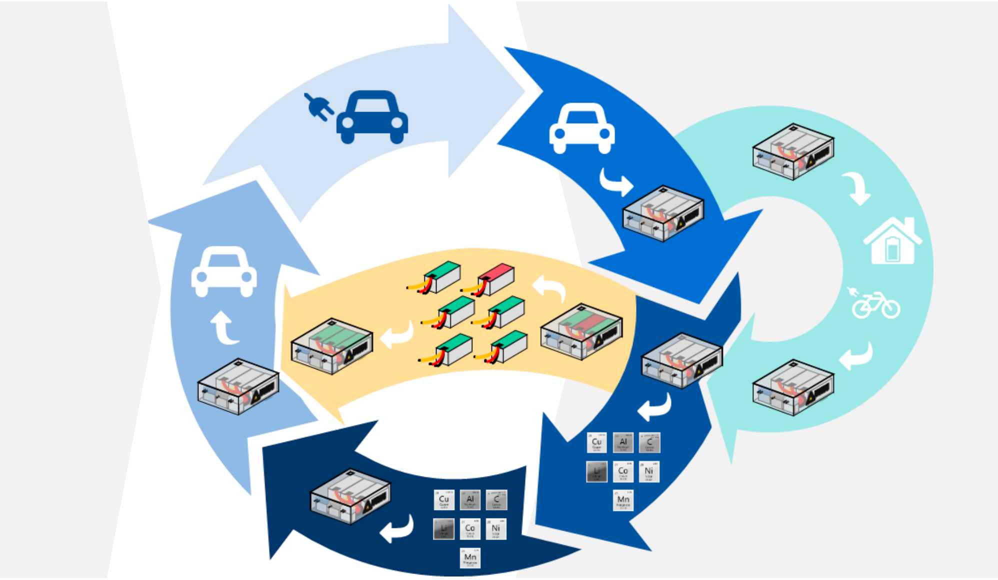


RECYCLING OF LITHIUM-ION BATTERIES

PEM | RWTH AACHEN UNIVERSITY

VDMA
Battery Production

BLB BATTERY LABFACTORY BRAUNSCHWEIG
Ein Zentrum der TU Braunschweig

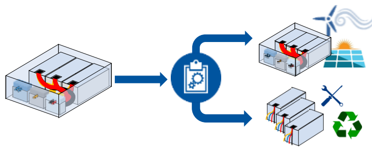
Overview

Lithium-ion battery (LIB)

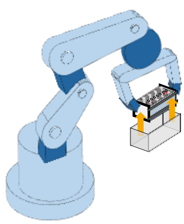
- The **amount** of lithium-ion batteries (LIBs) in their "end of life" (EoL) will increase significantly in the coming years due to the growing market penetration of electric vehicles, which is why new **concepts for recycling and raw material recovery** must be developed.
- The **process scrap** generated in battery production will ensure a need for higher recycling capacities in the near future.
- To implement **sustainable EoL concepts**, all players along the value chain from material synthesis to battery cell, battery module, and battery pack production to the use phase must address this issue.
- **Recycling rates** for individual materials (up to 95%) are proposed by the **CEID (Circular Economy Initiative Germany, acatech)*** and also envisaged by the **EU** in the **Battery Directive.**

- Fundamentals
  - Structure and function of LIB
  - Cost overview
  - Cathode composition
  - Anode composition
  - Life Cycle Assessment (LCA)
  - Sustainability
  - LIB recycling

- Handling EoL batteries
  - Risks in handling with LIB
  - Transport and storage
  - Characterization of LIB
  - LIB classification



- Disassembly
  - Challenges
  - Opening of the housing
  - Disassembly of the battery pack
  - Disassembly of the battery modules
  - Automation of the process



- Recycling
  - Overview
  - Mechanical recycling
  - Pyrometallurgy
  - Hydrometallurgy

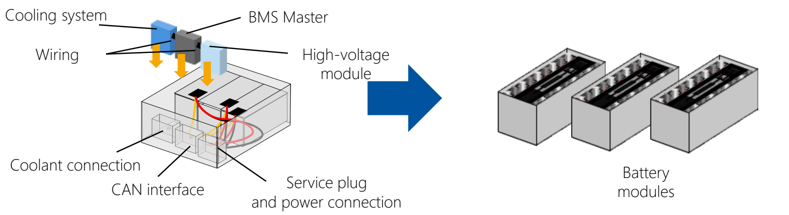
* Circular Economy Initiative Deutschland (Hrsg.): Ressourcenschonende Batteriekreisläufe – mit Circular Economy die Elektromobilität antreiben, *Kwade, A., Hagelüken, C., et al., acatech/SYSTEMIQ, München/London 2020.

Structure and function of LIB

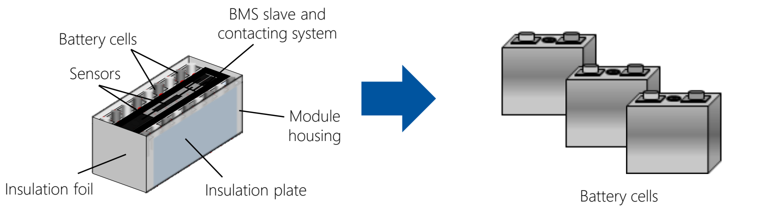
Fundamentals

Dealing with EoL-Bat | Disassembly | Recycling

Battery pack → Battery modules



Battery module → Battery cells

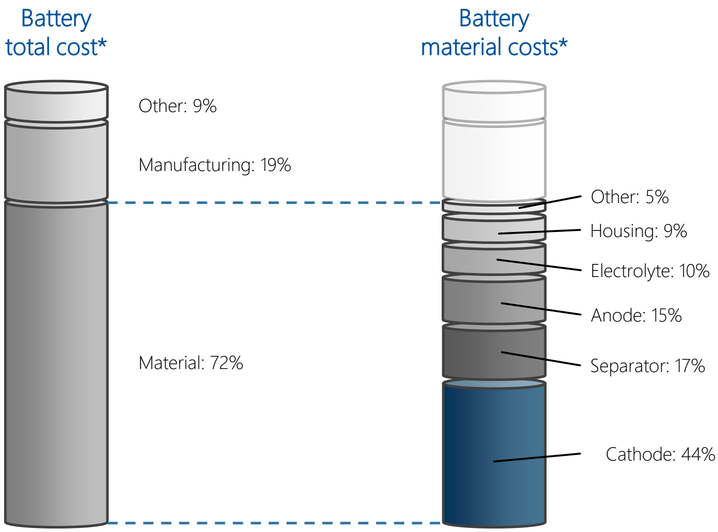


- The **battery pack** consists of several battery modules and other electrical, mechanical, and thermal components. It is connected and dimensioned differently depending on the required performance data.
- The **battery module** in turn consists of battery cells that can be connected in series or parallel.
- The **battery cell** exists in three different **cell formats**: cylindrical cells, prismatic cells and pouch cells. The general structure of the battery cell is independent of the respective cell type.
- In general, a battery cell consists of **anodes** and **cathodes** as well as the **separator,** which separates the electrodes from each other. Between them the ion-conducting **electrolyte** necessary for charge transport is located.
- The use of high-grade metals as the active material of the cathode makes it a **key component** in the field of recycling and it is therefore examined in more detail below.
- The **cathode** consists of a current conductor (often aluminum foil) and the active material, which often has one of the following compositions:
  - **NMC** (lithium nickel manganese cobalt oxide),
  - **LCO** (lithium cobalt oxide),
  - **LM(N)O** (lithium manganese (nickel) oxide),
  - **LFP** (lithium iron phosphate),
  - **NCA** (lithium nickel cobalt aluminum oxide)

