Challenges and opportunities of recycling Li-ion batteries

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Bachelor's thesis
LUT School of Energy Systems
Electrical Engineering

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ABSTRACT

Lappeenranta–Lahti University of Technology LUT
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This bachelor’s thesis examines the current state of Li-ion batteries recycling industry around the world and the possibilities and challenges within it. Also recycling methods and recycling emissions are studied. The research uses online sources.

The main problems include low profit, diversity and non-labelling among batteries, lack of recycling laws, reluctance to recycle cheap batteries, their components or substances and moving to cheaper chemistries.

Opportunities include growing market, possible local business growth and getting materials out of foreign suppliers. It also reduces EV prices and carbon footprint as well as save mineral resources and environment.

Laws or incentives are needed to support LIB recycling. Producer responsibility has shown great results in financing and supporting LIB recycling and standardization as well as labelling among batteries, in China.

TIIVISTELMÄ

Lappeenrannan–Lahden teknillinen yliopisto LUT
School of Energy Systems
Sähkötekniikka

Anniina Sorvisto
Mahdollisuudet ja haasteet litiumioniakkujen kierrätyksessä
2022

Kandidaatintyö
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Tarkastaja: Juha Pyrhönen
Ohjaaja: Juha Pyrhönen

Tämä kandidaatintutkimus tarkastelee litium-ioniakkujen kierrätyksen nykytilaa sekä sen mahdollisuuksia ja haasteita. Lisäksi tutkitaan kierrätysmenetelmiä ja kierrätyspäästöjä. Tutkimuksessa käytetään internetistä löytyviä tutkimuksia lähteinä.

Suurimpia ongelmia ovat kierrätyksen taloudellinen kannattamattomuus, akkukennojen moninaisuus ja merkitsemättömyys, kierrätyslakien puute, haluttomuus kierrättää halpoja akkuja, niiden komponentteja tai ainesosia ja kalliimmista metalleista halvempiin siirtyminen.

Mahdollisuuksia litiumioniakkujen kierrättämisessä ovat niiden jatkuvasti kasvavat markkinat, mahdollinen paikallisen liiketoiminnan kasvu ja materiaalien lokalisointi. Kierrättäminen vähentää sähköajoneuvojen hintaa, hiilijalanjälkeä sekä säästää mineraalivaroja ja ympäristöä.

Litiumioniakkujen kierrätyksen tukemiseksi tarvitaan lakeja tai kannustimia. Kiinassa tuottajavastuu on osoittanut hyviä tuloksia rahoittamalla ja tukemalla litiumioniakkujen kierrätystä ja standardisointia sekä akkukennojen merkitsemistä.

CONTENTS

LIST OF ABBREVIATIONS

BMS	Battery management system
CO_2	Carbon dioxide
EPR	Extended producer responsibility regulation
EU	European Union
GHG	Greenhouse Gasses
LCO	Lithium cobalt oxide battery
LFP	Lithium iron phosphate battery
LIB	Lithium-ion battery
LMO	Lithium manganese oxide battery
NCA	Lithium nickel aluminium oxide battery
NMC	Lithium nickel manganese cobalt oxide battery
PHEV	A plug-in hybrid electric vehicle
PRBA	Portable Rechargeable Battery Association
USA	United States of America

1. INTRODUCTION

Lithium-ion batteries (LIB)s are used in most rechargeable electronic devices and electric vehicles requiring high energy density that LIBs have. *(Chen, 2019)* Li-ion battery industry has grown exponentially in the late 20th century and continues to grow due to rising market of electric vehicles. LIBs gained compound annual growth rate (CAGR) of 24% from years 2015 to 2018. *(Breyer, 2020)* Still the LIB recycling business is at an infant stage which raises concerns about the future availability of lithium and cobalt when batteries alone possibly use over 10% of world cobalt reserves. *(Gaines, 2018)*. In 2015, Li demand was about 34.6 kt and in 2019 it raised to 49 kt. Breyer's study "Assessment of lithium criticality in the global energy transition and addressing policy gaps in transportation" *(Breyer, 2020)* shows that in the short term, supply and demand of lithium are at balance, but the long-term sustainability is endangered. There is a need for efficient recovery system to maintain lithium in circulation and prevent a future shortage. If recycling is not improved lithium shortage is met before the end of the century. Breyer's study suggests that at medium resources, Li resources are used by 2055. *(Breyer, 2020)*

EU's battery directive demands that 95% of cobalt, copper, nickel and 70% of lithium need to be regained from used Li-ion traction batteries by 2030. *(European Parliament)* EU sets relatively high requirements for Li-ion battery recycling but for example in Australia only 2% of the annual 3,300 tonnes of lithium-ion battery waste are recycled. *(Lithium-ion battery recycling. 2021)*

Lithium-ion batteries contain harmful substances to environment such as lithium, transition metal oxides, phosphates, aluminum, copper, graphite, organic electrolytes, lithium salts, and polymer separators. These substances end up in the environment if batteries are disposed of in landfills after use. Li-ion batteries in landfills could catch fire or cause hydrogen fluoride formation if the electrolyte is in contact with water or groundwater contamination if drained to the soil. Recycling lithium-ion batteries can save money, mineral resources and the environment from toxic materials and fire hazards in waste and transport equipment caused by LIB self-ignition. *(Beaudet, 2020)*

The intent of this study is to find out how much carbon dioxide emissions are generated when a lithium-ion battery is recycled into a new battery and what are the current challenges faced and what is the potential in LIB recycling industry. The research questions for this bachelor's thesis are: How are Li-ion batteries recycled across the world today? How much carbon emissions are generated of recycling a Li-ion battery versus producing a virgin Li-ion battery? How can the recycling be improved? What are the possibilities and challenges with LIB recycling?

1.1 Structure and types of LIBs

Desirable qualities with Li-ion batteries include lightweight, high discharge voltage and energy density, long life cycle, reduced memory effect, many supported charging/discharging cycles and fast charging abilities for rechargeable electric devices. *(Heelan, 2016) (Mossali, 2020)* The largest applications for LIBs are power tools, portable consumer electronics, and electric vehicles. *(Beaudet, 2020)* In electric vehicles LIBs are usually used in a parallel circuit because it is more efficient than one large battery.

