Tampere University of Applied Sciences

End-of-life Management for Large-scale Lithium-ion Batteries

A literature review

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ABSTRACT

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End-of-life Management for Large-scale Lithium-ion Batteries
A literature review

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Lithium-ion batteries are on the rise because of the trend for renewable energy and electric vehicles. Amassing in large quantities, posing possible and major consequence risk, lithium-ion battery waste critically needs a responsible handling plan. This thesis was to communicate a comprehensive battery waste management strategy to holders and future disposers of large-scale lithium-ion batteries. To meet this objective, a literature review was conducted on three types of documents, which are (i) international legislation and treaties, (ii) industrial reports conducted by consultant agencies, and (iii) publications in academic and scientific journals.

The journey of battery waste was identified. Planning for the end-of-life required a formal Decommission Plan, which oversaw risks factors, safety measures, and socio-environmental impacts. Waste transportation adhered to international treaties specifically applicable to end-of-life lithium-ion batteries. The containment of battery waste must be made of protective materials and properly labelled. Eligibility for second-life application was determined by batteries' remaining electric capacity storage and safety performance. Despite environmental benefits, second-life batteries were challenged by falling costs of new batteries. In term of recycling, hydrometallurgy was highly efficient and most popular recycling method for end-of-life lithium-ion batteries. After acid leaching, solvent extraction, and precipitation, cobalt, nickel, manganese, and lithium were extracted from battery scraps. Ultra-high heat pyrometallurgy was reportedly only operational at two recycling plants in Japan and Belgium; however, low-to-medium heat treatment were commonly implemented for removal and deactivation. Currently under development direct cathode recycling may have large potential in battery recycling. The refurbished cathode could be used in new battery manufacturing. For demonstration, this thesis included relevant examples of commercial recycling operators and a case study of a preliminary battery waste management plan for an R&D energy storage project.

This thesis conclusively recommends disposers of large-scale lithium-ion batteries to plan diligently for the decommission phase, follow laws in package and transportation, conduct battery diagnostic tests then consider for second-life, opt for hydrometallurgy recycling operators, meanwhile donate one battery module to research centers for developing direct cathode recycling technique.

Key words: large-scale lithium-ion batteries, waste management, recycling

CONTENTS

GLOSSARY

EOL	End-of-Life
EV	Electric vehicles
LFP	Lithium iron phosphate
Li-ion	Lithium-ion
LMO	Lithium manganese oxide
NCA	Nickel cobalt aluminium
NMC	Nickel manganese cobalt
SOC	State of health
SOH	State of charge

1 INTRODUCTION

1.1 The global demand for lithium-ion batteries and the need for their end-of-use management

The world is entering another industrial revolution. Fossil fuel has reached technological maturity and entered the obsolescence phase. The electric grid receives an increasing contribution from solar and wind farms; fully electrified transportation is competitive to internal combustion engine counterparts.

Among a wide array of technology for energy storage, and electrochemical technology for battery, rechargeable lithium-ion battery (Li-ion battery) owes its market popularity to competitive advantages in high energy with light weight and small volume, as well as long cycle life (Miao *et al.* 2019). Lithium-ion batteries are historically used in portable devices, namely laptops, smartphones, cameras, and household electronics. Contributing to the electrification of the transportation industry, these batteries are now so cheap, light, and efficient to enable the technological and economic feasibility of fully electric vehicles. In the grid and utility industry, lithium-ion large-scale energy storages serve in applications of grid stability, frequency regulation, peak shaving, and energy trading (EESE/EERA 2016). Li-ion energy storage is also prominent in integration with renewable energy sources of wind power, such as Tesla's 120 MWh Powerpacks at Hornsdale Wind Farm in Australia, or solar power, such as 590 battery packs with 4,200 rooftop photovoltaic panels at Joahn Cruyff Arena in Amsterdam, Netherland.

Driven by those phenomena, lithium-ion batteries experience a rocketing increase in demand and production, in addition to their already abundant uses in portable electronics. In 2019, there are accumulatively more than 1,250,000 tons of li-ion batteries on a global scale (Melin 2019). The question arises, what consequences will be when those batteries reach the end-of-life.

