved. In addition, direct recycling is particularly economically attractive if the recovered active materials have similar specific capacities to the current active material.

Key Takeaways

- Direct recycling is an interesting alternative to state-of-the-art recycling processes, as it enables shorter process times, lower costs, and lower emissions.
- As a rule, the recovered material does not yet meet the high demands of battery production.
- The scaling of direct recycling processes poses an additional challenge.

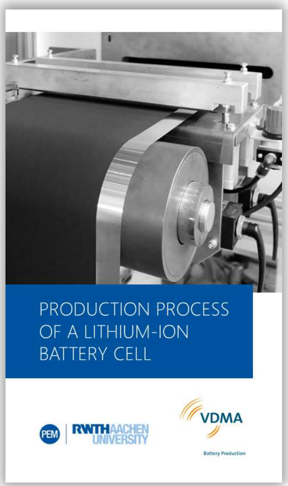
Gaines et al.: Direct Recycling R&D at the ReCell Center, 2021;
Yi et al.: Direct recycling technologies of cathode in spent lithium-ion batteries, 2021

Guides

on electric mobility

Available online

In various battery-related publications, the Chair of Production Engineering of E-Mobility Components (PEM) of RWTH Aachen University, in collaboration with VDMA, presents the different process and recycling chains from cell to battery pack and goes into detail about the manufacturing methods of the numerous components.



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The 'Production Process of a Lithium-Ion Battery Cell' guide provides a comprehensive overview of the production of different battery cell formats, from electrode manufacturing to cell assembly and cell finishing. Furthermore, current trends and innovation of different process technologies are also explained.

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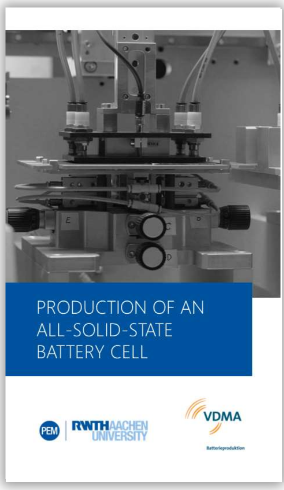

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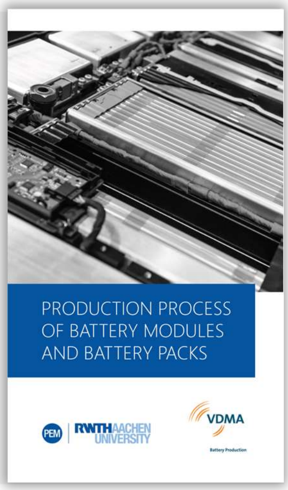

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