Main parts of a LIB cell are cathode, anode, electrolyte, and separator. Lithium ions travel from cathode to anode when battery has voltage. The separator prevents physical contact of the electrodes and provides an ionic conduction path for the liquid electrolyte. *(Heelan, 2016)* The commercially most used anode material in LIBs is graphite. Intercalated lithium compound is the commonly used cathode material. The electrolyte consists of lithium salts, such as $LiPF_6$, $LiBF_4$, $LiAsF_6$ or $LiClO_4$ and an organic solvent that is a mixture of alkyl carbonates, such as, dimethyl carbonate, ethylene carbonate, and diethyl carbonate. Porous polyolefin membranes are the most used separator materials currently. *(Heelan, 2016)*

The capacity of a LIB decreases when it is cycled and when it gains calendar age. How fast the capacity weakens depends on how the battery has been used, whether LIB has been used at its own or in a battery pack, and what type of electrode composition it has. *(Mengyuan, 2019)*

Li-ion traction batteries have three levels: battery system, modules, and cells. Battery system is the highest level that consists of battery modules. Battery modules consist of six components: cell contacting, cell fixation, thermal management, housing, battery management system (BMS) and the battery cells. *(Gerlitz, 2021)* Figure 1 displays a hypothetical plug-in hybrid electric vehicle (PHEV) battery-pack that has eight modules and 12 cells per module.

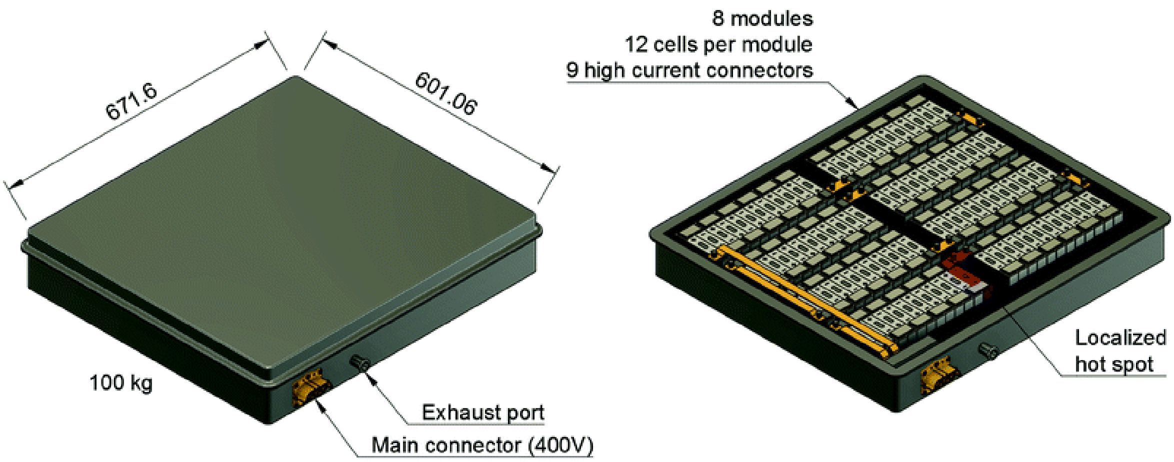

Figure 1. Hypothetical PHEV battery-pack. (Golubkov, A.W & Planteu, R & Krohn, P & Rasch, B & Brunnsteiner, B & Thaler, A & Hacker, V. 2018)

The LIB cell types include cylindrical, parallelepiped (often called prismatic), coin and pouch cells. Cylindrical cells have many cell rows in a module with arresters on opposite sides. Prismatic cells have either one-rowed or two-rowed modules and the arresters are always on the same side. Pouch cells have one-rowed modules with the arresters being either on the same side or the opposite sides. *(Gerlitz, 2021)*

A cylindrical cell is one of the first widely produced LIB types that consists of flat anodes, separators, and cathodes that are piled, rolled, and packed into a cylinder-shape. Cylindrical cells are used for automated manufacturing. The advantages with this cell type are its mechanical and thermal stability and the disadvantages are low packaging density and the need for many cells for same pack capacity with other cells. *(Fisher, 2021) (Arar, 2020)* Cylindrical cells are mostly used in portable electronics such as power tools, medical tools, laptops, and electrical bikes. *(BU-301a: Types of Battery Cells, Battery University, 2019)* Cylindrical cell is demonstrated in figure 3.

LIB prismatic cells consist of layers of electrodes, and separators rolled up or stacked into a cubical metallic or hard-plastic housing. Prismatic cells' capacity varies from several ampere-hours targeted for portable electronics to hundreds of ampere-hours designed for EVs. Prismatic cells have the advantage of optimal usage of space since the box-like packing but disadvantage of thermal heating. *(Arar, 2020)* Usage for this cell type include electric powertrain, energy storage systems, mobile phones, tablets, and laptops. *(BU-301a: Types of Battery Cells, Battery University, 2019)* Prismatic cell is visualized in figure 2.

Figure 2. Prismatic LIB cell. Types of lithium batteries: lithium cell format, OneCharge)

Figure 3. Cylindrical LIB cell. (Types of lithium batteries: lithium cell format, OneCharge)

LIB pouch cells are lightweight and offer an adaptable size and optimal usage of space. The layers of electrode and separator in pouch cell are stacked, and the cells are in a sealed flexible coil. Pouch cells have between 90%-95% packaging efficiency and increased energy density. *(Arar, 2020)* Small cells are mostly used for portable applications that need high load currents, such as drones and hobby appliances. Larger cells can serve in energy storage systems. *(BU-301a: Types of Battery Cells, Battery University, 2019)* Pouch cell can be seen in figure 4.

LIB coin or button cells have a low self-discharge meaning they have an ability to keep their charge for a long period of time. They are small and inexpensive to build but many coin cells have problems with swelling if charged too quickly. Coin cells consistency makes it suitable for watches, medical implants, car keys, hearing aids and memory backup. *(BU-301a: Types of Battery Cells, Battery University, 2019) (Lithium-Ion Batteries, Microbattery)* Coin cell can be found in figure 5.