Waste batteries have been accumulating in mass amounts on Earth each year, mostly resulted from discarded (smart)phones and laptops. A baseline study for end-of-life Li-ion batteries conducted in 2019 revealed that by 2018 there were

200,000 tons of Li-ion waste. In 2020, batteries from electric vehicles are reaching their predicted end of lifetime, and in the future of 2050, many large-scale energy-storages will also reach the decommission stage (Melin 2019).

Battery waste imposes major consequence risks on humans and the environment if left irresponsibly. For example, Li-ion batteries are prone to thermal runaway, which means a battery can heat itself excessively in extreme temperature condition. Because of thermal runaway, fire or explosion accidents have happened in storage facilities. If Li-ion batteries are to be discarded in landfills, the electrolyte as well as metals leach into soil and seep through underground water. If they are to be burnt indiscriminately, the combustion of fluoride content leads to the formation of hydrogen fluoride, a highly toxic gas. (Harper *et al.* 2019.) In fact, the Battery Directive 2006/66/EC imposes a ban on incineration and landfill of any industrial or automotive battery type, without proper prior treatment.

The supply chain for materials of lithium-ion batteries is also in great concern, particularly for cobalt being considered a critical raw material by the European Union. Most global supply for mined cobalt comes from a country with records of socially irresponsible practices on miners who are working in dangerous and abusive conditions. In addition, cobalt mining is a by-product of mining copper and nickel, therefore the supply of cobalt not only depends on its demand but also the demand for the parent metals. (Azevedo *et al.* 2018.) Therefore, industry actors attempt to prolong the lifespan of batteries, as well as viewing battery scrap as metal reservoirs to extract cobalt. These will reduce the reliance on mining sources and strengthen the supply chain.

For the above reasons, the need for a socially responsible, safe, minimum environmental impact, and economically viable waste management of lithium-ion batteries is a real and urgent matter.

1.2 Lithium-ion battery - a diverse technology

One Li-ion battery cell consists of two electrodes, which correspond to cathode and anode, terminals on each side for collecting the current, the conductive electrolyte, a separator, and the outer casing. When a cell is connected to a load, ionic lithium particles detach from the cathode, migrate in the electrolyte, pass the separator, deposit to the anode. The movement of lithium ions creates free electrons in the anodes; electrons then travel towards the anode-side current collector, flow through the load to cathode-side collector. The process reverses when the battery is in charging mode; lithium ions migrate from the anode back to the cathode. Figure 1 visualizes the components and operating mechanism for a Li-ion battery cell.

In actuality, the term "lithium-ion battery" generally describes a family of battery technology that utilizes that mechanism, but differs widely in materials for cathode, electrolyte, and anode. The composition for cathode materials are metal oxides; different metals are pure cobalt (Lithium Cobalt Oxide LCO), pure manganese (Lithium Manganese Oxide), a mixture of cobalt, manganese, nickel (Lithium Nickel Manganese Cobalt Oxide NMC), a mixture of cobalt and nickel doped with aluminum (Lithium Cobalt Nickel Aluminum NCA), and Lithium Iron Phosphate (LFP).

