Recycling strategies for End-of-Life Li-ion Batteries from Heavy Electric Vehicles

Iryna Samarukha
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<tr>
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<th>Examiner</th>
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<tr>
<td>2020-09-08</td>
<td>Peter Hagström</td>
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<td>Commissioner</td>
<td>Scania AB</td>
<td>Contact person</td>
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<td>Balasubramanian</td>
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<td>Prasanth</td>
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Abstract
The master thesis tackles the problem of recycling of end-of-life Li-ion batteries from heavy electric vehicles. The comparative analysis includes review of current global situation with batteries wastes and projections of materials that may be recovered. The transportation, pre-processing and two alternatives of recycling are considered. The modelling includes the evaluation of both economic parameters (revenue streams, costs breakdown) and environmental footprint (energy consumption and sources, water consumption, and emissions breakdown). The costs analysis has shown that transportation of spent LIBs as a hazardous wastes are 5.39 €/(t cells·km) on distance up to 200 km and 3.60 €/(t cells·km) if transportation distance is over 200 km. Modelling of recycling alternatives for different battery chemistries shows that the highest revenue is generated from NMC111 batteries in the hydrometallurgical recycling, Batteries without Cobalt and Nickel in electrode composition (LMO and LFP) generate comparably low revenue due to low value of recovered materials. The negative environmental impact of hydrometallurgical recycling, particularly, in emission of GHGs, energy and water use is more higher comparing to pyrometallurgical recycling. However, hydrometallurgy results in recovery of broader spectrum of materials of high quality.

Sammanfattning
Examensarbetet hanterar problemet med återvinning av uttjänta Li-ion-batterier från tunga elektriska fordon. Den jämförande analysen inkluderar en översikt över den nuvarande globala situationen med batteriavfall och utsprång av material som kan återvinnas. Transport, förbehandling och två alternativ för återvinning övervägs. Modelleringen inkluderar utvärdering av både ekonomiska parametrar (inkomstflöden, kostnadsfördelning) och miljöavtryck (energiförbrukning och källor, vattenförbrukning och uppdelning av utsläpp). Kostnadsanalysen har visat att transport av förbrukade LIB som farligt avfall är 5,39 € / (t-celler · km) på avstånd upp till 200 km och 3,60 € / (t-celler · km) om transportavståndet är över 200 km. Modellering av återvinningsalternativ för olika batterikemikalier visar att de högsta intäkterna genereras från NMC111-batterier i den hydrometallurgiska återvinningen, Batterier utan kobolt och nickel i elektrodekomposition (LMO och LFP) genererar jämförelsevis låga intäkter på grund av lågt värde på återvunna material. Den negativa miljöpåverkan av hydrometallurgisk återvinning, särskilt i utsläpp av växthusgaser, energi- och vattenanvändning är högre jämfört med pyrometallurgisk återvinning. Hydrometallurgi resulterar dock i återvinning av ett bredare spektrum av material av hög kvalitet.
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## Nomenclature

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<tr>
<td>GHG</td>
<td>Greenhouse Gases</td>
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<tr>
<td>LIB</td>
<td>Li-ion Battery</td>
</tr>
<tr>
<td>EV</td>
<td>Electric Vehicle</td>
</tr>
<tr>
<td>EU</td>
<td>European Union</td>
</tr>
<tr>
<td>R&amp;D</td>
<td>Research and Development</td>
</tr>
<tr>
<td>VOC</td>
<td>Volatile Organic Compound</td>
</tr>
<tr>
<td>CO</td>
<td>Carbon Monoxide</td>
</tr>
<tr>
<td>NOx</td>
<td>Nitrogen Oxides</td>
</tr>
<tr>
<td>SOx</td>
<td>Sulfur Oxides</td>
</tr>
<tr>
<td>PM10</td>
<td>Particulate Matter d≤10μm</td>
</tr>
<tr>
<td>PM2.5</td>
<td>Particulate Matter d≤2.5μm</td>
</tr>
<tr>
<td>BC</td>
<td>Black Carbon</td>
</tr>
<tr>
<td>OC</td>
<td>Organic Carbon</td>
</tr>
<tr>
<td>LFP</td>
<td>Lithium Iron Phosphate battery</td>
</tr>
<tr>
<td>LCO</td>
<td>Lithium Cobalt Oxide battery</td>
</tr>
<tr>
<td>NMC</td>
<td>Lithium Nickel Manganese Cobalt Oxide battery</td>
</tr>
<tr>
<td>NCA</td>
<td>Lithium Nickel Aluminium Oxide battery</td>
</tr>
<tr>
<td>LMO</td>
<td>Lithium Manganese Oxide battery</td>
</tr>
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Introduction

Transport sector is known to be a major consumer of fossil fuels and greenhouse gases (GHG) emitter. In 2017, energy consumption of transportation sector reached 2809 Mtoe (29,0% of total global). According to International Energy Agency report, transport sector consumed 2589 Mtoe of oil products, 105 Mtoe of natural gas, 84 Mtoe of biofuels and waste, 31 Mtoe of electricity (IEA 2020). In EU, transportation sector (excluding aviation and maritime transport) is the highest emitter outside EU Emission Trading system (followed by emissions from buildings, agriculture and other sectors) and is responsible for 35% of all GHG emissions (Transport & Environment u.d.).

Improvements of efficiency, electrification of transport and use of biofuels resulted in increase of emissions from transportation sector by only 0.6% in 2018 (compared with 1.6% annually in the past decade) (IEA 2019). Heavy-duty trucks and buses have only 10% of global vehicle stock but heavy-duty vehicles emissions share in around 46% with annual growth rate of 2.2% (ICCT 2018).

In order to prevent emissions growth from heavy-duty vehicles (including buses and medium-duty vehicles), many countries introduced various policy instruments that add obligations to both heavy-duty vehicles manufacturers and users. Thus, in 2019, China set fuel consumption limits for heavy-duty vehicles that aims tighten fuel consumption in all models by 12-15% comparing to 2014 (General Administration of Quality Supervision, Inspection and Quarantine of PRC 2019). India also introduced new requirements on energy efficiency of trucks and busses that require 5% reduction of GHG emissions (ICCT 2018). Comparing to other regions Canada and US have the most ambitious targets for CO₂ reduction. 20-years plan initiated in 2010 aims to decrease GHG emission from heavy-duty vehicles by 45% till 2030 (ICCT 2018). The adoption of the Paris Agreement (United Nations u.d.) made EU leaders agree on the target of -30% GHG emissions by 2030 compared to 2005 for sectors outside the EU Emission Trading System (Transport & Environment u.d.). EU introduced CO₂ emission performance standards (EU) 2019/1242 for new heavy-duty vehicles (EUR-Lex European Union Law 2019) and mandatory CO₂ emissions monitoring for trucks with a Gross Vehicle Weight above 3500 kg (EUR-Lex Access to EU Law 2018). However, EU regulations are expected to be more strict in future to meet the requirements of Paris.

Electrification of heavy-duty vehicles is prioritized in R&D of many companies as the most feasible alternative to fossil fuels. China, South Korea and Japan focus on both development of battery electric vehicles and H₂-vehicles for highly-populated urban regions where charging infrastructure constraints limit EV expansion. However, 95% of hydrogen comes from fossil fuels conjugated with CO₂ production and, therefore, can be considered only as solution against regional pollutions in cities. Moreover, trucks fuelled with hydrogen from renewables also require batteries for extra power and energy recuperation from brakes.

Reduction of environmental footprint and operational costs are the main advantages of using electric powertrains in busses and trucks. Moreover, some heavy-duty vehicles operate on established routes that make possible to optimize charging infrastructure, introduce highly efficient truck platooning and have consistency in demand prediction. However, environmental footprint of electric vehicle manufacturing is higher than in the internal combustion engine vehicles production process due to energy-intensive steps of batteries manufacturing, especially if batteries are produced in Asia (Hausfather 2019).

The global consumption of materials is expected to double in the next forty years, while annual waste production is projected to increase by 70% by 2050. Raw materials extraction, refining and transportation cause both significant environmental and social damage. For example, over 50% of total greenhouse gas emissions and more than 90% of biodiversity loss is associated with the resource extraction and processing. With the growth of transport electrification, the extraction of materials for batteries production has both environmental and social impact related to low working conditions on mining sites and use of child labor. For example, extraction of nickel, the main component of the NMC LIB, ranked as the 9th with the highest global warming potential and as the 7th most damaging metal to human health and ecosystems (Philip Nuss 2014). Nowadays, Cobalt is mostly sourced from the Democratic Republic of Congo where the child labor is used in artisanal mining (Öko-Institut e.V. 2011). Scaling up the circular economy
potentially can contribute to the achieving of climate neutrality, ensuring security of supply and support of local economies. And last but not least, relocation of elements across Earth’s surface leads to change of electromagnetic field of the planet that has more significant effect on climate change versus rising GHG level (Cnossen 2014). Considering abovementioned, recycling of spent LIB is considered as a necessary step in all developed economies.

On a company level, particularly, from the side of vehicle manufacturer, determination and implementation of optimal recycling strategy safe companies expenses related to handling of end-of-life vehicles, reduce supply risks related to raw materials extraction and transportation, and has a positive impact on environment, society and company values. Evaluation of hidden economic value of materials in spent batteries and costs and environmental footprint of different recycling technologies is the first necessary step in the development of the recycling strategy.
1 Background

LIB recycling aims to reduce the number of batteries being disposed as the wastes due to high toxicity and harmful impact on environment and human health if battery chemicals happened to pollute water or soil. Spent LIB contain substances that are highly explosive and carcinogenic, cause acute toxicity, severe irritation and chemical burns (Undisclosed 2019). The 2006/66/EC Batteries Directive prohibits the disposal of industrial batteries in landfills or by incineration. However, residues of any batteries that have undergone both treatment and recycling in accordance with this Directive may be disposed in landfills or by incineration. Environmental footprint of recycling is shown to be lower for some batteries even with the use of well-establish technologies. For example, according to life cycle assessment of Argonne National Laboratory, the energy consumed in virgin LiCoO2 production is around 147 MJ/kg LiCoO2 versus 52 MJ/kg LiCoO2 in pyrometallurgical recycling (Jennifer B. Dunn 2012).

Recycling has high economic potential. Thus, revenues from recycling has the potential to reduce the BEV & PHEV manufacturing costs due to cost reduction on battery production (20-30% of vehicle costs) (Melin, State-of-the-art in reuse and recycling of lithium-ion batteries – A research review 2019). Currently, gate fee, a fee that battery owner pays to a battery recycler for recycling and wastes handling, is the main source of revenue for recyclers and battery (or vehicle) manufacturers are responsible for this costs. Policy regulation and subsidies in EU could lead to organizational and technological development in recycling and make recycling self-sustainable due to declined operational costs and increased value of recovered materials. However, existing business models should consider: optimization of spent LIB processing technologies; proof-of-concept of existing solutions, scale-up and commercialization; operational cost reduction via automatization and increased materials recovery with respect to battery chemistry; investments in LIB recycling; optimization of collection schemes; and quality and circulation of recycled materials.