Figure 4. Pouch LIB cell. (Types of lithium batteries: lithium cell format, OneCharge)

Figure 5. Coin cell. (Lithium-Ion Batteries, Microbattery)

1.2 Different electrolytes for LIBs

Lithium-ion battery has six different commercialized electrolyte chemistries that are reviewed below. Table 1 shows comparison of different LIB types in terms of specific energy, cost, specific power, safety, lifespan and performance.

Lithium Nickel Cobalt Aluminium Oxide battery (NCA) has a high specific energy. It is moderate in performance, cost, specific power, and lifespan. The disadvantage with this battery is its low level of safety. Moderate lifespan and high specific energy render it fitting for electric vehicles and electric bikes. *(Nuamah, 2021)*

Lithium Nickel Manganese Cobalt Oxide battery (NMC) can have a high specific energy or power. It is used for example in electric powertrains, vehicles, and bikes due to its high specific energy. Good qualities also include relatively low cost and moderate safety, lifespan, and performance in comparison to the other LIBs. *(Nuamah, 2021)*

Lithium Manganese Oxide battery (LMO) has the advantage of low price. Moderate qualities include specific power, specific energy, and safety when compared to the other types of li-ion batteries. The disadvantages are low performance and lifespan. LMOs are utilized in power tools and medical devices. *(Nuamah, 2021)*

Lithium Iron Phosphate battery (LFP) offers high specific power, high level of safety but also long lifespan, and low cost. One disadvantage is low specific energy when compared to other types of LIBs. It has moderate performance. *(Nuamah, 2021)*

Lithium Titanate battery (LTO) has the advantage of high safety, performance, and long lifespan. It has also extremely fast recharge time and moderate specific power when compared to other LIBs. Disadvantages are high cost and low specific energy. Applications for LTOs include solar energy storages and smart grids. *(Nuamah, 2021)*

Lithium Cobalt Oxide (LCO) has low cost and high specific energy that makes it suitable for laptops and mobile devices. On the downside, it has low level of safety, specific power and short lifespan. Its performance is considered moderate. *(Nuamah, 2021)*

	Specific energy	*Cost*	*Specific power*	*Safety*	*Life span*	*Performance*
NCA	*high*	*moderate*	*moderate*	*low*	*moderate*	*moderate*
NMC	*high*	*low*	*high*	*moderate*	*moderate*	*moderate*
LMO	*moderate*	*low*	*moderate*	*moderate*	*short*	*low*
LFP	*low*	*low*	*high*	*high*	*long*	*moderate*
LTO	*low*	*high*	*moderate*	*high*	*long*	*high*
LCO	*high*	*low*	*low*	*low*	*short*	*moderate*

Table 1. Different LIB electrolyte types compared in terms of specific energy, cost, specific power, safety, life span and performance.

1.3 Current recycling systems

The two most used processes for lithium-ion battery recycling are pyrometallurgical and hydrometallurgical processes. Pyrometallurgical treatment has high temperature smelting to recover metals as an alloy that can be further separated with hydrometallurgy or used in other applications. Hydrometallurgy uses chemical leaching to dissolve metals. There is also direct recycling that is still at early stage, but it has potential to preserve old LIBs to new life cycle by separating the anode or cathode and reconditioning it. Figure 7 presents the steps for these recycling systems.

1.3.1 Pyrometallurgical recovery

Pyrometallurgy is the most used LIB recycling method used all over the world by major companies, such as Umicore, Dowa and Sumitomo in Japan, Accurec in Germany, Batrec and Nickelhütte Aue Gmbh in Germany. *(Larouche, 2020)*

In pyrometallurgy the batteries are melted at high temperatures in which the polymers that keep the battery cells together burn off and the components of the metal oxides are reduced to a metallic alloy consisting of cobalt, copper, iron, and nickel. (*Beaudet, 2020.)* Besides metallic alloy, smelting also produces slag, and gas. The metal alloy and slag can be more dissolved though hydrometallurgy into its separate components or used in other applications for example in cement industry. *(Mengyuan, 2019) (Harper, 2019)*

The advantages with pyrometallurgical recovery are that it is relatively safe and the exothermic reaction from polymer combustion reduces the required input energy. Pre-treatment or crushing are not necessary meaning fewer intermediate stages. *(Beaudet, 2020)*

The main disadvantages of pyrometallurgy are the relatively expensive gaseous wastewater treatment plants needed to avoid the release of toxic compounds into the air and water effluents. If used without hydrometallurgy, the valuable elements of LIBs, such as electrolyte, graphite, steel, aluminum, and lithium are lost as slag or exhaust gas in the process. That makes the recycling of cheap LIBs like LiFePO4, $LiMnO_2$ or $LiTiO_4$ unprofitable and economically inefficient. *(Li-ion battery recycling, a catalyzer to proper waste regulation, 2020) (Gaines, 2014)*

1.3.2 Hydrometallurgical metals reclamation

Hydrometallurgical metals reclamation uses aqueous substances to remove the desired metals from the cathode, usually at low temperatures. Before the actual process cells are shredded and sieved to remove pieces of metal. *(Gaines, L. & Dai, Q, 2021)* The first step is leaching that dissolves metals with acid, base or salt. After leaching, the metals are separated via ion exchange, precipitation, and solvent extraction. Lastly, wanted metals are recovered from solution as a metal, a metal salt, or a compound. Cobalt, the most valuable metal, can be regained in the form of sulphate, oxalate, hydroxide, or carbonate. Recovering the metals from the solution can be implemented by ionic precipitation, crystallization, electrochemical reduction, reduction with gas or electrolytic reduction. *(Brückner, 2020)*

Hydrometallurgy's viability depends on the flowsheet complexity, reagent systems, toxicity level of effluent, and consumption of water. Hydrometallurgy can separate different valuable elements like cobalt, nickel, manganese, and lithium to single element compounds. *(Larouche, 2020)*