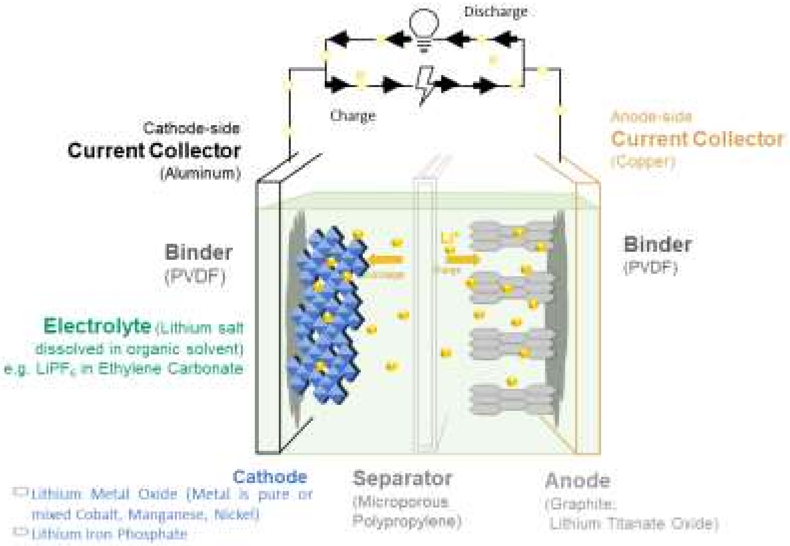


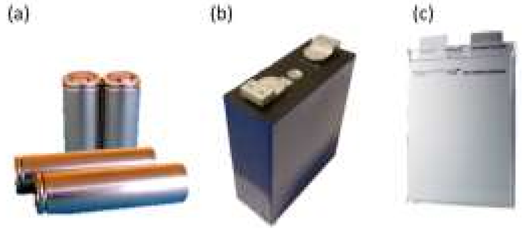
FIGURE 1. Operating mechanism, components, and components' materials in a Li-ion battery cell. Recreated work from Valio 2017.

The anode material is carbon-based graphite. Recent technological advancement introduces Lithium Titanate Oxide (LTO) in conjunction with the LFP cathode, but its application is still limited. The electrolyte is a lithium salt dissolved in an organic solvent, for an instance, $LiPF_6$ dissolving in ethylene carbonate. It also exists in gel-form in Lithium Polymer or solid-form in Solid-State batteries.

The variations in cathode and anode technology lead to different performance ratings among Li-ion types; therefore, each technology is more suitable in an application than other ones. Figure 2 displays these trade-offs in five parameters. *Specific energy* means the battery's electric storage capacity relatives to its volume. *Specific power* means the battery can output high current within a period of time. S*afety* shows, for example, a low chance of internal heating during full or high charge. *Performance* indicates the battery operation at extreme temperatures. *Life span* correlates to the number of charging cycles of the battery cell, and how long can the battery be used until deteriorated to 80% of its original capacity. (Battery University 2019.) Battery with high specific energy and specific power are crucial parameters for electric vehicles. However, those two parameters are not as important in stationary large-scale energy storage, where maximum capacity, cell degradation, life span, material cost are more concerning.

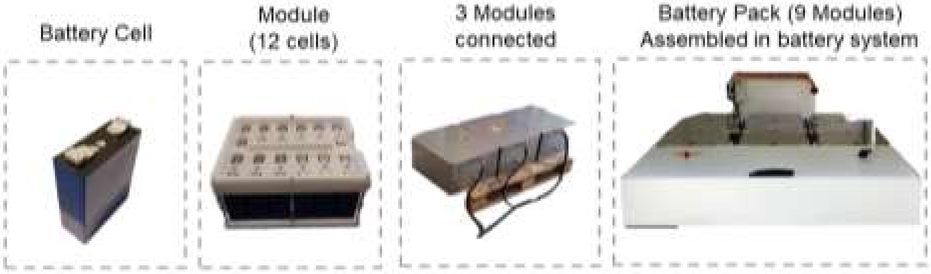
FIGURE 2. Relative comparisons among different Li-ion battery cathode technology on the market. Recreated work from Battery University 2019 and Miao *et al.* 2019

Apart from the material variations, size and shape also vary in lithium-ion cells. The cylindrical cell has the advantage of uniform manufacture and standardized sizing. The disadvantage is the inefficiency in physical space when connecting cylindrical cells together. The prismatic cell has a rectangular solid case, and the pouch cell takes the form of a bag. These two configurations have more efficient spacing when making a battery module from cells. However, they suffer from size expansion, which must be considered when building modules. The manufacturers for these two configurations may have their proprietary design, and therefore there may be no international standardized size. (Romare & Dahllöf 2017). Illustration for three cell shapes are displayed in picture 1.



PICTURE 1. Lithium-ion battery cell shapes. a) cylindrical cell, b) prismatic cell, c) pouch cell (Image source: Battery University 2019, Korpela 2018)

Lithium-ion batteries can deliver such high power and voltage for electric vehicle and energy storage uses because multiple cells are connected, in series or parallel, to each other and form a battery module. Battery modules are then connected to form the battery pack. The pack is then assembled to a system with peripheral electronics, for example, DC-AC inverter, Battery Management System, Thermal Management System (Hesse 2017). Picture 2 shows expansion layers of battery connections.



PICTURE 2. Levels of battery assembly: cell, module, pack, system. Photos from e-mail communication with the project manager of Tampere University of Applied Science's ENERVARA energy storage project.