Kampker et al. „Montageprozess eines Batteriemoduls und -Packs", 2018
Kampker et al. „Produktionsprozess einer Lithium-Ionen-Batterie", 2018

Cost overview

Fundamentals



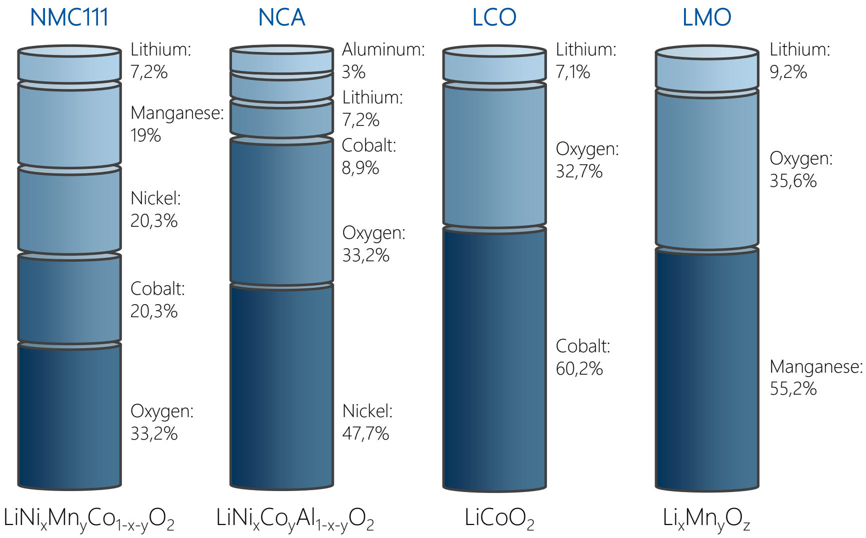
- The most expensive component of a battery electric vehicle (BEV) is the **battery pack**, accounting for up to 50% of the total cost.
- **Material costs account** for the main share of total battery costs in the manufacture of lithium-ion battery cells.
- The **cathode material** is the most expensive component of the battery cell, accounting for about 44% of the material costs.
- By using **high-quality cathode and anode materials**, there is a possibility of making the battery more powerful, more energetic, and thus more assertive.
- For the recovery of the high-grade cathode materials, **no continuous economic recycling process** currently exists industrially, and no industrial recycling solution exists for the anode materials.
- Materials from the battery system such as **steel** and **plastics** are already successfully separated from the remaining components by mechanical separation processes (via density, particle size, magnetizability, etc.), which is why these materials are not discussed in detail here.
- The raw materials **lithium, cobalt, nickel** and **manganese** are primarily the focus of current and future recycling processes, even though they only account for less than one third of the weight of the material composition of the entire battery system. In the course of the overall recycling rates of up to more than 70% to be achieved, the recovery of the anode material will also become increasingly important.

* Metastudy from the following sources: Kampker et al. "Component production of a lithium-ion battery cell," 2019; Friedrich et al. "Recovery of Valuable Metals from E-Waste and Batteries by Smart Process Design," RWTH Aachen, IME, 2019; Kwade, A., Haselrieder, W., Leithoff, R. et al. "Current status and challenges for automotive battery production technologies," Nat Energy 3, 290-300 (2018).

Cathode composition

Fundamentals

Dealing with EoL-Bat | Disassembly | Recycling



- The graph shows the mass fractions of the four **main cathode active materials** in relation to the collected used batteries in 2020.
- In addition to NMC111, **other variations** of NMC are used in BEV – e.g. NMC622 or NMC811/NCA – which have a higher **nickel content.**
- Unlike NMC, LCO and LMO consist of only one of the three metals; LFP contains iron. This results in **different properties** in terms of energy density, service life, and safety.
- LMO forms **different oxides** (e.g. LiMn2O4, Li2MnO3), so that only an average value over all mass fractions can be presented here.
- The highly **fluctuating raw material prices of these elements** are causing battery cell manufacturers to strive for raw material security, which can be achieved through new recycling technologies, among other things.
- These fluctuations also arise due to the **long supply chains**, as raw material deposits are limited to certain countries.

Projected used batteries collected, as of 2020*.

- NMC: 3,700 tons
- LCO: 2,700 tons
- LMO: 1,500 tons
- LFP: 250 tons

Disposal costs

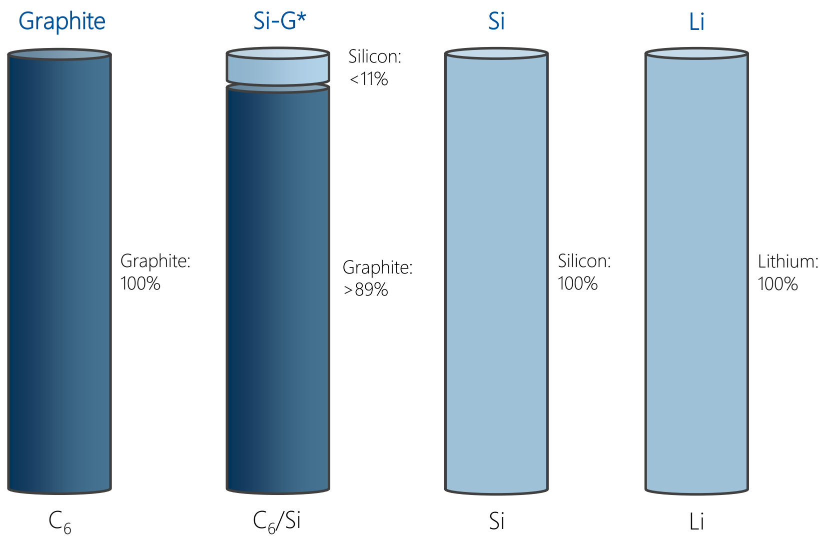
- Currently, companies that put batteries on the market have to plan for high disposal costs and build up appropriate reserves.