Furthermore, the increasing demand on raw materials for batteries production is the challenge in long-run prospective. Lithium, cobalt, nickel and artificial graphite are considered to be the critical materials and evaluation of recycling potential is important for security of supply. Covid19 shock showed that the global materials supply value chain is vulnerable and will cause the divestment from developing countries and focus on local supply and materials circularity. A projection of lithium demand in US showed that, in optimistic scenario for penetration of EV, recycled materials will start significantly impact on shortages of raw materials extraction by 2035 (Figure 1) (Linda Gaines 2018). According to projections of International Energy Agency, materials from battery wastes can cover 6.5% of all materials demand by 2030. By 2040, around 50% of all materials for LIB production may be from recovered in recycling (IEA 2020).

Recycling and refining materials from battery wastes has significant potential to decrease extraction of virgin materials. However, recycling of LIB as all other technologies should be considered in dimensions of economic, environmental and social impact. Moreover, volatility of market price of virgin materials, COVID19 economic crisis, political risks and upcoming battery recycling regulations may add new constrains and challenges.
1.1 Recycling potential of spent LIB

Li-ion batteries (LIB) are the cells assemblies with housing, control electronics and wires, and a cooling system. LIB cells design comprise a wide variety of cathode and anode systems, electrolytes and other components. There are five major LIBs cathode chemistries available on a market nowadays (Table 1): lithium-cobalt oxide (LCO) (Julien 2016), lithium-nickel-manganese-cobalt (NMC) (Noh, o.a. 2013), lithium-manganese oxide (LMO) (Jiang, Wang och Zhang 2016), lithium-nickel-aluminium oxide (NCA) (Doeff 2013) (Zhou, o.a. 2017), and lithium-iron phosphate (LFP) (Larouche, o.a. 2020).

<table>
<thead>
<tr>
<th>Cathode Material</th>
<th>LCO</th>
<th>NMC</th>
<th>LMO</th>
<th>NCA</th>
<th>LFP</th>
</tr>
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<tr>
<td>Average potential (V vs. Li^0)</td>
<td>3.7–3.9</td>
<td>3.3</td>
<td>3.8</td>
<td>3.8</td>
<td>3.3</td>
</tr>
<tr>
<td>First cycle discharge capacity (mAh/g at 0.1 C)</td>
<td>140</td>
<td>140–200</td>
<td>120</td>
<td>180–200</td>
<td>155–160</td>
</tr>
<tr>
<td>Specific energy (Wh/kg)</td>
<td>520</td>
<td>560</td>
<td>455</td>
<td>680–760</td>
<td>560</td>
</tr>
</tbody>
</table>

Figure 2 (A) shows the availability of end-of-life LIB for recycling by battery chemistry in 2018-2025. LCO batteries have been widely used in electronics and today reach their end-of-life and are already available for recycling. Recycling of LCO is highly profitable because of 17% content of Cobalt. High demand and scarcity of Cobalt resulted in the increase of Cobalt price by 300% up to 95400 $/t (Trading Economics 2020). Therefore, the automotive LIB market key trend is in the increased use of NMC batteries with low content of Cobalt and no Cobalt such as LFP and LMO batteries (Melin 2019). The main reason is in a massive decline in NMC battery prices in the last few years. Starting from 2015, when NMC became major LIB chemistry in the worldwide LIB market (~29%), followed by LCO (26%) and LFP (23%), interest in NMC batteries is continuously growing. Major global car manufacturers announced the application of NMC batteries in mass-produced EVs. For example, Tesla in China uses LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2 (NMC622) batteries supplied by CATL (Shirouzu och Lienert 2020). Both Tesla and Audi buy NMC batteries (LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2 (NMC111), LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2 (NMC622), LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2 (NMC811)) from LG Chem (Shirouzu och Lienert 2020) (LG Chem 2020). Therefore, the share of end-of-life NMC LIB is expected be higher comparing to other chemistries.

In 2018, over 68% of LIB recycling was held in China (Bernhart 2019), around 19% in South Korea and less than 5% were in EU (Figure 2, B). However, after adoption of the waste import ban in 2018 (UN
Environment 2018) electronic wastes from outside China are not accepted for recycling in China due to high negative environmental impact and now exported waste batteries are recycled in South Korea which makes recycling more expensive due to higher price on a workforce.

Obviously, the amount of recycled materials depends on the battery chemistry. Table 2 shows a comparison of content of different materials in LIB cells of various battery chemistries (Melin 2019). However, the choice of recycling technology determines the recovery efficiency of available materials and, therefore, potential revenue.

<table>
<thead>
<tr>
<th>Material</th>
<th>Price, USD/kg</th>
<th>Mass content in a cylindrical cell, %</th>
<th>Economic value of cell materials, USD/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Casing</td>
<td>NMC111</td>
<td>NMC253</td>
</tr>
<tr>
<td>Casing</td>
<td>Steel</td>
<td>0.29</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>Aluminium</td>
<td>1.8</td>
<td>10</td>
</tr>
<tr>
<td>Current Collectors</td>
<td>Aluminium</td>
<td>1.8</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>Copper</td>
<td>6.0</td>
<td>7</td>
</tr>
<tr>
<td>Anode Material</td>
<td>Graphite</td>
<td>1.2</td>
<td>18.1</td>
</tr>
<tr>
<td>Cathode Material</td>
<td>Manganese</td>
<td>2.4</td>
<td>6.1</td>
</tr>
<tr>
<td></td>
<td>Lithium</td>
<td>70.0</td>
<td>2.3</td>
</tr>
<tr>
<td></td>
<td>Cobalt</td>
<td>30.0</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td>Nickel</td>
<td>12.0</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td>Aluminium</td>
<td>1.8</td>
<td>0.4</td>
</tr>
<tr>
<td></td>
<td>Iron</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>Total value, USD/kg</td>
<td>5.42</td>
<td>5.02</td>
<td>5.19</td>
</tr>
</tbody>
</table>

Selection of recycling technology is a complex task that should take into consideration:

- recycling and collection facility location;
- transportation costs and charges;
- Pre-processing costs (disassembly and discharge);
- recycling efficiency and price;
- environmental and social impact of recycling;
- battery chemistry, state of health and state of charge.

Consolidation of information about available end-of-life LIB is beneficial for highly efficient consolidation materials streams and develop optimal business models. This will impact on development of recycling and refinery industry and keep the recycling costs down. Universal materials database also add value to materials supply industry and may secure the raw material reserves.

**1.2 Regulations for handling of end-of-life automotive LIB**

The 2006/66/EC Batteries Directive (EUR-Lex 2006) regulates automotive LIBs. Moreover, spent LIB handling from automotive transport is tackled in the regulation on road transport (ADR), product design and waste management regulation (Figure 3).

According to the Battery Directive Lithium-ion batteries are not specified as a separate battery type and fall under the category of industrial batteries and accumulators. Battery producers (or vehicle manufacturer if cells are produced outside EU) are obliged to take back waste batteries from end-users regardless of “chemical composition and origin”. Producers are also responsible for financing the end-of-life management of the products put on the market. The 2006/66/EC Batteries Directive defined the battery collection rate of 45% by 2016 as the percentage of mass of batteries collected in the calendar year in comparison to the average mass of batteries put on the market during the last three calendar years. Direct
disposal of spent LIB by means of landfill and incineration is prohibited. However, the treatment and recycling may be done outside of the EU. There is no focus on LIB, however, the new Battery Directive is expected to be adopted by the end of 2021 or beginning of 2022.

Recycling companies often propose full cycle of end-of-life LIB battery handling, from collection to recycling. However, so far, the number of spent EV LIB is negligible and collection is managed by car scraping and dismantling companies. Despite the still low volume, EV LIB in Sweden are currently collected, pre-processed by El-Kresten, a Swedish non-profit organization and one of the two main waste electrical and electronic equipment collection companies in Sweden. Stena Recycling AB, another waste electrical and electronic equipment collector, that is also looking into LIB recycling; however, it is still unclear which stage of the process the company will focus on. In Denmark, two companies, El Retur A/S and ERP, are responsible for battery collection, including LIB. The Producer Responsibility legislation in Denmark is supervised by the national authority, the Danish Product Responsibility System. The main battery collection service in Norway is provided by BatteriRetur AS, which handles discharging, dismantling and second life assessment. The large number of batteries in Norway has accelerated expansion of the collection market with more companies, such as Revac AS, Norskrisk, and ERP.

All the batteries collected in Netherlands, Belgium and Luxemburg are mainly recycled by Umicore in Antwerp, Belgium. Umicore invests heavily in technology development and in the expansion of the battery recycling industry. In 2018-2019, the company signed partnerships with Audi, BMW, LG Chem and Northvolt to expand their business across Europe. In France, the national regulations on Batteries are framed in the French Environmental Code, accompanied by three ministerial orders. The Environment agency ADEME closely monitors the waste battery systems and treatment facilities. UK and Ireland export spent LIBs to mainland Europe for recycling. Nevertheless, Japanese car-maker Nissan announced a launch of the production line of energy storage systems for households in UK which use second life batteries that are no longer fit for EVs. German legislation classifies Li-ion batteries as “other batteries”, which are generally classified as non-hazardous. In 2015, after strong criticism, the Committee for the Environment, Nature Conservation and Reactor Safety proposed an amendment to the legislation to treat Lithium-ion batteries as a separate type of battery. Due to concerns about competitive disadvantage of German recyclers caused by stricter regulations, the German Federal Council did not adapt this change. Nevertheless, the federal states of Germany have the right to tighten the national law inside their respective regions (ReCell 2020).
1.3 Collection and Transportation

According to the 2006/66/EC Batteries Directive (EUR-Lex 2006), EU battery manufacturer or EU EV manufacturer (if batteries are imported from outside of EU) are responsible for batteries recycling and disposal. Battery collection can be done by battery manufacturer/automotive company and can be delegated to the third party for organization of battery collection and recycling.