Advantages in hydrometallurgy include relatively low energy consumption since the low temperature compared to pyrometallurgy, lithium can be recovered from the process in carbonate form, and it has good efficiency on different battery chemistries. *(Mossali, 2020)* The disadvantages are that a large amount of solvent is needed, and the neutralization is expensive. It complicates the process that the cathode and anode need to be separated at the start and the design of the batteries can make it challenging. *(Mengyuan, 2019)*

Hydrometallurgical reclamation is a marginally used method alone. Many companies use both pyrometallurgical and hydrometallurgical methods at their recycling system, like Umicore in Belgium, Sumitomo in Japan, Glencore in Canada, Batrec in Switzerland, and Brunp in China. The most important companies using only hydrometallurgy are Retriev in Canada and US and Recupyl in France. *(Larouche, 2020)*

1.3.3 Direct recycling

In direct recycling the anode and cathode are separated from the battery, healed and then a new Li-ion battery is made with recycled electrodes. The materials do not have to be decomposed into separate elements since the materials are recycled to a new LIB. *(Mengyuan, 2019)*

There are different methods to direct recycling and different steps can be performed in different order. Battery can be shredded, and the electrolyte recovered. Then separation of the electrodes can be executed with mechanical, thermal, chemical, or electrochemical processes, for example precipitation, state or property change with temperature, chemical replacement or addition, gravity separation, distillation, froth flotation, surface modification, electrochemical processing, microwave, or ultrasonics. *(Mengyuan, 2019)* *(Gaines, 2021)* Then can occur cathode-healing, and building new cells with recycled cathode and anode. *(Sloopa, 2020)* Cathode healing means adding back the lithium that is lost. The amount of lost lithium can be estimated for example with spectroscopy. Cathode healing can include thermal, hydrothermal, redox mediator, ionothermal and electrochemical methods. One of many direct recycling processes is scalable cathode healing that is presented at figure 8. *(Gaines, 2021)*

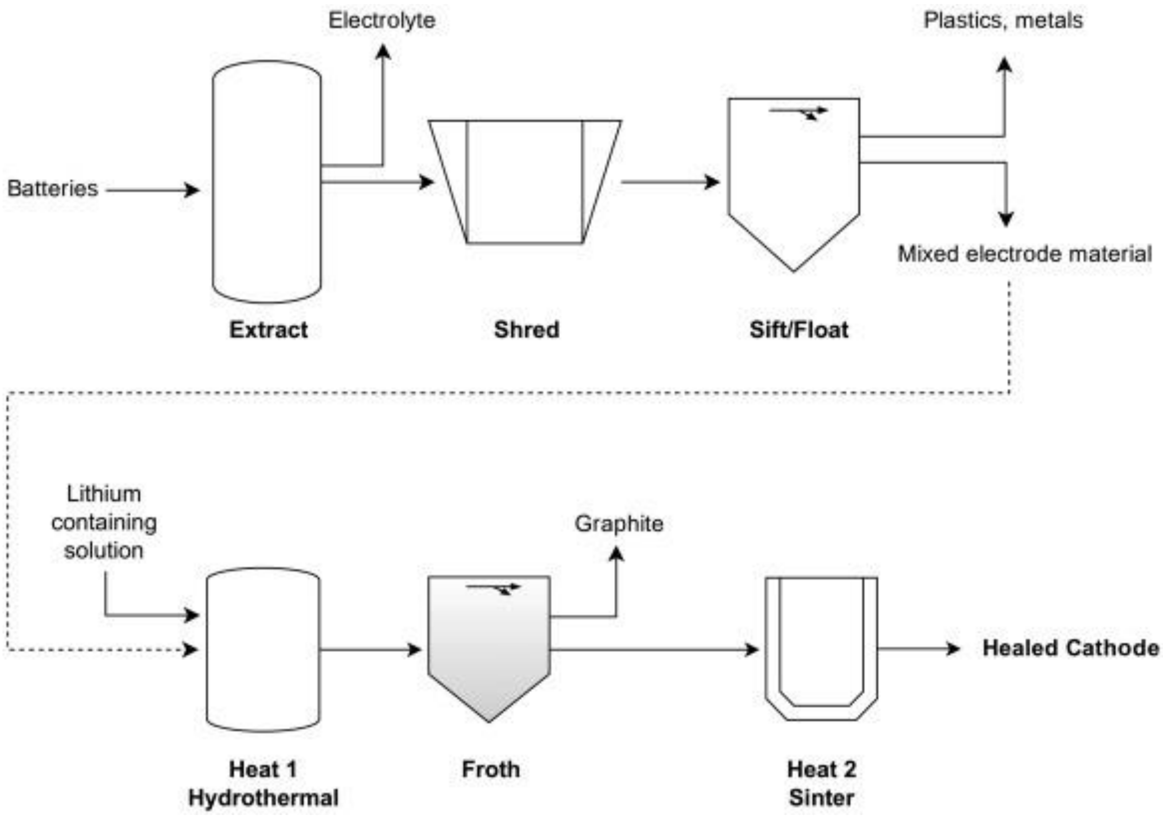

Figure 8. Scalable Cathode-healing™ process. (Sloopa, 2020)

The advantage of direct recycling is that almost all battery components except from separators are recovered and can be reused. It has lower energy consumption and emissions compared to pyrometallurgy and hydrometallurgy. *(Mengyuan, 2019)* Second life also cause less emissions than recycling. When recycling reduces emissions (g CO_2e/km) by 4%, second life reduces 22%. *(Hall, 2018)*

The disadvantage in direct recycling is the difficult battery sorting and treatment steps needed to preserve the materials. *(Mengyuan, 2019)* Direct recycling systems are created according to the cathode composition and thus the process must be designed for one type of battery at a time. Hence recycling varied batteries becomes difficult. Usually, the active cathode material has degraded and for recycled LIBs to be able to compete with new ones, the cathode needs to be healed with new lithium. *(Hughes, 2020)*

Recent progress in mechanical separation of cathode materials using selective flotation, magnetic separation and ultrasound offer promising results for improved efficiency. *(Larouche, 2020) (Hughes, 2020)*