* Friedrich et al. "Recovery of Valuable Metals from E-Waste and Batteries by Smart Process Design," RWTH Aachen University, IME, 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

Dealing with EoL-Bat | Disassembly | Recycling

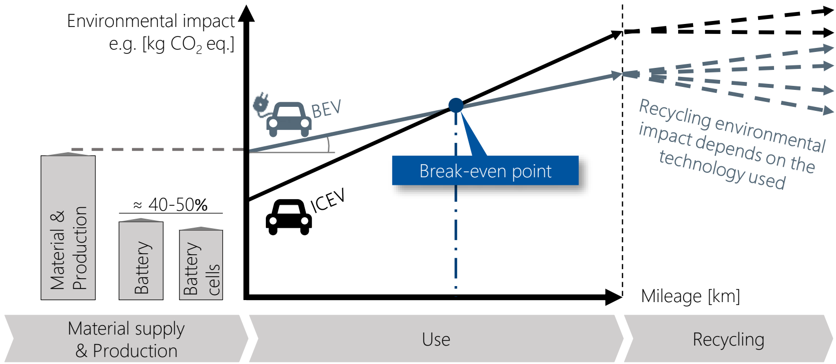


- The graph shows the mass fractions of the four **most important anode active materials** in relation to the state of the art and research. However, with regard to the collected end-of-life batteries in 2020, only the **graphite anode** is relevant.
- The mixture of graphite and silicon increases the energy density and the fast-charging capability, among other things. In the long term, the aim is to use **pure silicon** to maximize **energy density**, among other things. In addition, anodes made of lithium metal are being developed which have very high energy densities but bring with them the challenge of dendrite formation.
- **Challenges** in using pure silicon anodes include agglomeration of li-ions, formation of the SEI layer, and dendrite formation.
- If lithium is embedded in the silicon during the charging process, this results in a large **volume change** in the anode. To avoid damage to the anode, the amount of silicon added is crucial.
- To prevent the silicon anode from being damaged, it can be coated with **silicon oxide** (SiO_x). Since the use of SiO_x produces undesirable irreversible by-products such as Li_2O and Li_4SiO_4, the long-term aim is to use pure silicon.
- So far, the focus has been on the recovery of the metals within the LIB. However, the **recovery of graphite** is steadily gaining in importance.
- Through the **recycling** of graphite, it can be extracted or produced in Europe, despite the lack of natural deposits.

Chae et al. "Integration of Graphite and Silicon Anodes for the Commercialization of High-Energy Lithium-Ion Batteries," UNIST, Korea, 2019.

Life Cycle Assessment (LCA)

Fundamentals



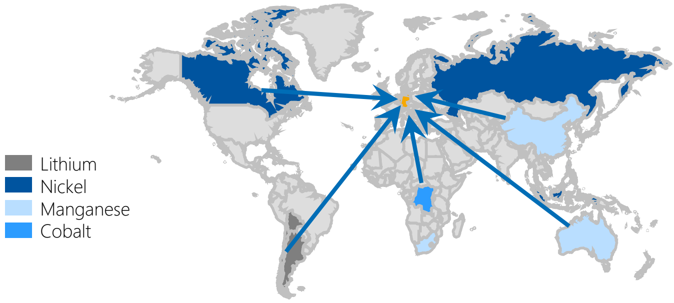
- Despite the high cost of the battery – especially due to the expensive raw materials – the **environmental aspect** should be considered intensively.
- At the beginning of their life cycle, BEVs have a larger CO_2 footprint than internal combustion engine vehicles (ICEVs) but a lower CO_2 impact in the use phase.
- The large initial CO_2 footprint results from **primary material processing** and **battery cell production.**
- The **variability** of environmental impacts during the use phase of BEVs depends on various factors and leads to different break-even points. Currently, this point is usually reached after 50,000 to 80,000 kilometers driven.
- Factors influencing the break-even point include the **energy mix** used during charging, **climate conditions** during use (season/time of day), topology, or **user behavior.**
- An energy mix with a **higher share of green electricity positively** influences the break-even point.
- In general, factors that **extend** battery **life** can **positively** influence the overall balance.
- The **recycling technology –** especially for the battery cells – also exerts a major influence on the overall environmental impact of a BEV and can affect the life cycle assessment positively in particular, but also negatively in unfavorable cases.
- An early **break-even point** and **effective recycling technology** must be achieved so that e-mobility can further reduce CO_2 emissions compared to ICEVs.

Regett et al. "Climate footprint of electric vehicles," 2018 I Agora Verkehrswende "Climate footprint of electric cars," 2019
Acatech et al. "Resource-efficient battery cycles," 2020 I Ciez et al. "Examining different recycling processes," 2019

Sustainability

Fundamentals

Dealing with EoL-Bat | Disassembly | Recycling



- In addition to the factors mentioned before, the break-even point also depends on the environmental balance of **battery material production.** At <0.01% (except for Mn), the materials are only slightly present in the earth's crust and are also not mined in Germany.
- The mining of the light metal lithium requires very **large quantities of water**, which results in falling groundwater levels in the salt lakes in the mining areas. The water depletion has a negative impact on the local agriculture and thus on the basic supply of the population.
- In addition, the extraction of these metals through **refining processes** significantly pollutes the environment. Quantitative statements about the exact proportion are difficult to make due to a lack of information.
- The **uneven distribution of raw materials** across different continents results in long transport routes, which are associated with high CO_2 emissions. For this reason, too, the further development of recycling technology to realize a local circular economy continues to gain in importance.

CO_2 emissions

- CO_2 emissions in 2019 for the production of NMC batteries for small cars were around 70-77 kg/kWh of battery capacity*. The value is strongly dependent on the energy mix used.

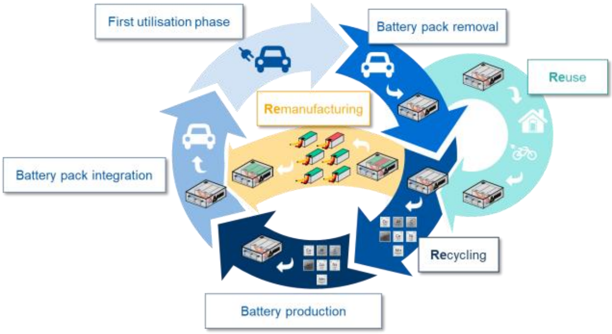
CO_2 saving

- A circular economy can greatly reduce CO_2 emissions generated by mining, refining and transporting raw materials.

Sustainability

- Improved recycling processes ensure that fewer limited raw materials will be mined in the future, lowering the price of the battery and reducing environmental impact.

* Using natural gas for heat generation in production I Emilsson et al. "Lithium-Ion Vehicle Battery Production," 2019.
Mayyas et al. "The case for recycling," 2018 I Agora Verkehrswende "Klimabilanz von Elektroautos," 2019.



- From the preceding sections, the need for a shift from the current linear economy to a **circular economy** is clear.
- The main goals of the circular economy are the unlimited reuse of materials by closing the product life cycle and the associated **reduced waste disposal** as well as the **reduction of dependence** on important primary materials.
- The classic value chain can be extended by reuse and remanufacturing. The terms reuse, remanufacturing, and recycling, which are relevant for a sustainable circular economy, can be summarized as **Re-X.**
- Since 12% of the total **greenhouse gas emissions** of EV batteries in EoL status are generated in Europe, this life cycle phase deserves special attention.
- In the following, the **handling of EoL batteries by means of recycling** is therefore explained in more detail.

Re-Use

- Lithium-ion batteries that still have sufficient residual capacity at the end of their service life in BEVs can possibly be used in other applications, e.g. stationary energy storage.

Remanufacturing

- Remanufacturing enables the extension of the first life cycle by preparing used batteries for reuse in BEVs by replacing or exchanging damaged components of the battery.