As a highly-flammable product, the transport of LIBs is highly regulated. International Carriage of Dangerous Goods by Road (ECE/TRANS/275 2019) also known as “ADR”, which regulates the packaging of high voltage LIBs to ensure safety during road transport. All batteries need to be individually packaged with an inner packaging that is covered with insulative non-combustive, non-conductive material in the empty space between batteries to prevent contact and an outer packaging to prevent excessive movement. In addition, defective batteries must be packed separately in a leak proof packaging with additional materials that can absorb leaking electrolyte. LIBs are classified as dangerous goods but not as hazardous waste, unless damaged or defective. Since the 2019 update of the ADR, LIBs are split into three categories, each with a different marking: new cells; defective or damaged cells; and batteries carried for disposal or recycling. The packaging requirement is complex, and especially complicated due to the high weight of LIB packs of up to 500 kg.

Figure 4 shows locations of collection and recycling companies in EU as of June 2020. Transportation of end-of-life LIB is considered non-hazardous in EU member states. However, EU can follow the US regulatory innovations trend and classify LIB as hazardous which will have a significant impact on transportation costs.

1.4 Pre-processing

Pre-processing may include LIB sorting, dismantling and discharge. Table 3 shows summary of pre-processing steps for different processes implemented by recycling companies or processes under development.

Sorting and size reduction (dismantling) need depends on recycling technology used. The main advantage of pyrometallurgical processes is that sorting and dismantling till cell level are not necessary because a mixture of LIBs (and even NiMH batteries in a bulk) can be recycled. Hydrometallurgical method requires sorting (and sometime storage of sorted batteries) which increase recycling costs and safety risks. As for today, automotive LIBs are manually dismantled and sorted (Mengyuan Chen 2019) therefore is the most labour-intensive step in recycling. Moreover, workers need to be highly qualified to manage safety risks due to high voltage and thermal runaway.
Table 3: Pre-processing steps for existing recycling processes

<table>
<thead>
<tr>
<th>Company / Process</th>
<th>Pre-Processing</th>
<th>Company / Process</th>
<th>Pre-Processing</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aalto University</td>
<td>-</td>
<td>OnTo</td>
<td>Discharge, Dismantling</td>
</tr>
<tr>
<td>Accurec</td>
<td>Sorting Dismantling</td>
<td>Recupyl Valibat</td>
<td>-</td>
</tr>
<tr>
<td>Akkuser</td>
<td>Sorting</td>
<td>Retriev Technologies</td>
<td>Dismantling</td>
</tr>
<tr>
<td>Battery Resources</td>
<td>Discharge</td>
<td>Sumitomo–Sony</td>
<td>Sorting, Dismantling</td>
</tr>
<tr>
<td>LithoRec</td>
<td>Discharge, Manual disassembly</td>
<td>Umicore ValEas™</td>
<td>Dismantling</td>
</tr>
</tbody>
</table>

There is some progress in automatization of sorted batteries. For example, computer vision algorithms were used to recognize labels on batteries, and then pneumatic actuators to segregate batteries into different bins according to their type of chemistry. Recent algorithms capable to recognize objects and materials on the basis of features such as size, shape, colour and texture. However, labels, QR Codes, RFID tags or other machine-readable features on key battery components and sub-structures with information about the chemical composition could simplify sorting process.

![Figure 5: Dismantling problems on different steps of vehicles LIB Disassembly (Harper, o.a. 2019)](image)

However, it’s a company decision to disclose the information that usually is the trade secret or patented data. Moreover, the creation of open source database for recycling requires the data format harmonization and reliable data protection technologies (Harper, o.a. 2019). The blockchain technologies are proposed to be applied to provide whole-life-cycle tracking of battery materials, including information and transparency on materials sources, ethical supply chains, battery state of health and utilization history (Bazilian 2018).

Only few countries in the world look into labelling standards for electric-vehicle batteries with respect to battery recycling. For example, the Society for Automotive Engineers and the Battery Association of Japan...
recommended labelling standards for electric-vehicle batteries. However, international cooperation in this field is expected to be even more beneficial. Thus, the launch of Battery Passport project was announced at the 50th annual meeting of the World Economic Forum in Davos this year. Battery Passport, a lifetime history and real-time monitoring service for EV batteries (Battery Passport 2020), was proposed by the Global Battery Alliance that was launched in 2017 and includes Google, Microsoft, Volkswagen, Honda, Enel, Umicore, ERG, the World Bank, UNICEF, OECD, UNEP and a number of other global industrial and policy players. However, fast implementation of the Battery Passport faces with the abovementioned challenge of information asymmetry, particularly, willingness or ability to volunteer data that are useful for recyclers (Forbes 2020).

Disassembly robotization of LIB packs is under development (Ian Kay 2020), however, there are very few working prototypes. The main challenges of automatic and semi-automatic dismantling are in variety of pack designs, no pack design optimization for recycling and lack of harmonized dismantling and testing protocols (Figure 5). Therefore, development of intelligent and flexible algorithms for robots operation are the key to success. Another challenge is in development of sensing systems for advanced robotic perception. Robot’s recognition system usually includes computer vision using three-dimensional RGB-D imaging devices, bespoke sensors from materials and battery experts, tactile and force-sensing, etc (Harper, et al. 2019).

There are several options for end-of-life LIB discharging. The choice of discharging method depends on LIB size and state of charge (SOC) (Figure 6). Discharge in 5 wt. % water solution of Na₂CO₃ and metal powder is cost efficient, safe, non-corrosive, and easy for robotization way for cells of any SOC and modules and packs with voltage under 500 V and low SOC. For large battery packs and modules the resistive (direct Ohmic) discharge with energy recovery is usually used in industry because of safety, minimal environmental impact, and energy efficiency. The revenues resistive discharge is quite modest and hardly can cover investments costs. For example, 60-kWh LIB pack at a 50% SOC and a 75% SOH has a potential 22.5 kWh for end-of-life recovery. However, process is associated with intense energy dissipation that requires cooling systems of flexible design. Process automatization is mandatory due to high risks of manual operations behind the battery management system (Nembhard 2019).

![Figure 6: LIB Discharging Decision Matrix](image)

However, in-process stabilization during opening is preferred in industry nowadays, as it minimizes costs. This method is particularly essential for physical LIB processing in the Recupyl (Tedjar 2013), Akkuser (Pudas 2010), Duesenfeld (Hanisch 2019) and Retriev (Smith 2013) processes. Large-scale EU recyclers stabilize LIB cells opening with CO₂ or Ar (with O₂ content less than 4%). CO₂ forms a passivating layer of lithium carbonate on lithium metal. The Retriev process (US/Canada) uses water hydrolysis of Li as a heatsink to prevent thermal runaway. The in-process stabilization is used for the damaged items.
1.5 Recycling

Nowadays, the capacity of recycling facilities in EU is approximately 30 000–40 000 tons of LIBs per year (Elementenergy 2019). The biggest facilities are located in Germany (Redux, Accurec, Duesenfeld, Volkswagen), Belgium (Umicore), Finland (Akkuser) and France (EDI, SNAM) (Figure 4, B) (ReCell 2020).

Recycling companies use pyrometallurgical and hydrometallurgical methods or their combinations (Table 4). Mechanical treatment (crushing, shredding, milling, grinding, gravity and magnetic separation, etc.) may be applied as a preliminary step depending on technology and equipment. For example, efficient mechanical processing steps results in high recycling efficiency (>90%) and low energy consumption (0.3 kWh/kg) of Akkuser process. High complexity of mechanical pre-processing decreases viability and costs comparing to simpler recovery processes. For example, the process developed in Aalto University achieve high quality and efficiency of materials recovery. However, it requires the efficient mechanical pre-processing steps, high energy consumption in pyrometallurgical step and various reagents in the hydrometallurgical recovery steps.

A shift from pyrometallurgical process toward combination of mechanical pre-processing and hydrometallurgical method is in line with the circular economy idea because it increases variety and usability of materials in a close-loop cathode production.

Majority of the state-of-art recycling processes were not initially designed for end-of-life LIB processing and are used for the recycling of other battery chemistries. However, growing share of LIBs in battery waste mix and high value of materials pushed for a redesign of recycling processes for LIB streams Umicore and Recupyl.

<table>
<thead>
<tr>
<th>Company Name</th>
<th>Location</th>
<th>Capacity [t/year] (in 2020)</th>
<th>Technology</th>
<th>Recovered Elements</th>
<th>Losses</th>
</tr>
</thead>
<tbody>
<tr>
<td>Umicore</td>
<td>Antwerpen, Belgium</td>
<td>7000</td>
<td>Pyrometallurgy (shaft furnace); hydrometallurgy (leaching solvent extraction)</td>
<td>Co, Ni, Cu, Fe, CoCl₂</td>
<td>Slag: Al, Si, Ca, Fe, Li, Mn</td>
</tr>
<tr>
<td>Akkuser</td>
<td>Nivala, Finland</td>
<td>4000</td>
<td>Mechanical processing (1st cutting, air filtration, cutting, magnetic separator)</td>
<td>Co, Cu powder, Fe</td>
<td>Non-ferrous metals</td>
</tr>
<tr>
<td>Duesenfeld</td>
<td>Wendeburg, Germany</td>
<td>3000</td>
<td>Mechanical, thermodynamic and hydrometallurgical processes</td>
<td>CoSO₄, NiSO₄, MnSO₄, Li₂CO₃, graphite</td>
<td>Electrolyte, Co, Al</td>
</tr>
<tr>
<td>Accurec Recycling GmbH</td>
<td>Krefeld, Germany</td>
<td>2500</td>
<td>Mechanical processing (Milling, separation, agglomeration, filtration, ambient); pyrometallurgical (vacuum thermal treatment, reduction); hydrometallurgical (H₂SO₄)</td>
<td>Li₂CO₃, co-alloy</td>
<td>Metallic alloy</td>
</tr>
<tr>
<td>EDI (Sarpi Veolia)</td>
<td>Dieuze, France</td>
<td>1080</td>
<td>Mechanical processing (grinding), cold hydrometallurgical</td>
<td>Cu, Al, Ni, Co, Mn, alloys, Li₂CO₃</td>
<td>Slag</td>
</tr>
<tr>
<td>SNAM SAS</td>
<td>Viviez, France</td>
<td>300</td>
<td>Pyrometallurgy and hydrometallurgical (under development)</td>
<td>Ni, Co, Fe</td>
<td>Slag with precious metals</td>
</tr>
</tbody>
</table>
Some processes, such as Sumitomo–Sony and Akkuser, were developed for LIBs with high content of Cobalt, one of the most expensive precious metal on the market. However, mass-produced EV LIB tend to shift from high Cobalt-content technologies due to element scarcity. Another challenge is that LIB with different SOH should be processed separately (Velázquez-Martínez, o.a. 2019). The battery recycling company Akkuser Oy receives end of life batteries from several countries in Europe such as Sweden, Norway, Spain, Denmark and Austria. The company recycles and processes hazardous materials from Lithium in a crushing process, sending the cobalt to refineries in Kokkola, where the mining industry is located. Other recycling companies, such as Crisolteq, recycle batteries through a hydrometallurgical process that treats the black mass (a mix of materials and chemical elements) on industrial scale.