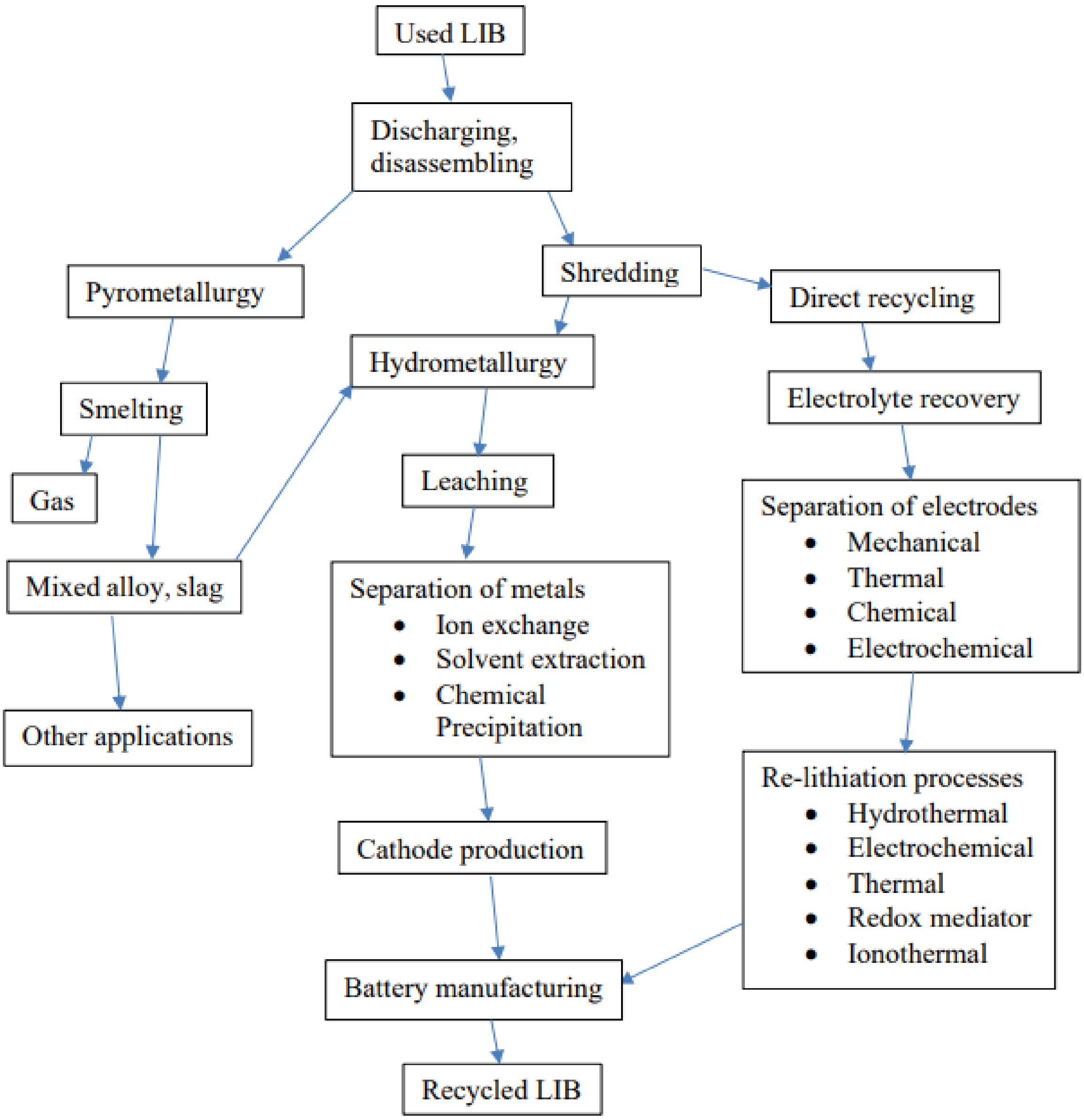

Figure 9. Main LIB recycling processes step by step.

2. METHODS

The research is carried out by collecting information from online studies on LIB recycling. The objective for references is to use studies of recent years and keep the information updated since the subject is timely, and research is constantly being made to evolve LIB recycling. The goal for seeking information is to answer the research questions and utilize mainly researches and reliable articles for references.

3. RESULTS

3.1 Recycling across the world

Over 50 companies in the world are recycling Li-ion batteries. *(Mengyuan, 2019)* China accounts for 45%, Japan 19%, Europe 15–18%, and Korea 7% of the total LIB production of the world. Companies in China and Korea pay better cost for LIB waste than companies in Europe or the USA because they reach better recycling efficiencies and so the recycling percentages are higher. Recycling companies in the USA, Canada, Europe and Japan have efficient process technologies for LIB recycling business, but do not have the amount of waste batteries to make it profitable for enough yet. *(Zhao, 2021)*