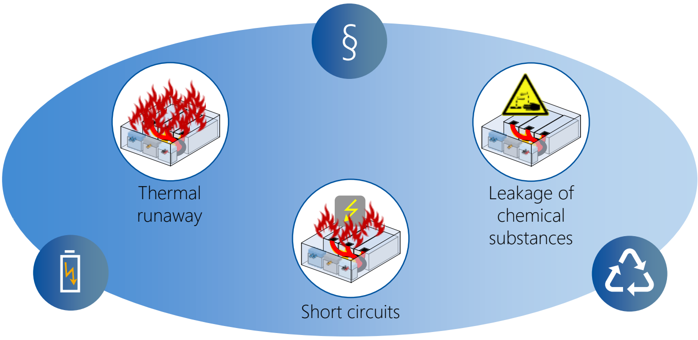
Recycling

- Recycling is required to recover raw materials from the battery through a safe process and return them to a battery production process.

Kampker et al. "International Conference on Engineering and Technology," 2018 I Emilsson et al. "Lithium-Ion Vehicle Battery Production", 2019.
Siret et al. "PEFCR for High Specific Energy Rechargeable Batteries," 2018 I Acatech et al. "Resource Efficient Battery Cycles," 2020.

Risks in handling LIB

Handling EoL batteries



- As the **demand for** LIB in electric vehicles increases, larger quantities of end-of-life batteries will be collected for recycling.
- Due to the high hazard potential, the conscious and careful handling of end-of-life batteries is particularly important.
- The existing **hazards** are:
  - Thermal runaway due to flammable substances and chemical chain reactions
  - Fire hazard due to internal or external short circuits of cells or modules
  - Leakage of chemicals, e.g. of the electrolyte, due to mechanical damage

Fire hazard

- Damage/overheating can lead to internal short-circuiting and ignition of the battery cells, with toxic gases escaping.
- The reactive decomposition products produced during battery fire can lead to an uncontrolled release of the stored energy (thermal runaway).

Short circuit hazard

- If the used batteries are not stored properly, short circuits can occur which lead to spontaneous combustion in the collection container.

Need for global security measures

- Transport and storage of the lithium-ion batteries must be carried out without damage and in compliance with international standards and guidelines.

Transport and storage

Handling EoL batteries

Fundamentals | | | | | Disassembly | Recycling

	IEC 62281	UN 38.3	ADR*	VdS 3103	End-of-life Vehicle Ordinance*
Trans-portation	Requirements for transport to ensure the safety of LIBs	Criteria and test procedures for LIB that should be met for transport	Rules for the packaging, labeling and transport of dangerous goods	No data	No data
Storage	General safety requirements for the location of LIBs	No data	No data	Instructions for handling LIB with regard to damage prevention	Action specification for storage

Lithium-ion batteries have been class 9 dangerous goods since 2009; the following guidelines and standards must therefore be observed when **transporting** LIB:

- IEC 62281: Safety of primary and secondary lithium cells and batteries during transport
- ADR compliant labeling
- UN 38.3: electrical, mechanical and thermal tests for transport safety of LIB. Reports on successful tests must be available.
- UN 3090: Lithium metal batteries
- UN 3480: Lithium-ion batteries

Lithium-ion batteries from BEVs are to be stored as hazardous materials. The following guidelines and safety measures must therefore be observed when **storing** LIB:

- VdS 3103: Loss prevention of lithium-ion batteries
- End-of-life Vehicle Ordinance: "Batteries must be stored separately in acid-resistant containers or on a drainless and acid-resistant surface."
- Safety cabinets according to EN 14470
- Fire protection container with certified fire resistance
- Storage only permitted if LIBs are tested according to UN 38.3.
- Medium capacity lithium batteries (>100 Wh and ≤12 kg) must be physically (at least 5 m) or structurally fireproof separated from other areas.

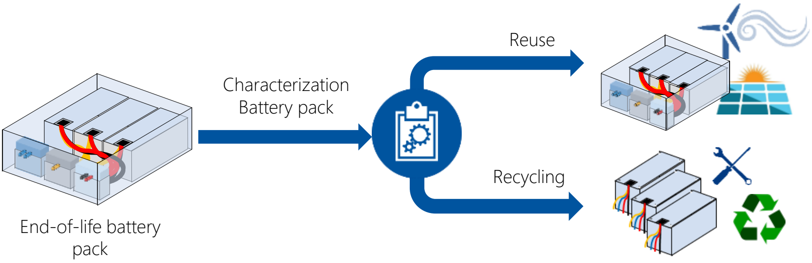
The aforementioned guidelines and safety measures apply in this form primarily in **Germany**, as each country in the EU has its **own battery legislation**.

* Legally binding
German Recycling, Battery Act, Section 5, 2020 I End-of-Life Vehicles Ordinance, 2020

Characterization of LIB

Handling EoL batteries

Fundamentals | Disassembly | Recycling



- Accurate characterization of EoL battery packs is accomplished through time-consuming and costly **electrical test methods**.
- If the usage and **condition data** from the initial usage phase is **available** in the vehicle, the battery's condition can be determined much faster and more economically.
- Since third parties are mostly unaware of the condition data so far (EU-wide battery passport is planned), economic test concepts must be developed.
- The following battery characterization **procedures** can be applied to incoming used batteries:

Incoming inspection

Quick

- At the end of the first life cycle, the LIBs are collected and inspected visually or by air pressure for obvious defects such as mechanical damage or leaks.

Data evaluation

Quick

- By evaluating the usage data such as self-discharge, charge/ discharge processes, and state variables (e.g. cell temperature), a decision can be made on the further use of the battery without any major measurement effort.

SoH determination

Time-consuming

- Capacity and power determinations are costly and time-consuming, as they require the use of expensive measuring equipment.
- By measuring the chemical and physical properties of the LIB, such as the cathodic galvanostatic pulses or the AC* measurement, the battery condition can be determined.

Resistance determination

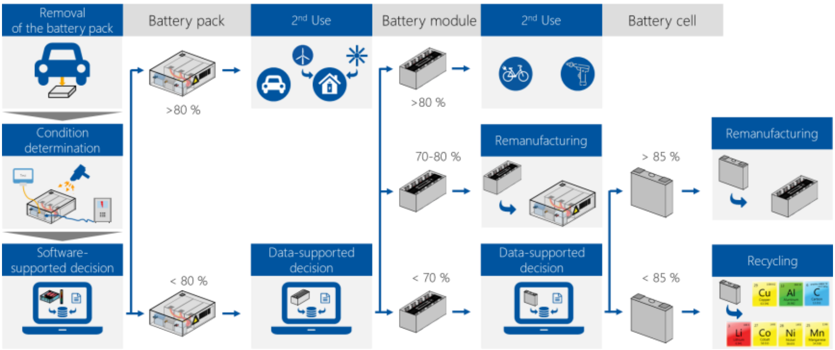
Time-consuming

- Due to various degradation processes (electrolyte decomposition, deposition on the electrode surfaces, etc.), the internal resistance increases.
- This is determined via short current pulses or with electrochemical impedance spectroscopy at high frequencies and provides information about the condition of the battery.