2 RESEARCH OBJECTIVES & METHODOLOGY

This thesis's objective is to clarify the waste management strategy for large-scale lithium-ion energy storage, with an emphasis on the recycling processes. The primary target audience of this thesis is any holder of such batteries who ceases to use and wants to dispose of those bulk. In addition, this thesis can be used as a reference material to pinpoint the current state and the missing link of a sustainable waste management strategy for lithium-ion batteries. Based on it, future studies, research, innovation, and development projects can be conducted.

The scope of this thesis focuses on lithium-ion batteries for large-scale energy storage applications. The scope also considers a possibility that the batteries have first use in electric vehicles and second use in stationary energy storage. Within the variations in material choices, most information reports that large-scale lithium-ion energy storage will commonly use cathode of NMC, NCA, and LFP, graphite anode, and lithium salt in a solvent electrolyte (Adolfsson Tallqvist 2019, Harper *et al.* 2019, Or 2019, Korpela 2018). Regarding cell shapes and sizes, all configurations and various sizes are reportedly used in the large-scale application (Hesse *et al.* 2017).

This thesis will be conducted as a literature review and will analyze secondary data. Legislative documents, industrial reports, and academic researches and reviews are types of literature. Relevant legislations were viewed at international treaties, European Union Directives, and Finnish national Acts. Scientific literature for researches, experiments, and reviews were obtained from ResearchGate, ScienceDirect, Nature. The search terms include the object "electric vehicles batteries", "Lithium-ion batteries", "energy storage", with the subject "recycling", "recycling process(es)", "end of life". Industrial reports are obtained by having the search term in the Google engine. The reports compiled or commissioned by reputable organizations, such as Business Tampere, U.S. Energy Storage Association, Swedish Energy Agency, are included in this thesis's research range.

3 END OF LIFE MANAGEMENT FOR LITHIUM-ION BATTERIES

Waste Framework Directive (2008/98/EC) defines waste management as the comprehensive journey of collection, transportation, storage, recovery or other appropriate treatment strategies, monitoring, and reporting. This chapter will follow that guideline and present relevant results. First, considerations for decommissioning, collection, and transportation of Li-ion batteries are presented. Next, the second-life application of Li-ion batteries is reported. Finally, current recycling on commercial scale is described, with some mention for innovative concepts on laboratory or pilot scale to approach battery recycling.

3.1 Decommission, Collection, Transport

Li-ion batteries are stated to have their service lifetime ten to twenty years in grid energy storage (EPRI 2017), or seven years as second-life energy storage (discusses further in chapter 3.2 Second-Life Reuse). These estimations are based on the time when the battery can only store 80% of its original capacity. A battery management system can provide necessary data to predict and indicate when that time happens so that energy storage operators can plan beforehand.

When a battery energy storage receives decommission decisions, battery packs will be detached from peripheral electronics such as the battery management system, coolant system, inverter. Battery packs may also be dismantled to a manageable unit for convenience of transportation. During these operations, there are possible, major consequence occupational risks of high voltage and thermal runaway. Therefore, it is advisable for the disposers of energy storage to carefully assess risk issues, safety plans, disturbance to the surrounding environment including humans and nature in a formal Project Decommissioning Report (EPRI 2018).

For the collection of waste, the Waste Framework Directive (2008/98/EC) institutes the Extended Producer Responsibility on waste producers, which states that the responsible party must set up a collection scheme for the waste. Battery

Directive (2006/66/EC) stipulates this responsibility specifically on producers of battery waste. Therefore, the legal obligation is put on battery distributors to handle the decommissioned energy storage. Battery operators and battery suppliers should communicate and cooperatively scheme and finance for the end-of-use management plan.

Li-ion batteries are categorized as Class 9A dangerous goods, under a larger classification for Miscellaneous Hazardous Materials. Transportation of lithium-ion batteries follows rigorous legislations for different transportation modes (e.g. road, rail, or air), as well as special provisions and packaging instruction accordingly to specific cases.