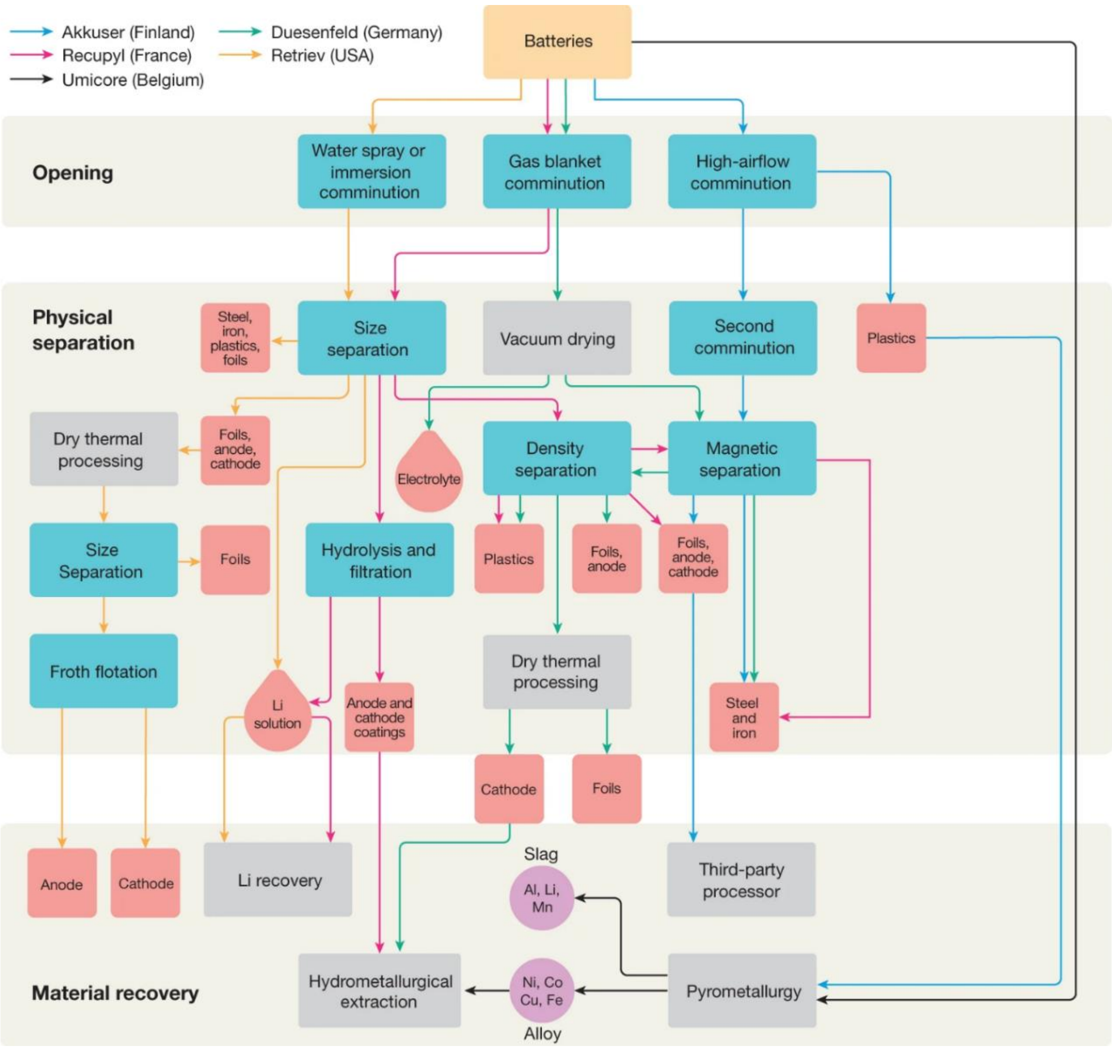

Figure 9. Different recycling systems in Europe and USA. (Harper, 2019)

Across the world recycling methods for LIBs are almost solely hydrometallurgical. The products from the process are typically chemical compounds such as nickel sulphate, cobalt sulphate, or lithium carbonate. More than one third of the recycling companies also produce precursors, for example, NMC or NCA cathodes for LIBs. *(Melin, 2019)* Five different recycling companies' (Akkuser, Recupyl, Umicore, Duesenfeld and Retriev) processes are displayed step by step in figure 9.

3.1.1 Regulations and incentives promoting recycling

The EU legislation requires that end-of-life vehicles should have been planned in a way that regaining, reusing, and recycling is easy. EU directive demands the free-of-charge shipment of end-of-life vehicles to recycling centres and forbids landfill and combustion of their batteries. The EU Battery Directive regulations insist for pickup, processing, recycling, and disposal of batteries to be paid by battery producers in relation to their market share. *(Zhao, 2021)* Germany’s regulations demand all parties within the battery value chain to take equivalent responsibility of recycling batteries. *(Zhao, 2021)*

Landfill disposal of household batteries is banned in 2006 Californian and 2010 New York battery recycling Acts. The Californian Act also classified Li-ion batteries hazardous because of the immoderate rate of Cobalt, Copper, and Nickel. It demands retailers to provide a system to gather used batteries for reuse, recycling or proper disposal and pay for it. In New York LIBs must also be labelled appropriately to ease the recycling process. *(Zhao, 2021)*

The Japanese government has provided support for LIB recycling and required producers to recycle them from 2000. In 2001, a regulation that focused on recovery and reuse of small rechargeable batteries such as LIBs came into effect. In 2013, a regulation on the advancement of reusing and recycling of used small electronics was implemented that envisioned the parts for manufacturers, retailers, processing companies, consumers, and the government in battery recycling. *(Zhao, 2021)*

In China, producer responsibility system was implemented in the beginning of 2016 through the execution of recycling policies concerning EV battery recycling systems and second life of EV batteries in other utilizations. Recycling companies and retailers get financial aid from government. Car manufactures are required to let the people to repair, give away, or exchange their used batteries. Unique IDs are used to trace batteries to ease recycling and second-life sorting. *(Zhao, 2021)*

In Australia first to prohibit landfilling e-waste and make its recycling compulsory was the Capital Territory in 2005. Later also Victoria in 2019 and South Australia in 2010 had e-waste landfill prohibited. The battery National Stewardship Scheme, formed in 2020, requires battery producers and importers to pay a fee per battery for end-of-life processing. *(Zhao, 2021)*

3.1.2 Recycling industry and non-profit companies

In the USA, recycling is mostly achieved by non-profit organizations and industry associations. The non-profit trade battery industry group Portable Rechargeable Battery Association, PRBA, promotes LIB recycling business and regulations in the USA. PRBA has created experimental battery recycling programs in Vermont, New Jersey, and Minnesota. PRBA provides LIB recycling services through its member companies including Battery Solutions, Call2Recycle®, Umicore, KBI, and Retriev Technologies. *(Zhao, 2021)*

In Germany, GRS Battery Foundation was founded as a company to collect the used batteries for recycling under the German Battery Act. It was constructed and funded by

German battery manufacturers and the electrical and electronics industry association, ZVEI. *(Zhao, 2021)*

In China, non-governmental recycling platforms have formed in Suhuishou, Aihuishou, and Huishoubao. Over 100 enterprises have the resources to dissolve LIBs and other e-waste in China. *(Zhao, 2021)* Some of the industrial LIB recycling companies are Zhejiang Huayou Cobalt, Taisen Recycling, Jinqiao Group, Brunp, GEM and Jiangxi Ganfeng Lithium. GEM has 13 automated battery disassembling and recycling centres which also manufactures the cathode precursors in China. *(Pagliaro, 2019)* Large battery manufacturers usually own recycling companies and products from battery recycling are sold to battery manufacturers. *(Pagliaro, 2019)* China can support the recycling by the economic value gained from material recovery, which is why China is the most recycling country in terms of the number of recycled batteries. *(Mayyas, 2018)*