* Alternating Current

LIB classification

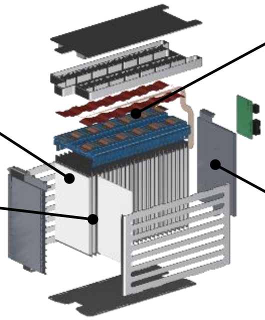
Handling EoL batteries



- With the help of various test procedures, the LIBs are classified according to their current **State of Health (SoH)** after their first phase of use and assigned to the respective applications.
- After removing the battery pack from the vehicle, **quick tests** (visual examination, internal resistance) are used to determine the battery condition and perform a **presort.**
- After the first phase of use of the LIB, the SoH is 80% on average. If the quick tests indicate that the **battery pack** is in good condition, the SoH is determined accordingly. Subsequently, the following procedure can be followed (if direct recycling is not possible):
- The pack can be used in a "2nd Use" without further treatment if the SoH is still above 80%. Otherwise, the battery packs are disassembled and characterized at module level.
- **Battery modules** with a SoH of over 80% can be transferred to "2nd use" with little effort. Modules with a SoH of between 70% and 80% are suitable for rebuilding battery packs (remanufacturing). If the modules have a SoH of less than 70%, they are dismantled and characterized at cell level if the battery module design permits.
- **Battery cells** with a SoH of more than 85% can be used to build battery modules (remanufacturing). Cells with a SoH of less than 85% are recycled.
- For economic reasons, this battery classification process should be carried out within **short test times** and with **low equipment** and personnel costs.
- To classify the LIB, the battery packs and modules must be disassembled. For this reason, the **disassembly** of LIB is explained below.

Challenges

Disassembly



- Disassembly LIB packs from electric vehicles is a **complex, time-consuming** and **cost-intensive process** due to the wide **variety of** battery pack **designs** and interconnect technologies used.
- Due to the significant differences in battery pack design – depending on the manufacturer and vehicle model – **different special tools** and a **high degree of flexibility** in the disassembly process are required.
- The **requirements** of battery cell producers and the requirements imposed by remanufacturing/recycling are very different.
- To simplify disassembly, a **new design** is needed for the battery pack, where components are assembled with detachable connections and good accessibility.
- This allows the remanufacturing and recycling process to be more efficient under controlled conditions.
- **The individual steps of the disassembly process** are described in more detail below.

Requirements of the battery cell manufacturers

- High security
- Low weight
- Low cost
- Low space requirement

in the selection of contacting and connecting components

Requirements for remanufacturing/recycling

- Avoidance of poorly soluble compounds within the modules for non-destructive disassembly at cell level
- Good accessibility of components for quick disassembly

Opening of the housing

Disassembly

Manual disassembly of the battery pack from the Renault Fluence.*

- At the start of disassembly, the **battery pack** is **discharged** and the power is temporarily stored or fed into the grid.
- Next, the battery pack **housing is opened** by, for example, removing the cover by loosening the screws along the edge of the cover.
- As the sealing rings between the battery cover and the housing differ depending on the battery type, **high forces** sometimes have to be exerted **to remove the cover**. Depending on the sealing material, the seal may be damaged during removal.
- Inside the housing are the battery modules and other components such as the cooling system and power electronics.
- The **disassembly of these components** from the battery pack is now considered in more detail.

Special tools

- Insulated tools and ESD workstations are used when working on the HV battery pack to avoid the risk of employee electrocution.

Trained employees

- Disassembly is carried out by specially trained personnel, as work is carried out at a connection voltage of 60 V DC (live working, battery risk awareness – electrician).

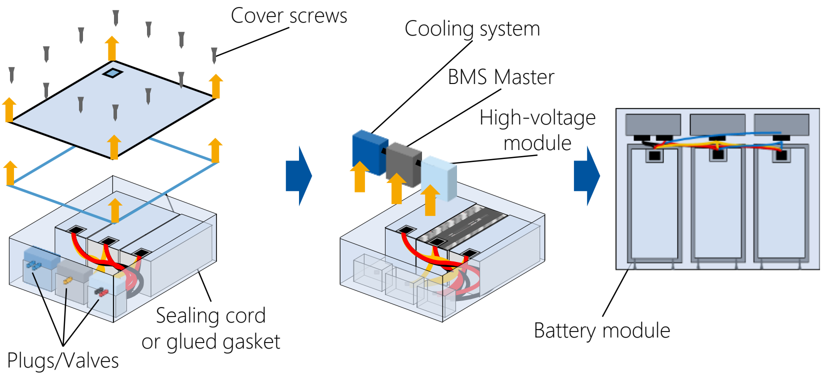
Plant/staffing requirements

- Depending on the size of the battery, the lid is removed by two people or a crane.

* PEM of RWTH Aachen University, database, 2020

Disassembly of the battery pack

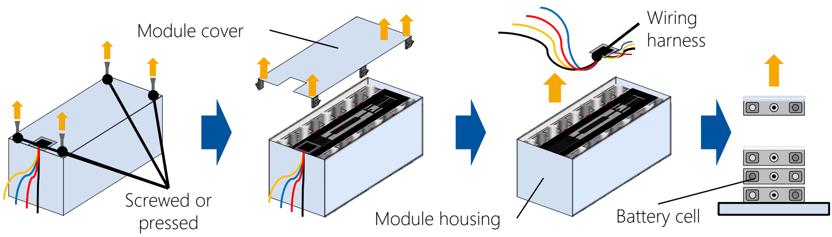
Disassembly



- After removing the housing cover, the battery components can be **disassembled** from the **battery pack**. To do this, the wiring will be removed first, then the cooling system and the high-voltage module, and finally the battery modules will be removed.
- The **cabling of** the BMS master with the cooling system, the module slave board, and the high-voltage module are removed first.
- The HV and LV harnesses connecting the modules to **peripherals** are next to be removed.
- The screwed, complex **high-voltage module** is then extracted using an insulated screwdriving tool.
- To remove the **cooling system**, it is first separated from the cooling elements of the packing case.
- For this purpose, the **coolant** should first be **removed** from the system with an extraction system in the case of liquid-cooled battery packs in order to **prevent** the coolant from leaking and thus potential **short circuits**.
- The metallic cold plates and the cooling system components can be recycled well due to their composition.
- The **battery modules** are attached to the battery pack housing via screw connections. **Additional fixation** by adhesives, foams or waxes are optionally used by the manufacturers and are correspondingly **difficult to release**.
- Depending on the design, the battery packs consist of less than ten to 48 modules.

Disassembly of the battery modules

Disassembly



- For further disassembly, the **battery module** must now be **opened** so that the battery cells can be removed.
- The **module cover** is either screwed or pressed**.** It is removed with the aid of insulated screwdrivers or a force/pressure insert.
- The **module housing is** made of **different materials** (e.g. metals or plastics) depending on the battery design. The design of the housing also depends on this.
- Metal housings consist of a deep-drawn housing tray and a cover. Plastic housings usually have an end plate and are clamped to the battery cells.
- After opening the module, the **cell contacts can be separated to remove** the cells. For safety reasons, the cell contacts are covered or taped with **insulating material**.
- In most cases, the cells in the module housing cannot be removed without damaging them because they are glued together for **thermal and electrical insulation**.

Module from prismatic cells

- The cells are braced to the housing using a bandage and then are glued.
- The cell contacts are welded and cannot be separated non-destructively.

Module from pouch cells

- The cells are inserted into a frame, tensioned by means of springs, and then are glued.
- The cell contacts are welded and cannot be separated non-destructively.