### 1.5.1 Pyrometallurgical LIB recycling

Pyrometallurgical recycling is the major commercialized technology nowadays. Pyrometallurgy involves application of heat for metals extraction and purification. Umicore (Belgium), Accurec (Germany), Batrec (Switzerland), Nickelhütte Aue GmbH (Germany) use pyrometallurgical process which is followed by hydrometallurgical steps to extract valuable metals from the black mass.

Figure 7 shows a process diagram of generic pyrometallurgical LIB recycling. Pre-processed (shredded or intact) LIBs first are processed in a smelter where LIB electrolyte and plastics are burned for energy, and carbon, graphite and aluminium are oxidised in a reduction reaction with the metals. Iron, copper, nickel, and cobalt are sedimented in matte and rest of materials, including Al₂O₃ end up in a slag. The matte is leached with the acids with further extraction and precipitation of cobalt and nickel compounds. Lithium can be potentially extracted from slag. However, due to lack of data lithium extraction is not included.

Co and Ni compounds can be used for the cathode production. Slag also may be used for cement production or as an aggregate for pavement (Argonne National Laboratory 2019). An overage materials recovery efficiencies for pyrometallurgical recycling are following: Copper – 90%; Iron – 90%; Co²⁺ in output – 98%; Ni²⁺ in output 98% (Li Li 2018) (Chunwei Liu 2019) (Argonne National Laboratory 2019). Lithium compounds and aggregate (from slag) recovery efficiency are not included in the analysis due to insufficient data. Aluminium usually is landfilled in a slag. All other components are burned for energy.
Attachment 1 shows the technological process with the assumed equipment for generic pyrometallurgical recycling of end-of-life LIBs.

1.5.2 Hydrometallurgical recycling of LIB

Hydrometallurgical recycling uses water as a solvent to extract and recover valuable elements from various complex mixes of compounds. There is a growth of interest in hydrometallurgical approach in recycling because it is low-cost, energy efficient and has proven to have low environmental footprint comparing to direct physical and biological methods (Siqin Xiong 2020). However, these parameters differs a lot for the various inlet complexity, reagent schemes, recycling efficiency, effluent toxicity, and water consumption (Larouche, o.a. 2020).

Sulphate acid is the most common leaching agent in hydrometallurgical recycling. H₂SO₄ leaching has the following reaction with Co-, Ni-, and Mn-based active materials (Meshram, Pandey och Mankhand 2014):

\[
2\text{LiM}(\text{III})\text{O}_2(s) + 3\text{H}_2\text{SO}_4 \rightarrow 2\text{MSO}_4(aq) + \text{Li}_2\text{SO}_4(aq) + 3\text{H}_2\text{O} + 0.5\text{O}_2(g)
\]  

where M is Co, Ni, or Mn.

Hydrogen peroxide is the most popular reducing agent which reacts with cathode materials as following:

\[
2\text{LiMO}_2(s) + 3\text{H}_2\text{SO}_4 + \text{H}_2\text{O}_2(aq) \rightarrow 2\text{MSO}_4(aq) + \text{Li}_2\text{SO}_4(aq) + 4\text{H}_2\text{O} + \text{O}_2(g)
\]  

Hydrochloric acid is another system that was studied for application in hydrometallurgical recycling. There are two alternatives of reaction with evolution of oxygen (Meshram, Pandey och Mankhand 2014) and chlorine gas (Larouche, o.a. 2020):

\[
2\text{LiM}(\text{III})\text{O}_2(s) + 6\text{HCl}(aq) \rightarrow 2\text{M(II)Cl}_2(aq) + 2\text{LiCl}(aq) + 3\text{H}_2\text{O} + 0.5\text{O}_2(g)
\]

\[
2\text{LiM}(\text{III})\text{O}_2(s) + 8\text{HCl}(aq) \rightarrow 2\text{M(II)Cl}_2(aq) + 2\text{LiCl}(aq) + 4\text{H}_2\text{O} + \text{Cl}_2(g)
\]

HNO₃, organic acids, and other mineral acids or alkaline leaching agents are used in a leaching step.

Figure 8 depicts a process diagram for a generic hydrometallurgical recycling. Spent LIBs after preliminary discharging and disassembly are shredded and undergo a low temperature calcination. On this stage binder and electrolyte are burned off. Other materials undergo several physical separation steps to segregate aluminium, copper, steel as metal scraps and plastics. Then black mass is leached, extracted with solvents with following precipitation of cobalt, nickel and manganese compounds, and potentially Li₂CO₃.
For both pyrometallurgical and hydrometallurgical recycling the exhaust gas treatment is necessary step to remove fluoride emissions, the product of combustion and/or decomposition of the electrolyte.

An average materials recovery efficiencies for hydrometallurgical recycling are following: Copper – 90%; Steel – 90%; Aluminium – 90%; Graphite – 90%; Plastics – 50%; Lithium carbonate – 90% (Li⁺ equivalent in output); Co²⁺ in output – 98%; Ni²⁺ in output – 98%; Mn²⁺ in output – 98%; electrolyte solvents and salts – 50%. Around 50% of plastics and electrolyte are burned for energy. Carbon black and PVDF are landfilled.

There are many research regarding the use of combination of physical methods for recycling such as direct and indirect physical recycling which allows to avoid change of chemical composition of battery components and, thus, directly recover electrode materials. Industrial biotechnology proposes the application of bacteria for bioaccumulation and extraction of valuable materials and the use of organic acids or enzymes in bioleaching process. However, all these projects are lab-scale or under prototype development and due to lack of industrial operation data are not included in analysis. Nevertheless, batteries recycling is an evolving field and evaluation of innovative technologies should be included in future analysis.
2 Objective

The main goal of the research is to perform techno-economic analysis and comparison of end-of-life Li-ion batteries recycling technologies with respect to the battery chemistry and state-of-health, module and pack design, destiny and revenue from recycled materials, environmental impact.

Following tasks were determined as necessary steps to achieve the goal:

- determine methodology of research: develop a model for the calculation of recycling revenue and environmental footprint and determine system boundaries;
- analyse input parameters: chemical composition of Li-ion batteries, materials of modules and packs;
- compare main recycling technologies and recycling efficiency for Li-ion batteries of different chemistries with technological inputs and outputs;
- analyse the market of recycled materials and revenue streams;
- evaluate transportation and collection costs and environmental footprint of LIB wastes transportation.

The scope of the research covers transportation and recycling steps for major LIB chemistries that are used in electric vehicles.
3 Methodology

The calculations and modelling were done with the use of MS Excel and Matlab based on data collected from literature, interviews with stakeholders and technical documentation. The EverBatt model by Argonne Collaborative Center for Energy Storage Science was used to evaluate economic and environmental indicators. The model includes the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model, and Battery Performance and Cost (BatPaC) model.

Comparing to methodology in GaBi, Ecoinvent, Majeau-Bettez et al (2011), Ellingsen et al (2014), Amarakoon et al (2013) and others, the advantages of EverBatt model are that it includes both environmental impact and costs projections, provides insight into the relative impacts of different recycling paths, is in free access, has transparent calculation model and flexible data entry (Argonne National Laboratory 2019). Simplification to the minimum economic, energy, and environmental impacts and generic recycling technologies, and no user data entry support are the main disadvantages of the model.

3.1 System boundaries

Figure 9 summarizes a system boundaries that includes input parameters such as amount of spent LIBs and battery chemistry, materials, water, energy, labor and capital costs.

System outputs include recovered materials and revenues from product sales, as well as associated wastes and emissions. Impact from transportation and collection depends on distance from end user to collector and from collector to recycler. Transportation costs of recovered materials from recycler to external buyer or batteries manufacturer are not included. The comparison analysis is limited to the major commercialized recycling technologies: pyrometallurgical and hydrometallurgical recycling.

3.2 Cost Analysis

Net costs of recycling are determined as a sum of transportation and recycling costs and minus revenue

$$C_{\text{recy,net}} = C_{\text{transport}} + C_{\text{recycling}} - R.$$  \hspace{1cm} (5)

Revenues from recycling are calculated as

$$R = \sum_i m_i \times u_{p_i},$$  \hspace{1cm} (6)

where $m_i$ is the mass of recovered material $i$, and $u_{p_i}$ is the unit market price of material $i$ (Table 5).
Table 5: Value of recovered materials (Scrap Register 2019)

<table>
<thead>
<tr>
<th>Material</th>
<th>Material price (€/kg)</th>
<th>Material</th>
<th>Material price (€/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminum</td>
<td>1.14</td>
<td>Ni\textsuperscript{2+} in Ni salt/oxide</td>
<td>9.91</td>
</tr>
<tr>
<td>Copper</td>
<td>5.79</td>
<td>Co\textsuperscript{2+} in Co salt/oxide</td>
<td>45.00</td>
</tr>
<tr>
<td>Steel</td>
<td>0.26</td>
<td>Mn\textsuperscript{2+} in Mn salt</td>
<td>2.72</td>
</tr>
<tr>
<td>Plastics</td>
<td>0.09</td>
<td>Electrolyte organics</td>
<td>0.13</td>
</tr>
<tr>
<td>Lithium carbonate</td>
<td>6.93</td>
<td>Graphite</td>
<td>0.25</td>
</tr>
</tbody>
</table>

3.2.1 Transportation and collection costs

Transportation and collection costs include spent LIB transportation from end user to the collection site and transportation costs from collector to recycler. Geographically, transportation of retired LIB and recycling are expected to be done around Sweden, Norway, Belgium and Germany.

Transportation on the distance greater than 110 km is assumed to be done with heavy heavy-duty truck (25 t). Short-distance transportation (under 110 km) is done by medium-duty tracks (8 t). Ocean tankers, train, and barge are included in scenario for transportation over 1000 km.

LIB are classified as hazardous wastes in some EU countries and regions. Due to additional safety measures, permission and charges transportation costs are higher for transportation of hazardous wastes (Table 6).

Table 6: Transportation costs for LIBs

<table>
<thead>
<tr>
<th>Transport type</th>
<th>Transportation cost (€/ton-km)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Non-hazardous materials</td>
</tr>
<tr>
<td>Rail</td>
<td>0.03</td>
</tr>
<tr>
<td>Heavy heavy-duty truck (25 t)</td>
<td>0.08</td>
</tr>
<tr>
<td>Medium heavy-duty truck (8 t)</td>
<td>0.08</td>
</tr>
<tr>
<td>Ocean tanker</td>
<td>0.01</td>
</tr>
<tr>
<td>Barge</td>
<td>0.01</td>
</tr>
</tbody>
</table>

The transportation cost is then calculated as follows:

$$c_{\text{transport}} = \sum_{i} d_i \times u_i + \sum_{j} d_j \times u_j,$$

where $d_i$ and $d_j$ are the distances for transportation of nonhazardous and hazardous materials, respectively, and $u_i$ and $u_j$ are unit costs of transportation.