In Australia, battery collection service is handled by organizations and retail stores, such as Mobilemuster, Aldi, Australian Battery Recycling Initiative (ABRI) and Officeworks, but LIB recycling industry still does not exist in there. Even though Australia is a major LIB producer, the recycling there is still minor. LIBs are mostly sent overseas for processing. *(Zhao, 2021)*

The Japanese battery producers that were made responsible for recycling formed recycling companies to manage with e-waste. For example, companies Nissan and Sumitomo formed 4R Energy to recycle batteries. Japanese unions are also advancing the recycling industry. *(Zhao, 2021)*

3.2 Difficulties with recycling Li-ion batteries

Significant problem with recycling LIBs is that manufacturing a new battery generally remains cheaper than recycling. However, considering the high demand of materials in the future recycling of battery materials must be realized. Current commercialized technologies are limited and recycling methods for Li-ion batteries do not make enough profit for the business to grow. *(Beaudet, 2020)* Mostly used recycling systems cannot recover all components of the battery and in direct recycling which recovers most components the battery’s capacity can weaken. There is discussion about the second life of a battery after its capacity has fallen to 80% of the original. However, at some point recycling must take place.

LIB manufacturers are progressively using more nickel and less cobalt because of cost and concerns of cobalt shortage. Cobalt is the main revenue for recycling business so replacing expensive cathode metals with cheaper ones are harming the recycling business. *(Chen, 2019)*

Li-ion batteries have many different materials in a cell that need to be recycled. Li-ion battery for automobile usually have 100 or more individual cells which could be recovered intact or distinguished. The chemical compositions of electrodes in cells vary with producer and what battery is used for, especially cathode materials are changing. Without standardization and labelling of LIBs it is difficult to build a sustainable recycling system. *(Gaines, 2014)* In most of the current recycling systems, lithium-ion traction batteries are only dismantled to a module level because of the high cost of disassembling, and then shredded. Disassembling battery packs into smaller structures the purity of different materials increases and offset later recycling processes. *(Gerlitz, 2021)* With proper labelling automated LIB disassembly systems are easier to make and automated disassembly could also reduce the risk of electric shocks or other injuries for technicians that dismantle LIBs. *(Hughes, 2020)*

Recycling still stays unequal as batteries that contain cobalt and nickel (NCA, NMC) are preferred in recycling facilities instead of those that do not contain high value metals (LMO, LFP). *(Gaines, 2014)* This has resulted to 2.00$/kg cells gate fee on LFP battery recycling in California. *(Samarukha, 2020)* Incentives, landfill fees and technical improvements for process efficiency could increase the recycling of low cost LIBs. The minor available information on LIB recycling economics makes investment by entrepreneurs in this industry unattractive. *(Mayyas, 2018)*

3.3 Possibilities of LIB recycling

The business of recycling LIBs will expectedly increase significantly in the next decade producing billions of dollars in revenue, tax income and employment. It is estimated that in the year of 2042 12 million tonnes of LIBs will be recycled with a market CAGR of 22%. *(Holland, 2021)* Investing or making business around recycling LIBs can boost local economic.

Since Li-ion batteries have been categorized as class 9 hazardous material, there are distinctly defined packaging, shipping, documentation, and labelling standards for transferring LIBs that significantly raise transportation cost. *(Mengyuan, 2019)* The high cost of transporting supports small local recycling facilities. *(Gaines, 2018) (Beaudet, 2020)*

Recycling LIBs in countries that originally have no lithium market, recycling could bring LIB materials to local market and reduce reliance on foreign suppliers. Material extraction locates in conflict zones like Congo where large part of production functions with armed compulsion and child labour. *(Gaines, 2018) (Melin, 2019)* Materials are imported from countries with monopolistic market power or environmental problems caused by the lithium mining. *(Beaudet, 2020) (Lohani, 2020) (Melin, 2019)* Production should be traceable, and mining should be focused on areas that have regulations about emissions and child or forced labour.

It is estimated that 30–50% of EV lifecycle GHG-emissions are coming from LIB manufacturing and mineral extraction. *(Beaudet, 2020)* Recycling LIBs can reduce EV's lifecycle emissions.

One way to further LIB recycling is extended producer responsibility regulation (EPR). It would make the battery producer responsible for funding collecting and recycling LIBs but also contribute battery designing that supports recycling. This approach is already utilized in China where car manufacturers and importers are responsible for end-of-life LIB collection, repurposing, and recycling. EPR is also used in EU that regulates the manufacturers to pay the cost of picking up, handling, storing, and recycling waste batteries. *(Gaines, 2018)*

3.4 CO_2 emissions from LIB recycling

Information about CO_2 -emissions from recycling Li-ion batteries is limited due to lack of shared data within the LIB recycling industry. Emissions depend on battery type, the number of cells the battery has, transportation distance, energy source and recycling method used.

Emilsson and Dahllöf estimates in their study "Lithium-Ion Vehicle Battery Production" *(Emilsson, 2019)* that with a 100% electricity powered cell producing and battery pack assembling factory, producing a new NMC111-LIB from virgin materials emits 61-106kg CO_2-eq/kWh battery capacity when varying the electricity mix from a clean (0kg CO_2-eq/kWh) to a fossil fuel rich (1kg CO_2-eq/kWh). Varying the electricity only when natural gas is used for heating the emissions range from 70-77kg CO_2-eq/kWh battery capacity.

Iryna Samarukha's study *(Samarukha, 2020)* gives specific information about different recycling methods CO_2 emissions (kg CO_2-eq/kg cell) and the revenue received when recycling LIBs from heavy electric vehicles. Another study about recycling emissions is Darlene Steward, Ahmad Mayyas and Margaret Mann's research *(Steward, 2019)* about economics and recycling LIBs from end-of-life vehicles. These studies are used in the next sections for comparison.