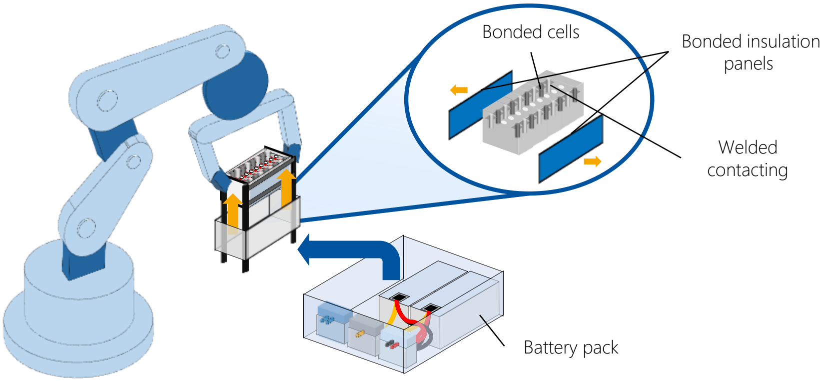
Module from cylindrical cells

- The cells are fixed in place with the aid of cell holders. The resulting spaces between the battery cells are used for cooling.
- Contact is made with one metal plate per pole, which is welded to the battery cell on both sides. When the welded connections are separated, the cell poles are damaged.

Kampker et al. "Assembly process of a battery module and pack," 2018.

Automation of the process

Disassembly



- Battery disassembly represents an **elementary process step** for both reuse and recycling of the batteries.
- Currently, only a few components are manually removed from the battery in the industry before recycling.
- In order to implement disassembly industrially and economically, the disassembly **processes** must be **partially/automatically.**
- Due to the many **different battery designs** and the use of **poorly soluble compounds,** there is as of yet no suitable process for disassembling LIB packs.
- So far, **partially automated disassembly steps** by human-robot collaborations have already been used, as they are easier to implement than fully automated disassembly.

Challenges

- Complete automation of the disassembly process is very complex to implement and therefore very cost-intensive.
- Due to the lack of process experience, there are not yet any concrete proposals for solutions, e.g. for the sensors required and for the disassembly tools for loosening the connections.

Opportunities for improvement

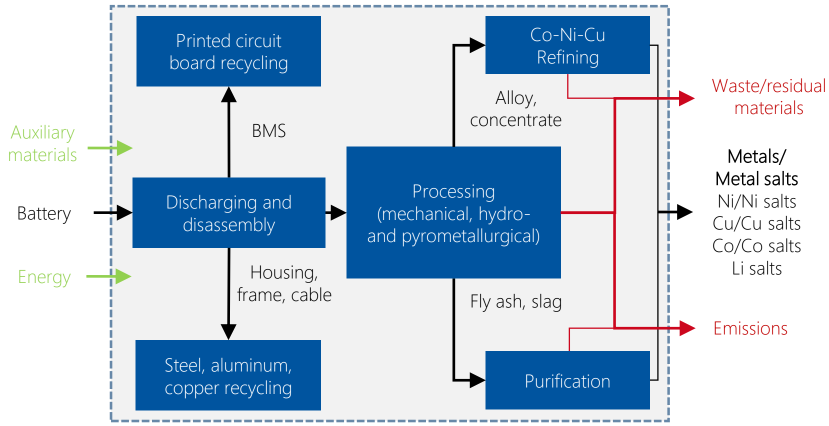
- The process can be improved with a new design of the battery pack, which is also being developed for remanufacturing/ recycling.
- Battery packs must be standardized on as broad a basis as possible so that fully automated disassembly is economically advantageous.
- Through the use of AI, automated disassembly processes will be learned and implemented.

System limits of battery recycling

Recycling

Fundamentals | Handling of EoL-Bat | Disassembly | Recycling

(*)



- The recycling process of Li-ion batteries requires batteries as input stream as well as raw/auxiliary materials and energy. It generates different output streams via several intermediate processes, especially valuable materials (metal salts), but also emissions and waste/residual materials.
- Different components require different ways of reprocessing or recycling, which should be chosen to be as energy-efficient as possible.
- The high diversity of components used in batteries leads to a complex process design, which consists of conventional recycling processes as well as more specialized processes for secondary raw material production.
- The superior goal of all processes is high recycling rates with high-value and non-contaminated output streams.
- "In order to be able to consistently determine the (usually positive) environmental effects of resulting recyclates in comparison with each other as well as with comparable primary materials, the recycling processes are to be evaluated primarily according to their energy and/or their CO_2 intensity and, if necessary, according to other environmental factors, beyond the recycling rates achieved, and the effects are to be allocated to the recyclate." (*)
- The evaluation of LIB recycling requires the holistic consideration and balancing of the applied process area.
- The balancing of the processes requires precise knowledge of the composition of the batteries as well as comprehensive life cycle analyses for the economic and ecological design of the processes.

* Circular Economy Initiative Germany (ed.): Battery Life Cycles. Driving Electric Mobility with the Circular Economy, *Kwade, A., Hagelüken, C., et al, acatech/SYSTEMIQ, Munich/London 2020.

Main process steps

Recycling

Discharging

Battery pack

↓

Electrical energy

Disassembly

Battery modules

↓

Housing, BMS, cables, busbars, screws

Cell processing

Battery module/cell

↓

Purified material fractions (e.g. copper)

Hydro-metallurgy

Active materials

↓

Co, Ni, Mn – sulfates (SO_4), Li_2CO_3

- The recycling process of Li-ion batteries after discharge and disassembly at module, cell, or electrode level consists of several sub-steps, each releasing different products/value materials.
- The process sequence requires separation of the composite components and materials after disassembly, for example by mechanical disassembly or by thermal processes.
- The complex composition of Li-ion batteries requires a large number of process steps to be combined for a high recovery rate (>90%).

Challenges

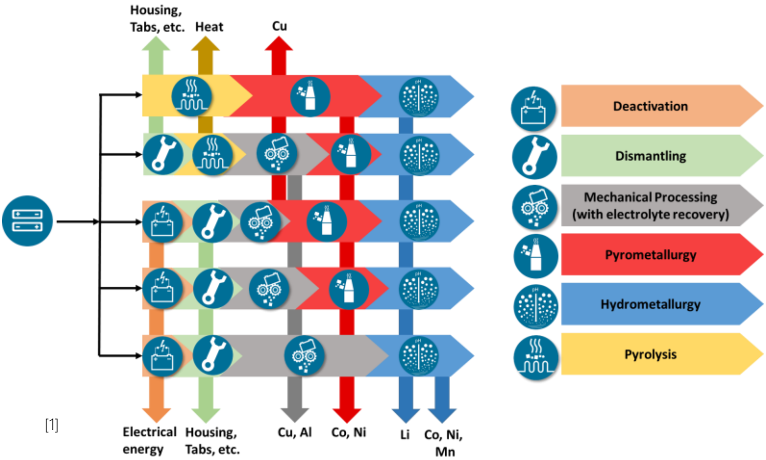
- The diversity of the components requires a large number of process steps that can be combined in different ways.
- Effective recycling of materials with a recycling rate of lithium >90% has not yet been implemented industrially.
- Current industrial technologies do not recover all recyclables.
- Recovered battery materials should have qualities that lead to reuse.