3.2.2 Dismantling and discharge costs

As for today pack and modules dismantling is performed manually. Robotic dismantling is evolving and was piloted in several projects, however, it still has several challenges that varies from standardization of battery packs which undesirable because can limit car manufacturers to implementation of self-learning artificial intelligence that will handle uncertainties and complexities.

Therefore, the manual dismantling was assumed. Also, it was assumed that one pack could be disassembled by one qualified worker during 8-12 hours depending on the pack design complexity.

3.2.3 Recycling costs

To develop the model for recycling costs the model for production costs of generic chemical plant was used (Max S. Peters 2003) with modifications adopted for recycling (Argonne National Laboratory 2019). Production costs model includes calculation of total capital investment and total product costs (Table 7).
The equipment costs were taken from EverBatt model, price quotes, public database, expert opinions, and literature. Price of individual equipment items, utilities and materials are given below for recycling alternatives. A direct labor rate is assumed to be 18 €/hr. The utilities costs assumptions: electricity - 0.062 €/kWh; natural gas – 0.13 €/m³; water – 0.84 €/m³. The cost of waste disposal are assumed as following: landfill (tip fee) – 40 €/ton; wastewater discharge – 1.24 €/m³.

Plant operation conditions are assumed as following: actual processing – 20 hr/day and 320 days/yr.; plant life – 10 years; capacity – 1000 t/yr., throughput – 100 t/yr., continuous process. The cost of equipment of various sizes and plant energy consumption varies with the plant throughput. The costs of equipment and energy rating curves are included with an assumption that 2 pieces of equipment are needed in the process.

For analysis, no addition gate fees on end-of-life LIB were assumed.

Costs of equipment and energy rating curves are based on costs and power rating data for each equipment item (Argonne National Laboratory 2019) and are included in the model in following equations:

\[ C_{\text{equip}} = (a + L^b) \times r_{\text{adj}} \times r_{\text{ex}} \]  
where \( L \) is the design capacity of the equipment (t/hr); \( a \) is the market price of equipment in year X or assumed recent market price of equipment unit; \( b \) is the equipment-specific cost coefficients (see 3.4.3); \( r_{\text{adj}} \) is the adjustment coefficient to convert the equipment prices according to the Chemical Engineering Plant Cost Index annual index; \( r_{\text{ex}} \) is an exchange rate (0.84 EUR = 1 USD).

\[ ER = (m \times L^a + p) \times r_{\text{ex}}, \]  
where \( m, n, \) and \( p \) are the equipment-specific energy rating coefficients (see 3.4.3).

### 3.3 Environmental impact

Environmental impact includes calculation of energy and water use, and GHGs emissions breakdown for LIB wastes transportation and recycling activities.
3.3.1 Environmental impact of transportation

Environmental impact of transportation is assumed to be the same for nonhazardous and hazardous materials and is calculated as follows:

\[ EI_{\text{transport}} = \sum d_{i,j} \times e_{i,k} \]  

(10)

where, \( e_{i,k} \) – environmental impact from each emission category, and \( d_{i,j} \) is the transportation distance.

3.3.2 Environmental impact of recycling

EverBatt model (GREET) is used to evaluate the environmental impact of batteries recycling. It includes total energy use, water consumption, different categories of air pollutant emissions, and total GHG emissions. Energy use category includes breakdown on fossil fuels (coal, natural gas, petroleum) and non-fossil fuels. Emissions breakdown includes following categories: volatile organic compound (VOC), carbon monoxide (CO), nitrogen oxides (NOx), sulfur oxides (SOx), particulate matter d\leq10\mu m (PM10), particulate matter d\leq2.5\mu m (PM2.5), black carbon (BC), and organic carbon (OC). GHGs summarize impact from carbon dioxide (CO\(_2\)), methane (CH\(_4\)), and nitrous oxide (N\(_2\)O).

An overall environmental impact is evaluated as total of environmental intensities of materials and energy input:

\[ EI_k = \sum m_i \times e_{i,k} + \sum q_i \times e_{i,k} + P_k , \]  

(11)

where \( EI_k \) – environmental impact/emission of category k for the recycling process, \( m_i \) – mass of material \( i \) (kg) consumed, \( e_{i,k} \) – GHG emissions associated with consumption of 1 kg of material \( i \) (GREET), \( q_i \) – energy consumed in the process (MJ), \( e_{i,k} \) – GHG emissions associated with consumption of 1 MJ of energy, \( P_k \) – CO\(_2\) emissions (stoichiometric calculations) produced in combustion or thermal decomposition of the materials (e.g., combustion of graphite in the pyrometallurgical recycling process) (Argonne National Laboratory 2019).

<table>
<thead>
<tr>
<th>Table 8: Carbon contents of battery parts</th>
</tr>
</thead>
<tbody>
<tr>
<td>PVDF</td>
</tr>
<tr>
<td>0%</td>
</tr>
</tbody>
</table>

\( P_k \), the recycling process CO\(_2\) emissions are calculated as:

\[ P_k = \sum_i (P_{\text{combustion}} + P_{\text{decomposition}} + P_{\text{losses}}), \]  

(12)

where \( P_{\text{combustion}} \), \( P_{\text{decomposition}} \), and \( P_{\text{losses}} \) are CO\(_2\) emissions from \( i \) step of technological process. \( P_{\text{combustion}} \) accounts for the emissions from combustion of graphite, electrolyte, plastics, carbon black and binding materials. It is calculated stoichiometrically as following:

\[ P_{\text{combustion}} = \sum_i m_i \times \frac{w(C)_i}{w(CO_2)} , \]  

(13)
where $m_j$ – mass of material $j$ that is combusted, $w(C)_j$ – carbon content of material $j$ (Table 8) (Argonne National Laboratory 2019), $w(CO_2)$ – carbon content in CO$_2$.

CO$_2$ emissions from process decomposition account thermal decomposition of Li$_2$CO$_3$, CaCO$_3$, Na$_2$CO$_3$ based on stoichiometry. 10% CO$_2$ loss is assumed for the recycling processes that use supercritical CO$_2$ extraction (Argonne National Laboratory 2019).

![Figure 10: EU-28 net electricity generation mix, 2017](image)

Recycling facilities are located around EU that is why the net electricity generation mix for EU-28 based on 2017 Eurostat data is used (Figure 10) (Eurostat 2020). Combustible fuels includes oil products, natural gas, and biomass.

The process is considered to be an open-loop. Therefore, no allocation of recovered materials to any application and no materials conversion have been considered.

### 3.4 Input data

Input data includes the information about chemical composition of batteries and materials feed, the materials and energy inputs in generic pyrometallurgical and hydrometallurgical recycling processes, and equipment costs.

#### 3.4.1 LIB chemical composition

The comparison is limited to six major types of batteries present on a market: NMC111, NMC622, NMC811, LFP, NCA, and LMO. Lithium Cobalt Oxide batteries are not included because they found very little application in electric vehicles due to their high price. Table 9 summarize input data of material composition of cells received from literature and validated for Life Cycle Assessment studies (Argonne National Laboratory 2019).

<table>
<thead>
<tr>
<th></th>
<th>NMC111</th>
<th>NM622</th>
<th>NMC811</th>
<th>LFP</th>
<th>NCA</th>
<th>LMO</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cathode</td>
<td>34.7%</td>
<td>32.4%</td>
<td>31.1%</td>
<td>32.7%</td>
<td>30.6%</td>
<td>40.8%</td>
</tr>
<tr>
<td>Graphite</td>
<td>19.4%</td>
<td>21.0%</td>
<td>20.6%</td>
<td>16.8%</td>
<td>22.1%</td>
<td>14.1%</td>
</tr>
<tr>
<td>Carbon black</td>
<td>2.3%</td>
<td>2.2%</td>
<td>1.7%</td>
<td>2.2%</td>
<td>2.1%</td>
<td>2.7%</td>
</tr>
<tr>
<td>Binder: PVDF</td>
<td>3.0%</td>
<td>2.9%</td>
<td>3.6%</td>
<td>2.7%</td>
<td>2.9%</td>
<td>3.0%</td>
</tr>
<tr>
<td>Copper</td>
<td>15.7%</td>
<td>16.1%</td>
<td>15.7%</td>
<td>13.9%</td>
<td>16.7%</td>
<td>15.0%</td>
</tr>
<tr>
<td>Aluminium</td>
<td>8.2%</td>
<td>8.4%</td>
<td>8.2%</td>
<td>7.5%</td>
<td>8.6%</td>
<td>7.8%</td>
</tr>
<tr>
<td>Electrolyte: LiPF6</td>
<td>2.2%</td>
<td>2.2%</td>
<td>2.6%</td>
<td>3.4%</td>
<td>2.3%</td>
<td>2.2%</td>
</tr>
</tbody>
</table>
Total value of recovered materials from module and pack parts (excluding cells) varies in the range of 120 – 150 EUR/pack. Pack dismantling labour costs are higher than value of recovered materials. Therefore, for simplification the value of battery materials are used. However, pack modules parts are useful for transportation purpose (see 1.3), therefore, it is reasonable to do pack discharge and dismantling as at recycling site.

Table 10 shows theoretical values of material inputs for various LIB cells types. Values are calculated from abovementioned cell parts mass (Table 9), chemical composition (molar content of elements) and molecular mass (Li – 6.94 g/mol; Co – 58.933 g/mol; Ni – 58.693 g/mol; Mn – 54.938 g/mol; O – 15.999 g/mol; P – 30.974 g/mol; F – 18.998 g/mol; Al – 26.982 g/mol; Fe – 55.845 g/mol).