Information about recycling emission is only found on NCA, NMC, LMO and LFP batteries therefore those are reviewed and compared.

3.4.1 NCA cells recycling

Iryna Samarukha reports that CO_2-emissions with pyrometallurgical recycling are 2.19kg CO_2-eq/kg cell and 2.13kg CO_2-eq/kg cell from hydrometallurgical recycling method. NCA recycling becomes unprofitable if batteries are transported over 800km. Hydrometallurgy has higher CO_2 emissions and is more expensive because of the production and transportation of reagents. *(Samarukha, 2020)*

The calculated income from recycling 1kg of NCA cells are 3.41€/kg cells for pyrometallurgical recycling and 3.54€/kg cells for hydrometallurgy. *(Samarukha, 2020)*

3.4.2 NMC recycling

Samarukha's study informs that emissions from NMC11 recycling with pyrometallurgy are 2.1kg CO_2-eq/kg cell and with hydrometallurgy 2.13kg CO_2-eq/kg cell. *(Samarukha, 2020)* Steward, Mayyas and Mann's study states similar results with pyrometallurgy and gives also results about direct recycling. When using virgin raw materials in NMC333 battery, emissions are estimated to be 9kg CO_2-eq/kg cell. When recycling NMC333 battery with pyrometallurgy carbon dioxide emissions decline by 78% and when recycling with direct recycling by 94%. Recycling NMC333 with pyrometallurgy then emits 1.98kg CO_2-eq/kg cell and recycling with direct recycling 0.54kg CO_2-eq/kg cell. *(Steward, 2019)*

Revenue from pyrometallurgy is calculated to be 4.68€/kg cell for NMC111battery, 3.77€/kg cell for NMC622-battery and 3.15€/kg for NMC811-battery. Revenue from hydrometallurgy is 4.99€/kg cell for NMC111-battery, 4.01€/kg cell for NMC622-battery and 3.33€/kg cell for NMC811-battery. Hydrometallurgy is generally more profitable but there is a lot of variation depending on the cathode composition. *(Samarukha, 2020)*

3.4.3 LMO recycling

Samarukha's study reports emissions from recycling LMO cells are 1.95kg CO_2/kg cell in pyrometallurgical recycling and 2.15kg CO_2-eq/kg cell from hydrometallurgical recycling. *(Samarukha, 2020)* Steward, Mayyas and Mann's study states that making LMO battery with virgin materials produces 5kg CO_2-eq/kg cell. Recycling with hydrometallurgy is estimated to emit 5% less CO_2 emissions and with direct recycling 10% less. Emissions from hydrometallurgy is then 4.75kg CO_2-eq/kg cell and direct recycling 4.5kg CO_2-eq/kg cell. *(Steward, 2019)*

Recycling LMOs using pyrometallurgy produces revenue 0.75€/kg and using hydrometallurgy 1.49€/kg. *(Samarukha, 2020)* LMO batteries' low value makes the recycling not profitable.

3.4.4 LFP recycling

CO_2 emissions generated from recycling LFP cells are 2.16kg CO_2-eq/kg cell for pyrometallurgical method and 2.36kg CO_2-eq/kg cells for hydrometallurgical. *(Samarukha, 2020)*

Revenue from pyrometallurgical recycling is 0.69€/kg cell and from hydrometallurgy 0.80€/kg cell. *(Samarukha, 2020)* Recycling of LFP batteries has the lowest revenue compared to LMOs, NMCs and NCAs.

3.4.5 Transportation

Transportation of 1 kg of used LIBs emits 12.7g CO_2-eq/100km due to fossil-based fuels. *(Samarukha, 2020)* That makes the emissions for 1kg of LIBs 0.127g CO_2 per kilometre.

As transportation costs are the most expensive part of the recycling process, international agreements, and regulation to reduce transportation costs internationally is important. *(Gaines, 2014)*

4. CONCLUSIONS

Recycling methods across the world for LIBs are mostly hydrometallurgical. The end products are typically chemical compounds such as cobalt sulphate, nickel sulphate or lithium carbonate and more than one third of the recycling companies also produce precursors, for example, NMC or NCA cathodes for LIBs.

Recycling industry struggles to make profit from recycling the lower value batteries such as LMO and LFP batteries and thus higher value LIBs are more commonly recycled, and only high value ingredients are recovered. LIB manufacturers are using higher nickel and lower cobalt chemistries because of cost and adequacy concerns of cobalt which further decrease the value of recycling. Additionally, batteries vary heavily in type and components so standardization and marking system is needed to make recycling efficient and safe.

LIB recycling industry has a lot of opportunity since the market for LIBs continues to grow and regulations or incentives for LIB recycling are necessarily needed at some point. Recycling business can build local economic value, bring materials into the local market, and decrease dependence on foreign suppliers that may produce materials at the cost of human rights or environment. Laws about recycling and incentives can increase LIB recycling and producer responsibility has shown great results executing LIB recycling in China.

Pyrometallurgical recovery is relatively risk-free and simple way to recycle LIBs but fails to recover all elements and offers low economic and environmental benefits. Hydrometallurgy needs low energy consumption, lithium can be recovered, and it has good efficiency, but it is relatively expensive, and separation of electrodes is needed. Direct recycling can recover almost all components, has low energy consumption and emissions but needs battery standardization to be functional.

Most of the CO_2 emissions are generated from transportation. Hydrometallurgical recycling emits more CO_2 emissions than pyrometallurgical recycling in all battery types except in NCA battery's case. Direct recycling emits least CO_2 emissions.

The best economical value is obtained from recycling NCA and NMC batteries. Cobalt, nickel, and metallic materials are currently largest source of revenue for recycling companies. Hydrometallurgy is more profitable than pyrometallurgy with all battery types because it recovers higher proportion of material and pyrometallurgy requires expensive gaseous wastewater treatment plants.

When NMC333 batteries are recycled with pyrometallurgy CO_2 emissions decrease by 78% and with direct recycling by 94%. Recycling can reduce emissions significantly and prevent shortage of lithium and cobalt resources.

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