Opportunities for improvement

- *Design for recycling* approaches could reduce complexity and simplify recycling process management.
- Combination of mechanical, thermal, and hydrometallurgical process steps to increase the overall recycling rate compared to processes focused on pyrometallurgy
- Establishment of a functional, environment-friendly and safer process

Process routes in battery recycling

Recycling



- Different approaches to Li-ion battery recycling have been implemented in industry and academia: mechanical treatment, thermal treatment, pyrometallurgy, hydrometallurgy.
- The combination of recycling approaches enables diverse process routes that vary in effectiveness and have advantages and disadvantages depending on the focus of material recovery (e.g. nickel vs. lithium).
- The recycling rates for the individual recovered recyclables are largely determined by the selected combination of process steps.

Challenges

- Combinations of process steps require precise process knowledge.
- Diversity of battery packs and materials requires robust and variable processes.
- Hazards due to electrical, chemical, and thermal faults must be counteracted.
- Material losses in the processes and through transport between them must be prevented.
- Materials must be recovered to "battery grade" whenever possible.

Opportunities for improvement

- In various disassembly processes, components containing pollutants can be removed at an early stage to prevent them from being fed into the subsequent recycling process.
- Minimization of energy use
- Graphite, electrolyte, and conducting salt recovery
- Automation in the area of transfer to other processes
- Robustness of processes against process impurities

[1] 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. Metals 2021, 11, 291. https://doi.org/10.3390/met11020291

Mechanical recycling

Recycling

Shredded material

Separator

Current collectors

Black Mass

- Mechanical crushing (shredding) of battery cells and modules, if necessary small packs (exception: direct feeding to pyrometallurgy)
- Release of the recyclable materials in the form of shredded material containing all the valuable substances
- Safety risks due to electrolyte residues, which should be removed by drying or pyrolysis before the following process steps and may sometimes be available for a new use
- Mechanical separation of „Black Mass" (e.g. Co, Ni, Mn, C), current-conducting foils and separator parts by a combination of crushing, drying, sorting, and classification processes
- Further processing of the valuable materials in metallurgical and chemical processes

Challenges

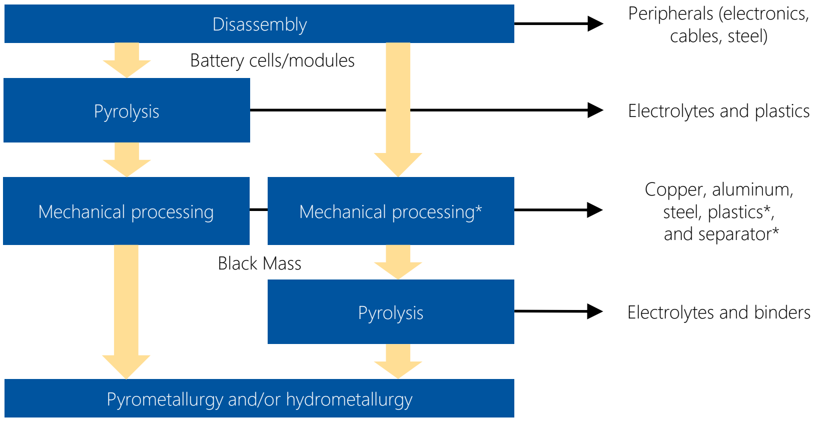
- The separation of materials by type requires a high level of process engineering effort.
- Only discharged cells can be fed into the process.
- Removal of electrolyte components is very challenging, since components of the solvents have very different boiling points.

Opportunities for improvement

- Integration of thermal steps for complete removal of electrolyte components, to generate a high safety level.
- Grade separation to improve further processing in hydro-metallurgical steps.
- Minimization of the hazard potential due to electrical, thermal and mechanical effects.

Pyrolysis

Recycling



- Prior to mechanical reprocessing, cell deactivation or treatment of the black mass during mechanical recycling can be carried out by means of pyrolysis.
- Cells are deactivated in a vacuum at up to 400 °C, where volatile electrolyte components as well as binders and polymeric components can be removed.
- Black mass can also be pyrolyzed at up to 700 °C after separation from the shredded material to remove electrolyte components as well as binders and polymeric components.
- The removal of fluorine-containing components in particular during pyrolysis positively influences hydrometallurgical processing.

Challenges

- Removal of fluorine-containing conducting salt from the cells or the black mass
- Carrying out pyrolysis with avoidance of oxidizing reactions
- Recovery of anode active material for reuse in battery applications

Opportunities for improvement

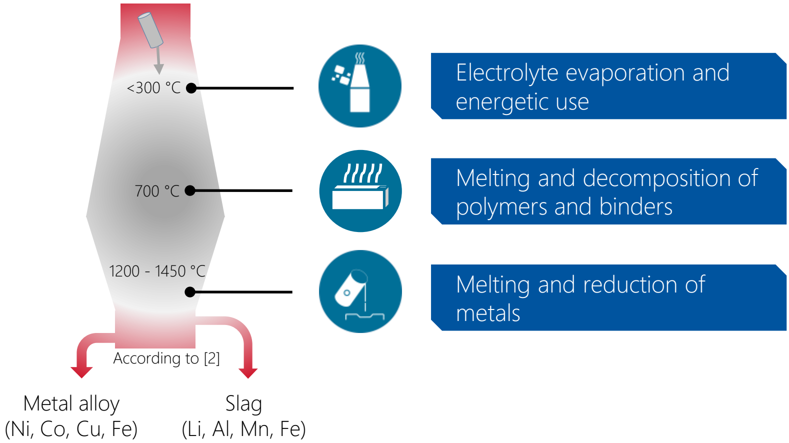
- Integration of electrolyte recovery with thermal process control
- Avoidance of HF formation from fluorine-containing substances
- Minimization of energy input during pyrolysis to reduce the ecological footprint

Pyrometallurgy

Recycling

Fundamentals | Handling of EoL-Bat | Disassembly | Recycling

Overview | Mechanical | Pyrometallurgy | Hydrometallurgy



- Pyrometallurgical processes can be performed directly with battery modules or following mechanical steps.
- Application of high-temperature processes enables production of alloys and slag containing mainly the valuable materials copper, cobalt, and nickel, as well as lithium.
- Loss of manganese and aluminum due to high temperatures
- Fluorides, which pose challenges in further processing, can be removed by high temperatures.
- Economic efficiency given with high proportions of recyclables
- High robustness against impurities

Challenges

- Recovery of valuable materials pyrolyzed in current processes (manganese, aluminum, polymers)
- Energy efficiency only given at high throughput
- Recovery of electrolyte so far not provided without mechanical processing, which provides a step for electrolyte recovery

Opportunities for improvement

- Combination with mechanical processes
- Removal of non-recoverable valuable materials before thermal application through further process steps
- Integration of electrolyte recovery to increase the overall recycling rate
- Energy-intensive process steps should be reduced to a minimum to improve the eco-balance.

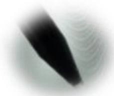
[2] M. Mohr, M. Weil, J. Peters, Z. Wang, Recycling of Lithium-Ion Batteries, Encyclopedia of Electrochemistry: Batteries, 2020, DOI: 10.1002/9783527610426.bard110009.