### Table 10: Materials feed with 1 kg of spent LIB cells depending on battery chemistry

<table>
<thead>
<tr>
<th>Material</th>
<th>NMC111</th>
<th>NMC622</th>
<th>NMC811</th>
<th>LFP</th>
<th>NCA</th>
<th>LMO</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li (in cathode and electrolyte)</td>
<td>0.026</td>
<td>0.024</td>
<td>0.023</td>
<td>0.016</td>
<td>0.023</td>
<td>0.017</td>
</tr>
<tr>
<td>Co (in cathode)</td>
<td>0.071</td>
<td>0.039</td>
<td>0.019</td>
<td>-</td>
<td>0.028</td>
<td>-</td>
</tr>
<tr>
<td>Ni (in cathode)</td>
<td>0.070</td>
<td>0.118</td>
<td>0.150</td>
<td>-</td>
<td>0.149</td>
<td>-</td>
</tr>
<tr>
<td>Mn (in cathode)</td>
<td>0.066</td>
<td>0.037</td>
<td>0.018</td>
<td>-</td>
<td>-</td>
<td>0.248</td>
</tr>
<tr>
<td>Copper</td>
<td>0.157</td>
<td>0.161</td>
<td>0.157</td>
<td>0.139</td>
<td>0.167</td>
<td>0.150</td>
</tr>
<tr>
<td>Aluminium</td>
<td>0.082</td>
<td>0.084</td>
<td>0.082</td>
<td>0.075</td>
<td>0.086</td>
<td>0.078</td>
</tr>
<tr>
<td>Graphite</td>
<td>0.194</td>
<td>0.210</td>
<td>0.206</td>
<td>0.168</td>
<td>0.221</td>
<td>0.141</td>
</tr>
<tr>
<td>Carbon black</td>
<td>0.023</td>
<td>0.022</td>
<td>0.017</td>
<td>0.022</td>
<td>0.021</td>
<td>0.027</td>
</tr>
<tr>
<td>PVDF</td>
<td>0.030</td>
<td>0.029</td>
<td>0.036</td>
<td>0.027</td>
<td>0.029</td>
<td>0.030</td>
</tr>
<tr>
<td>Plastics</td>
<td>0.022</td>
<td>0.022</td>
<td>0.021</td>
<td>0.019</td>
<td>0.023</td>
<td>0.021</td>
</tr>
<tr>
<td>Electrolyte organics</td>
<td>0.124</td>
<td>0.126</td>
<td>0.144</td>
<td>0.188</td>
<td>0.126</td>
<td>0.122</td>
</tr>
<tr>
<td>Fe (as metal oxides)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.116</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>P</td>
<td>0.005</td>
<td>0.005</td>
<td>0.005</td>
<td>0.007</td>
<td>0.005</td>
<td>0.004</td>
</tr>
<tr>
<td>Al (as metal oxides)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.004</td>
<td>-</td>
</tr>
<tr>
<td>F</td>
<td>0.017</td>
<td>0.017</td>
<td>0.019</td>
<td>0.025</td>
<td>0.017</td>
<td>0.016</td>
</tr>
</tbody>
</table>

### 3.4.2 Material and energy inputs in recycling processes

Data about material and energy consumption requirements were taken from EverBatt and GREET Models, patents and literature (Dunn 2014) (Xie 2015). Table 11 summarize materials inputs per 1 kg LIB cells recycled in generic pyrometallurgical and hydrometallurgical methods. For simplification, it was assumed that the same amount of reagents are used for the recycling of all selected battery chemistries.
Table 11: Materials inputs for generic pyrometallurgical and hydrometallurgical recycling

<table>
<thead>
<tr>
<th>Material</th>
<th>Costs of consumed chemicals (EUR/kg of chemical)</th>
<th>Material inputs (kg/kg cells)</th>
<th>Pyrometallurgical</th>
<th>Hydrometallurgical</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ammonium Hydroxide</td>
<td>0.39</td>
<td></td>
<td>0.031</td>
<td></td>
</tr>
<tr>
<td>Hydrochloric Acid</td>
<td>0.13</td>
<td></td>
<td>0.21</td>
<td>0.012</td>
</tr>
<tr>
<td>Hydrogen Peroxide</td>
<td>0.62</td>
<td></td>
<td>0.06</td>
<td>0.366</td>
</tr>
<tr>
<td>Sodium Hydroxide</td>
<td>0.34</td>
<td></td>
<td>-</td>
<td>0.561</td>
</tr>
<tr>
<td>Limestone</td>
<td>0.11</td>
<td>0.30</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>Sand</td>
<td>0.05</td>
<td>0.15</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>Sulfuric Acid</td>
<td>0.05</td>
<td></td>
<td>-</td>
<td>1.08</td>
</tr>
<tr>
<td>Soda Ash</td>
<td>0.13</td>
<td></td>
<td>-</td>
<td>0.02</td>
</tr>
<tr>
<td>Water</td>
<td>0.0014</td>
<td></td>
<td>-</td>
<td>3.79</td>
</tr>
</tbody>
</table>

Energy consumption includes diesel, natural gas and electricity consumption. Diesel consumption is assumed to be 20 l/hr (or 0.6 KJ/kg cells) for wheel loader which is used for loading/unloading and transportation within the recycling plant. A natural gas consumption, 2.5 MJ/kg cells, is assumed for hydrometallurgical recycling. Electricity consumptions per 1 kg of recycled cells are following: 4.68 MJ – pyrometallurgy, and 0.125 MJ – hydrometallurgy.

3.4.3 Equipment costs

Equipment calculation costs were adapted from EverBatt model (Argonne National Laboratory 2019) and literature. Assumed equipment for pyrometallurgical recycling technology is given in attachment A and includes following items:

- Hopper (bottom, bolted, carbon steel, 5000 ft³ bin volume) and conveyor (belt, open, short, 42 inch wide, 100 ft long) for inlet batteries processing;
- Smelter (incinerator, rotary kiln, hazardous feed material, atmospheric pressure) and gas treatment system with top torch, water quench, and bag house;
- Conveyor (belt, open, short, 42 inch wide, 100 ft long) for pyrolyzed material and granulator (agglomerator, disk with motor, stainless 304) for produced matte;
- Leaching tank (reactor, mixer/settler, stainless 304, atmospheric to 25 psi);
- Precipitation tank (reactor, mixer/settler, stainless 304, atmospheric to 25 psi) for Cu recovery and filter press (filter, plate and frame, 200 ft² filter area, stainless 304);
- Precipitation tank ((reactor, mixer/settler, stainless 304, atmospheric to 25 psi) for Fe recovery and filter press (filter, plate and frame, 200 ft² filter area, stainless 304);
- Solvent extraction unit (reactor, mixer/settler, stainless 304, atmospheric to 25 psi);
- Wheel loader (diesel-fuelled) for feeding material to system and material handling;
- Water treatment unit.

Table 12 summarize equipment and power rating coefficients to calculate cost and energy rating functions for pyrometallurgical recycling (see 3.2.3). Cost coefficients were calculated from actual equipment prices from chemical equipment catalogues and converted recent prices, based on the annual chemical engineering plant cost index reported in chemical engineering magazine. The detailed calculation of the costs coefficients are given in the Appendix A: Equipment costs and Appendix C: Equipment cost and energy rating curves of EverBatt documentation and user manual at https://www.anl.gov/egs/everbatt (Argonne National Laboratory 2019).
Table 12: Cost and power rating coefficients for equipment in pyrometallurgical recycling

<table>
<thead>
<tr>
<th>Equipment item</th>
<th>Cost coefficients</th>
<th>Power consumption coefficients</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>b</td>
</tr>
<tr>
<td>Hopper</td>
<td>38,700</td>
<td>0</td>
</tr>
<tr>
<td>Conveyor</td>
<td>102,600</td>
<td>0</td>
</tr>
<tr>
<td>Smelter</td>
<td>6,137,979</td>
<td>0.48</td>
</tr>
<tr>
<td>Gas treatment</td>
<td>3,000,000</td>
<td>0</td>
</tr>
<tr>
<td>Granulator</td>
<td>29,902</td>
<td>0.6671</td>
</tr>
<tr>
<td>Leaching tank</td>
<td>473,892</td>
<td>0.4481</td>
</tr>
<tr>
<td>Precipitation tank</td>
<td>473,892</td>
<td>0.4481</td>
</tr>
<tr>
<td>Filter press</td>
<td>173,000</td>
<td>0</td>
</tr>
<tr>
<td>Solvent extraction unit</td>
<td>473,892</td>
<td>0.4481</td>
</tr>
<tr>
<td>Wheel loader</td>
<td>150,000</td>
<td>0</td>
</tr>
<tr>
<td>Water treatment</td>
<td>1,000,000</td>
<td>0</td>
</tr>
</tbody>
</table>

The assumed equipment for generic hydrometallurgical recycling is given in the Attachment 2. The following equipment is assumed:

- Conveyors (belt, open, short, 42 inch wide, 100 ft long) and gyratory crusher are used for mechanical processing and loading of battery material;
- Calciner (incinerator, cylindrical, low hazard feed material) to remove Carbon and PVDF with gas treatment;
- Wet granulator (agglomerator, disk with motor, stainless 304) to separate black mass;
- Density separator (cyclone separator, heavy duty) for the separation of olefin plastics from metals;
- Froth flotation cell (Reactor, jacketed and agitated, stainless 304, atmospheric to 25 psi) to separate anode from cathode;
- Filter press (filter, plate and frame, 200 ft² filter area, stainless 304) to provide an anode filter cake;
- Leaching tank (reactor, mixer/settler, stainless 304, atmospheric to 25 psi) with filter press (filter, plate and frame, 200 ft² filter area, stainless 304);
- Solvent extraction unit (reactor, mixer/settler, stainless 304, atmospheric to 25 psi);
- Precipitation tank (reactor, mixer/settler, stainless 304, atmospheric to 25 psi) and filter press (filter, plate and frame, 200 ft² filter area, stainless 304);
- Dryer (steam tube dryer, class II, 304 stainless steel);
- Wheel loader (diesel-fuelled) for feeding material to system and material handling;
- Water treatment unit.

The coefficients for the cost and energy consumption evaluation for hydrometallurgical recycling is given in the Table 13.

Table 13: Cost and power consumption coefficients for equipment costs calculation for hydrometallurgical recycling

<table>
<thead>
<tr>
<th>Equipment item</th>
<th>Cost coefficients</th>
<th>Power consumption coefficients</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>b</td>
</tr>
<tr>
<td>Conveyor</td>
<td>102,600</td>
<td>0</td>
</tr>
<tr>
<td>Crusher</td>
<td>106,512</td>
<td>0</td>
</tr>
<tr>
<td>Calciner</td>
<td>1,313,832</td>
<td>0.512</td>
</tr>
<tr>
<td>Gas treatment</td>
<td>3,000,000</td>
<td>0</td>
</tr>
</tbody>
</table>
3.4.4 Unit prices of recovered battery materials

Unit prices for recovered materials are important parameters for the calculation of potential revenue from trading of recovered materials.