Hydrometallurgy

Recycling

Black Mass

- Active materials in black mass
- Cross contamination with Al, Fe, Cu possible



Acid leaching

- Dissolving of metal components
- Strong bases and acids necessary

Solid's separation

- Metal salt solution of the active materials
- Separation of solid residues

Precipitation

- Precipitation of the transition metals
- Li still in solution (LiOH, Li2CO3)

- Hydrometallurgy follows mechanical and/or pyrometallurgical processing.
- Recovery of pure non-ferrous metals from active materials
- Processed active material (black mass) is dissolved by means of acid digestion. Non-dissolved solids can be removed.
- The transition metals (nickel, manganese, cobalt) are specifically precipitated as salt and made accessible for resynthesis.
- Chemical processes used are leaching/extraction, crystallization, precipitation.

Challenges

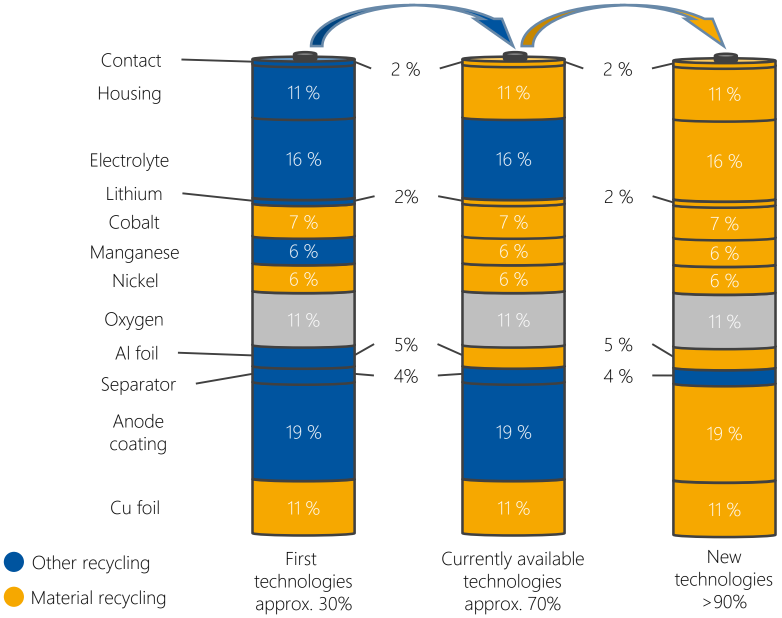
- The influence of feedstock contamination on hydrometallurgical processing and achievable material yields is not well understood.
- Cross-contamination can affect the quality and yield of the recovered material obtained and thus limit the efficiency of the recirculation.
- Scale-up of known processes for recovery of "battery grade" raw materials to industry-relevant size (LiOH, Li_2CO_3, transition metal sulfates)

Opportunities for improvement

- Avoidance of cross-contamination through upstream separation processes must be ensured.
- Increasing the robustness of hydrometallurgical processes against impurities
- Removal of fluoride ions via upstream process steps
- No complete material separation
- Graphite recovery

Process combinations

Recycling



- The combination of process technologies can perspectively increase the total recovery rate (without O_2) to >90%.
- Current available technologies can recover about 70% of the recyclables for the material cycle.
- Pure pyrometallurgical processes return approx. 30% of the valuable materials to the cycle, but Ni, Co, and Cu do so with very high yields.
- Linking the technologies increases the rate of recoverable recyclables, but also the complexity of the process.

Challenges

- The linkage requires a great deal of effort in terms of plant engineering.
- Impurities would have to be removed as early as possible in the process, as the influence is not sufficiently clarified.
- The cost structure must guarantee profitability.

Perspective

- Material loop management is essential for the EU with low resource abundance for competitiveness.
- Process combination(s) must always ensure a high overall recovery rate (at least 70%) with acceptable CO_2 emissions and at the same time a high rate for Ni, Cu, and Co (>95%).

The chair "Production Engineering of E-Mobility Components" (PEM) of RWTH Aachen University has been active in the field of lithium-ion battery technology production for many years. The field of activity covers both automotive and stationary applications. Through a multitude of national and international industry projects in companies of all value creation levels as well as central positions in renowned research projects, PEM offers extensive expertise.

The BLB is a research center of the TU Braunschweig and forms a transdisciplinary research platform for the development of circular production as well as diagnostic and simulation methods of current lithium-ion batteries and future technologies such as solid-state batteries and lithium-sulfur batteries. BLB unites 13 professorships from three universities (Braunschweig, Clausthal, Hannover) as well as battery experts from PTB, and it brings together the necessary competence along the value chain for electrochemical battery storage in Lower Saxony.

PEM
Chair of Production Engineering of
E-Mobility Components
Drill 12
52072 Aachen

www.pem.rwth-aachen.de

Battery LabFactory Braunschweig
Technical University of Braunschweig
Long Kamp 19
38106 Brunswick

www.tu-braunschweig.de/blb

Authors

PEM RWTH Aachen University

Dr.-Ing. Dipl.-Wirt.-Ing. Heiner Hans Heimes
Executive Chief Engineer
H.Heimes@pem.rwth-aachen.de

Prof. Dr.-Ing. Achim Kampker M.B.A.
Founder/Head of Institute
A.Kampker@pem.rwth-aachen.de

Christian Offermanns, M. Sc.
Chief Engineer
C.Offermanns@pem.rwth-aachen.de

Kim D. Kreisköther, M. Sc.
Group Leader Battery Components & Recycling
Kim.Kreiskoether@pem.rwth-aachen.de

Battery LabFactory Braunschweig

Prof. Dr.-Ing. Arno Kwade
Speaker Battery LabFactory
Head of Institute for Particle Technology
A.Kwade@tu-braunschweig.de

Stefan Doose, M. Sc.
Deputy Head of Research Departments
Battery Process Engineering
Institute for Particle Technology
S.Doose@tu-braunschweig.de

Marco Ahuis, M. Sc.
Research assistant
Institute for Particle Technology
M.Ahuis@tu-braunschweig.de

Dr. Peter Michalowski
Head of Research Department
Battery Process Engineering
Institute for Particle Technology
P.Michalowski@tu-braunschweig.de

Battery Production

The German Engineering Federation (VDMA) represents more than 3,200 companies in the medium-sized mechanical and plant engineering sector. The Battery Production department focuses on battery production technology. Member companies supply machines, plants, machine components, tools and services along the entire process chain of battery production: from raw material preparation to electrode production and cell assembly to module and pack production.

Abfall- und Recyclingtechnik

The VDMA Waste and Recycling Technology Association represents almost all German and European manufacturers of waste and recycling technology. Both manufacturers of complete plants and of individual components are represented in the association. The medium-sized industry has profound expertise in the field of processing and recycling of secondary materials.

VDMA
Battery Production
Lyoner Straße 18
60528 Frankfurt am Main

www.vdma.org

VDMA
Waste and Recycling Technology
Lyon Straße 18
60528 Frankfurt am Main

art.vdma.org

Authors

VDMA

Dr. Sarah Michaelis
Division Manager Battery Production
Sarah.Michaelis@vdma.org

Ehsan Rahimzei
Project Manager Battery Production
Ehsan.Rahimzei@vdma.org

Dr. Sarah Brückner
Managing Director Waste and Recycling Technology Trade Association
Sarah.Brueckner@vdma.org

Karl Rottnick
Project Manager Waste and Recycling Technology Trade Association
Karl.Rottnick@vdma.org

Frankfurt am Main, June 2021
PEM RWTH Aachen University,
BLB TU Braunschweig and VDMA
(in-house printing), 1st edition
ISBN: 978-3-947920-12-9