Table 14: Prices of recovered materials

<table>
<thead>
<tr>
<th>Materials</th>
<th>Unit Prices (EUR/kg)</th>
<th>Materials</th>
<th>Unit Prices (EUR/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminium</td>
<td>€ 1.09</td>
<td>Ni^{2+} in product</td>
<td>€ 9.49</td>
</tr>
<tr>
<td>Copper</td>
<td>€ 5.54</td>
<td>Co^{2+} in product</td>
<td>€ 43.09</td>
</tr>
<tr>
<td>Steel</td>
<td>€ 0.25</td>
<td>Mn^{2+} in product</td>
<td>€ 2.60</td>
</tr>
<tr>
<td>Plastics</td>
<td>€ 0.08</td>
<td>Electrolyte organics</td>
<td>€ 0.13</td>
</tr>
<tr>
<td>Lithium carbonate</td>
<td>€ 6.64</td>
<td>Graphite</td>
<td>€ 0.24</td>
</tr>
</tbody>
</table>

Table 14 summarize prices of materials on scrap market (Plastics Markets 2019) (Recycler's World 2019) (Argonne National Laboratory 2019)
4 Results and Discussion

Modelling of costs and environmental impact of recycling is important part of analysis for vendors selection. Smart transportation and recycling strategies could safe many, decrease company risks and minimize environmental footprint of battery and vehicle lifecycle. The modelling is also could be applied in policy analysis for development of subsidizing strategy in battery recycling in order to achieve materials circularity and sustainable market growth. Analysis includes different scenarios of recycling facilities locations and recycling technologies. Model also takes into consideration costs and environmental footprint from recycling for low-Cobalt (NMCs, NCA) and no-Cobalt (LFP and LMO) battery chemistries to analyse potential revenue streams.

4.1 Transportation costs and environmental impact

Transportation of spent LIBs adds costs, economically and environmentally. Costs are associated with the energy consumption, ensuring of safety of the hazardous LIBs on the way, and compensation of potential risks on environment and human health.

Modelling of transportation costs accounts:
- transportation from end user to collector;
- transportation from collector to recycling facility;
- permit fees.

4.1.1 End-of-life LIB collection and transportation costs

Spent LIBs transportation are considered in two scenarios: according to hazardous and non-hazardous materials transportation. Because of potential threats to the environment and public safety, transport of hazardous materials is given special attention by governmental agencies. Transportation company has to take care of packaging and cooling of spent LIBs to avoid accidental damage, spill, or ignition of batteries. There costs for transportation of hazardous wastes are significantly higher (see 3.2.1).

LIBs waste is usually transported by truck over public highways. Only a very small amount of LIBs is transported by rail or barge. Highway shipment is the most common because road vehicles can gain access to most industrial sites and recycling facilities. Railroad trains and inland waterway transportation require expensive siding facilities and are suitable only for very large waste shipments and is assumed only for distances over 1000 km. Maritime transportation is assumed for the scenario if spent LIBs are send for recycling to South Korea or China (8000 km).

Transportation costs increases linearly and starts from 0.27 €/kg spent batteries for 50 km transportation distance if hazardous regulation is applied (Figure 11). With this regulations, spent LIBs transportation from
EU to recycling facilities in South Korea or China will cost 28.27 €/kg spent cells for the transportation on distance of 8000 km.

If non-hazardous transportation policy is applied, the costs of transportation on 50 km distance is 0.0043 €/kg LIBs. Transportation on a 8000 km distance will costs only 0.63 €/kg LIBs. The application of non-hazardous wastes regulations on end-of-life LIB transportation makes recycling a profitable business in countries with low-cost labour and facilitates circularity of materials. However, risks of inflammation and leakage are extremely high. Therefore, non-hazardous regulations are applied only to the recovered in recycling materials. So, in order to minimize risks and costs, the closest to end user recycling facilities should be selected.

First 200 km are the most expensive (Figure 13), with net transportation costs of 5.39 €/(t cells·km) for 100 km transportation distance, comparing to 3.60 €/(t cells·km) and less if transportation distance is over 200 km. First, this is related to higher labour costs if medium-heavy track (8 t) is used. Second reason is potentially in higher energy use (see 4.1.2.1). A smart planning and prediction of battery wastes availability play important role to keep optimum track load and labour planning.
4.1.2 Environmental impact of spent LIB transportation

Waste generator (battery producer or vehicle manufacturer) is responsible for any consequences of end-of-life LIB mismanagement. However, transportation companies are also obliged to eliminate a negative environmental impact and safety risks of wastes collection and transportation. Spent LIBs transportation requires energy and emits pollutions from combustion of fossil fuels. The analysis includes evaluation of energy and water use, and GHGs composition associated with transportation.

4.1.2.1 Energy consumption

The model includes fuel-cycle energy consumption for the following two source categories: fossil fuels (coal, natural gas, and petroleum) and non-fossil fuels. Energy use increase with distance from 0.17 MJ/kg cell for transportation on 50 km distance till 6.11 MJ/kg cell for 8000 km transportation.

![Figure 14: Energy use for spent LIBs transportation](image)

However, net energy consumption (Figure 15) decrease with distance growth and remains highest (as well as net costs) for the first 200 km with medium-heavy truck transportation.

![Figure 15: Net energy consumption in transportation](image)

The analysis of energy use by the source was made for the scenario of 8000 km transportation and shows that 87.36% of energy comes from petroleum, 10.95% comes from natural gas, 1.19% is from coal, and only 0.5% is from non-fossil fuels (Figure 16). The share of fossil fuels is the highest in the short-distance transportation (Figure 14).
4.1.2.2 Water consumption

Water consumption is comparably low and reach only 0.378 L/kg cells in the scenario of spent LIB transportation on a distance of 10000 km. Water consumption is associated with fuel extraction and production. Fuels production from fossils requires less water comparing to fuels that derive either indirectly from fossil fuels (e.g., through electricity generation) or directly from biomass. Previously, it was shown that the lowest water consumption and withdrawal rates are for vehicles using conventional petroleum-based gasoline and diesel, non-irrigated biofuels, hydrogen derived from methane or electrolysis via nonthermal renewable electricity, and electricity derived from nonthermal renewable sources (Carey W. King 2008).

As a comparison, the vehicles running on electricity and hydrogen based upon fossil fuel and nuclear steam-electric power generation withdraw 5−20 times and consume nearly 2−5 times more water than by using petroleum gasoline. The water intensities of transport operating on biofuels derived from irrigated crops are 28 and 36 times more for corn ethanol (E85) for consumption and withdrawal, respectively. For soy-derived biodiesel the average consumption and withdrawal rates are 8 and 10 times more than for diesel and gasoline (Carey W. King 2008).

4.1.2.3 Emissions of transportation

The transportation sector generates largest share of greenhouse gases (GHGs) emissions. Thus, transportation of 1 kg of spent LIBs emits 12.7 g CO₂ eq. per 100 km and 467.2 g CO₂ eq. per 10 000 km GHGs comes from burning fossil fuels (Figure 18).

Figure 17: Water consumption associated with spent LIBs transportation

Figure 18: GHGs and CO₂ share in emissions from spent LIBs transportation
GHGs emissions come from fossil-based fuels combustion and 95.5-96.2% of emissions are CO₂ produced in exothermic oxidation of carbohydrates. Figure 19 summarize simulation results of CO, NOₓ, CH₄, SOₓ, and N₂O emissions, volatile organic compounds (VOC), and particle pollutions (PM10, PM2.5, BC, and OC) for various transportation distance. Black carbon (BC) and organic carbon (OC) aerosols are formed by incomplete combustion and is identified as having potentially significant impacts on climate change, particularly at regional scales. Due to higher energy intensity of first 200 km is also more polluting.

Sulphur Oxides (SOₓ), nitrogen oxides (NOₓ), particulate matter (PM) in the emissions breakdown increase more gradually with the distance comparing to other emissions components. SOₓ, NOₓ and PM are produced in the combustion and energy transformation processes (propulsion and energy production) and in the main challenge of maritime transport energy transition as its directly emitted to the atmosphere. Therefore, with the increase of the transportation distance the emission intensity growth as well.

Therefore, international transportation of LIB wastes is associated with additional costs and risks. With the respect to hazardous materials transportation regulations, sending spent LIBs on the distance over 1000 km makes recycling unprofitable even for the batteries with high content Cobalt. Every 100 km of transportation emits GHGs from 4.7 to 12.7 g CO₂ per 100 km depending on transportation type and is associated with fossil fuels use.
4.2 Recycling comparison of LIBs

Battery pack is the main component of electric vehicle which is necessarily needs to be recycled due to potentials negative impact of dump disposal and valuable materials content. Battery composition is defined by battery manufacturer and often is an industrial secret. Modules and packs are designed and optimized to meet customer needs, provide high performance and safety, and have overall materials and assembly costs as cheap as possible. Recycling often includes dismantling and discharge pre-processing and cells materials decomposition with further extraction of recovered materials. Recycling revenue and environmental footprint is calculated for battery packs materials, and cells with various content of Cobalt (NMC and NCA) or Cobalt-free (LMO and LFP).

4.2.1 Pack disassembly and discharge

Material selection and assembly method as well as component design are very important to determine the cost-effectiveness of battery modules and battery packs. Battery pack composition was assumed from literature data (N. Lewchalermwong 2017) (Rangarajan 2018). Around 90% of battery pack in heavy electric vehicles is aluminium alloys, 9% is mixed electronics and 1% is rubber and plastics.

![Figure 20: Pack materials weight and economic value](image)

<table>
<thead>
<tr>
<th>Material</th>
<th>Theoretical economic value of materials</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminium alloys</td>
<td>€ 155.50</td>
</tr>
<tr>
<td>Electronic wastes (mix)</td>
<td>€ 16.79</td>
</tr>
<tr>
<td>Total</td>
<td>€ 172.35</td>
</tr>
</tbody>
</table>

Assuming that the pack dismantling is done manually, the labor costs of pack dismantling will be 144 €/pack. However, due to various pack design and pre-processing requirement for different technologies, the dismantling costs could also vary. Additionally, the utilities, on site transportation and loading costs, stuff training and certification, administrative costs, insurance and externalities should be included. However, these expenses are included in the model as a part of recycling activities. Therefore, we assume best case scenario where the value of recovered materials from pack (Figure 20) covers the dismantling and discharge costs. Both dismantling and discharge are done at recycling site despite of the battery state of health and chemical composition of electrodes. So, the main revenue streams come from the cells recycling.

4.2.2 Recycling of the Lithium Nickel Manganese Cobalt Oxide batteries

Batteries with the Lithium Nickel Manganese Cobalt Oxide electrodes (NMC) are mainly used for vehicles that require high specific energy and power, safety, performance and lifespan. That is why NMC is the battery choice for electric powertrains. Due to comparable high Cobalt content NMC batteries are expensive, but interesting to recycle. The following NMC chemistries are considered in the model: LiNi0.33Mn0.33Co0.33O2 (NMC111), LiNi0.6Mn0.2Co0.2O2 (NMC622), and LiNi0.8Mn0.1Co0.1O2 (NMC811). The analysis includes comparison of recycling revenue streams and costs breakdown, and environmental footprint for pyrometallurgical and hydrometallurgical recycling.
4.2.2.1  **NMC cells recycling revenue and costs breakdown**

The economic benefits of both pyrometallurgical and hydrometallurgical recycling processes are very promising for NMC cells. Figure 21 summarize potential revenue of recycling facility per 1 kg cells recycled. The hydrometallurgical recycling gives higher profit comparing to pyrometallurgical method thanks to higher material recovery efficiency.

![Figure 21: Revenue from recycling NMC batteries (per kg cells recycled)](image)

<table>
<thead>
<tr>
<th>Cell Type</th>
<th>Hydrometallurgical Recycling</th>
<th>Pyrometallurgical Recycling</th>
</tr>
</thead>
<tbody>
<tr>
<td>NMC111</td>
<td>Copper, 17%</td>
<td>Copper, 18%</td>
</tr>
<tr>
<td></td>
<td>Ni²⁺ in product, 14%</td>
<td>Ni²⁺ in product, 15%</td>
</tr>
<tr>
<td></td>
<td>Co²⁺ in product, 63%</td>
<td>Co²⁺ in product, 67%</td>
</tr>
<tr>
<td>NMC622</td>
<td>Copper, 21%</td>
<td>Copper, 22%</td>
</tr>
<tr>
<td></td>
<td>Ni²⁺ in product, 29%</td>
<td>Ni²⁺ in product, 31%</td>
</tr>
<tr>
<td></td>
<td>Co²⁺ in product, 44%</td>
<td>Co²⁺ in product, 31%</td>
</tr>
<tr>
<td>NMC811</td>
<td>Copper, 25%</td>
<td>Copper, 26%</td>
</tr>
<tr>
<td></td>
<td>Ni²⁺ in product, 44%</td>
<td>Ni²⁺ in product, 31%</td>
</tr>
<tr>
<td></td>
<td>Co²⁺ in product, 25%</td>
<td>Co²⁺ in product, 47%</td>
</tr>
</tbody>
</table>

*Figure 22: Cell Recycling Revenue Breakdown for NMC LIBs*
The highest revenue of 4.99 €/kg cells recycled comes from hydrometallurgical recycling of NMC111 cells. The recycling revenue breakdown shows that Cobalt and Nickel compounds generate main share of the profit (Figure 22).

An analysis of costs breakdown shows that the recycling costs for hydrometallurgical method are equal despite of the NMC chemistry (Figure 23). The main share of expenses is associated with the materials purchase and consumption. In pyrometallurgical recycling share of costs for utilities and labour costs are higher. Utilities costs comes from the energy consumption in high-temperature pyrolysis. Also, pyrometallurgical method with subsequence hydrometallurgical recovery requires highly qualified labour for both high-temperature pyrometallurgy operation and chemist for leaching and precipitation processes.

Therefore, taking into consideration transportation costs, the recycling of NMC batteries is profitable or costs neutral if recycling facility is located on distance closer than 700-800 km from end user if hazardous regulation for wastes transportation is applied.

### 4.2.2.2 Environmental footprint of NMC batteries recycling

Pyrometallurgical recycling is associated with the higher use of utilities (energy) in the process. However, analysis of total energy use shows that hydrometallurgical recycling has higher energy intensity due to high energy consumption in materials production and transportation (Figure 24). Natural gas is the main energy source for both recycling alternatives.

Energy and water use, and GHGs emission are the same for NMC battery chemistries because same technological processes are applied. Hydrometallurgical method consumes over 13.2 liters of water per 1 kg cells recycled and pyrometallurgical recycling uses only 4.9 liters of water per 1 kg cells recycled (Figure 25).
4.2.3 Recycling of the Lithium Nickel Cobalt Aluminium Oxide batteries

The Lithium Nickel Cobalt Aluminium Oxide (LiNiCoAlO$_2$) batteries (NCA) is another type of LIB with high Cobalt content. NCA batteries are used in majority of Tesla vehicles in the 18650 cell that delivers an high specific energy of 3.4Ah per cell or 248Wh/kg (Battery University 2020).
The calculated revenues from recycling of 1 kg of NCA cells are 3.41 €/kg cells and 3.54 €/kg cells for pyrometallurgical and hydrometallurgical methods, respectively. The main revenue sources from NCA batteries recycling are also from recovery Cobalt and Nickel compounds, and Copper (Figure 28). Hydrometallurgy also allows recycle graphite electrode and aluminium that adds revenue. The recovered materials have high quality and can be used for production of batteries (Argonne National Laboratory 2019).

Materials is also the main expense category (75%) in the hydrometallurgical NCA recycling (Figure 29) as well as utilities and labour in pyrometallurgy. Figure 30 Error! Reference source not found. summarize the energy use in recycling processes and here hydrometallurgical recycling requires almost two times more energy comparing to pyrometallurgy.

Emission breakdown for NCA recycling is similar to NMC recycling because the process and materials use ratio are the same (Figure 32). The biggest impact in hydrometallurgical recycling also comes from SOx, NOx and CH4 emissions.
Similarly to NMC cells, NCA recycling is unprofitable if end-of-life cells are transported on the distance over 800 km. Hydrometallurgical recycling has higher negative environmental footprint and more costly due to production and transportation of reagents.

4.2.4 Recycling of the Lithium-ion Manganese Oxide batteries

The lithium ion manganese oxide battery (LiMn$_2$O$_4$, Li$_2$MnO$_3$, or LMO) is the type of Li-ion batteries that does not contain Cobalt. LMO/NMC blend batteries are used in BMW and Nissan vehicles. LMO batteries are safe, have high specific energy and specific power and comparatively cheap.

However, recycling of LMO batteries is low-profitable due to low price and high market availability of Manganese compounds. Copper is the only source of revenue of 0.75 €/kg LMO cells in the pyrometallurgical recycling. The revenue from hydrometallurgy is 0.86 €/kg LMO cells recycled (Figure 33).

Hydrometallurgy is 15% more expensive comparing to pyrometrical recycling and 75% of costs is utilized on materials purchase, 20% are labor-related costs and only 5% is used on utilities. In pyrometallurgical recycling the costs breakdown is following: 42% - labor, 36% - utilities, and 20% - materials.

Energy use (Figure 34), water consumption (Figure 35) and emissions (Figure 36) from LMO cells recycling are similar to recycling of batteries with high Cobalt content. However, if we consider transportation costs, recycling of LMO batteries is self-sustainable and should be included in batteries price formation. Another way is subsidization of recycling industry and development of internal recycled materials market that competes with virgin materials supply.
4.2.5 The Lithium Iron Phosphate batteries recycling

The lithium iron phosphate battery (LiFePO₄ battery) or LFP battery (lithium ferrophosphate). In May 2020, tesla announced that LFP batteries will be used in mass production of Tesla vehicles. LFP batteries contain neither nickel, nor Cobalt that make this LIBs inexpensive. Moreover, LFP batteries have comparably high energy density and are proven to be safer thanks to stabilization of the redox energies.

Copper is the main source of revenue for LFP batteries (Figure 37). The modelling shows that 1 kg of LFP batteries generate 0.69 € and 0.80 € of revenue in pyrometallurgical and hydrometallurgical recycling, respectively. As well for other battery chemistries, hydrometallurgy has high costs of materials and pyrometallurgy required the se of utilities. 1 kg of LFP recycling consumes 5.69 litres of water in pyrometallurgical recycling and 13.31 litres of water in hydrometallurgical recycling. Figure 38 shows the energy consumption breakdown for LFP batteries.
Total emissions generated in recycling are 2.16 kg CO$_2$ eq./kg cells recycled and 2.36 kg CO$_2$ eq./kg cells recycled for pyrometallurgical and hydrometallurgical methods, respectively. SOx emissions is the main contributor of non-CO$_2$ GHGs emissions in hydrometallurgical recycling.

Therefore, recycling of LFP batteries is not interesting for recyclers due to low potential revenue and high recycling costs. For this reason recycling facilities in California have gate fee 2.00 $/kg cells on recycling of LFP batteries (Argonne National Laboratory 2019). Similar fees or governmental support in EU for the batteries that do not contain Cobalt or Nickel.
5 Conclusions

Li-ion batteries recycling is an important segment in Li-ion battery supply chain. It is profitable business with an existing materials prices. Recycling of Li-ion batteries with high content of Cobalt and Nickel generate significantly more revenue comparing to the Lithium-ion Manganese Oxide and Lithium Iron Phosphate batteries. Cobalt, nickel and metallic fractions are currently main source of revenues. The recycling of 1 kg batteries using pyrometallurgical recycling technology generates following revenue: NMC111 – 4.68 €/kg cell; NMC622 – 3.77 €/kg cell; NMC811 – 3.15 €/kg cell; NCA – 3.41 €/kg cell; LMO – 0.75 €/kg cell; LFP – 0.69 €/kg cell. Hydrometallurgy allows the recovery of additional cells components and generates higher revenue: NMC111 – 4.99 €/kg cell; NMC622 – 4.01 €/kg cell; NMC811 – 3.33 €/kg cell; NCA – 3.54 €/kg cell; LMO – 1.49 €/kg cell; LFP – 0.80 €/kg cell.

Transportation costs is the main factor that determines profitability of recycling. First 200 km of transportation is the most expensive. 99% of transportation is fossil fuels based. However, transportation on distance over 800 km with hazardous regulations applied consumes all potential profit from battery recycling. However, battery pack can be used as a transportation case that may decrease transportation costs and secure transportation safety.

Pack dismantling is expensive and dangerous part in recycling pre-processing due to high labour costs and low economic value of pack and module materials.

GHGs emissions and water use is generally higher for hydrometallurgical method, but pyrometallurgy does not give high materials recovery and is limited for optimization. Thus, GHGs emissions from pyrometallurgical recycling are 2.17, 2.26, 1.98, and 2.16 kg/kg cells for NMC, NCA, LMO, and LFP, respectively. GHGs emissions intensity for hydrometallurgy are 2.28, 2.28, 2.27, and 2.37 kg/kg cells for NMC, NCA, LMO, and LFP, respectively. Water consumption in pyrometallurgical recycling of spent Li-ion for the NMC, NCA, LMO, and LFP batteries are 4.92, 5.68, 5.69, and 5.69 L/kg cell, respectively. Water use in the hydrometallurgical recycling is significantly higher and have the following calculated values: NMC – 13.25 L/kg cell; NCA – 13.25 L/kg cell; LMO – 13.31 L/kg cell; LFP – 13.31 L/kg cell.
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Attachments

Attachment 1: Process technology equipment for generic pyrometallurgical recycling
Attachment 2: Process technology equipment for generic hydrometallurgical recycling