For instance, large-scale Lithium-ion batteries, intended for transportation on road and for recycling, will follow the freight code UN 3480, ADR Special Provision 377, and the Package Instruction P909. Therefore, the batteries' terminals are sealed with insulation tape to prevent external short circuits. Containers should provide prevention and protection against thermal runaway, have a strong, impact-resistant outer casing, and secure the batteries with cushion material. In addition, markings on the containers should include the UN specification code, the Class 9 cautious label, and the text specifying its destination's purpose as shown in picture 3. Detailed specifications for containers and markings are referred to the ADR Book 2017. (ADR Book 2017, Lithium Battery Service GbR 2017.)

PICTURE 3. Markings on a container when transporting lithium-ion batteries for recycling

3.2 Second-Life Reuse

The waste hierarchy outlined in the Waste Framework Directive (2008/98/EC) refers that re-using is a more desirable strategy than recycling. Therefore, battery waste is expectedly receiving second life and refurbishment treatment. Currently, electric vehicle manufacturers are pioneers in projects of second-life batteries; they often form partnerships to implement the batteries from their retired vehicles in technological and business ventures of energy storage (table 1).

TABLE 1. Examples of electric vehicles batteries' second life in energy storage

Automakers	Partner	Second-life Battery Project
Hyundai	Wärtsilä	1 MWh energy storage system consists of Hyundai IONIQ Electric batteries
BMW	Bosch	2.8 MWh energy storage facility with 500 BMW i3 batteries
Mercedes-Benz	Mobility House, Getec Energie	8.96 MW/ 9.8 MWh energy storage plant from 1,920 batteries modules from EV
Nissan	Eaton, Mobility House	250 packs from Nissan Leaf, in 590-pack energy storage with PV system in Joahn Cruyff Arena, Amsterdam

State of health (SOH) is crucial for deciding if the battery is suitable for second-life applications. At the remaining capacity of 70 to 80% original specification, Li-ion batteries still can offer service while their safety and degradation performance are ensured. Moreover, a diagnostic test is even recommended to be conducted on each battery module or even on each cell, because at a certain degrading point, known as "aging knee", battery cells will degrade so fast that they must be denied in either vehicles or stationary storage for safety concerns.

In addition to batteries' state of health upon first life retirement, their performance during the second life must also be evaluated. During second life operation, depth of discharge, charge-discharge rate (C-rate), and cycles per day are determinants to the longevity and safety of batteries, hence influencing the refurbishment efforts and costs for the re-used batteries. Besides successful demonstration by automakers and partners, piloting and R&D projects have been re-using li-ion batteries in numerous scenarios, such as off-grid with solar cells, supporting national grid, utility ancillary service, EV charging station, or uninterrupted backup power, to testify the suitability of second-life batteries in such applications.

Refurbishment process for retired batteries is technically challenging. The battery pack may consist of modules with different chemistry types, and with different state of health. Due to different voltage operations, energy, and power capabilities among modules, the battery pack requires a more advanced management system (Energy Storage Association 2020, Martinez-Laserna 2018). For one solution, Hart and colleagues (2014) proposed an algorithm for the management system of second-life energy storage consisting of mixed NMC and LFP modules.

The profitability of second-life energy storage must be attractive enough to be competitive with new batteries on the market. The trading price of re-use batteries is estimated as $60 to $300/kWh (Melin 2018); in a baseline study in the context of the U.S. in 2012, second-hand batteries were estimated to cost $44 to $180/kWh (Martinez-Laserna 2018, Neubeuer *et al.* 2012). Meanwhile, the cost of a new battery pack is decreasing and expectedly at €150/kWh ($177/kWh) in 2030 (EESE/EERA 2016); however, Statista reports that the price of a lithium-ion battery pack is at $156/kWh in 2019. The refurbishment operations must be more effective so second life batteries may be competitive with the price of new ones.

Re-use of EV batteries in stationary energy storage brings environmental benefits in multiple ways; that has been illustrated in scientific literature (Melin 2019). Energy storage is complementary and shown to promote the market penetration and growth of renewable energy, especially solar power in household, commercial, or industrial settings. The utility industry has the potential to greatly reduce greenhouse gas emission, if it substitutes gas or diesel power plant with large-scale stationary energy storage and photovoltaic for peak shaving. Not only excelling fossil fuel, but lithium-ion batteries are also a great replacement for lead-acid, which is a battery type previously used in many utility applications but entails risks of human toxicity and environmental hazard because of high lead concentration. The option to use second-life batteries reduces the requirement for virgin and newly produced battery packs, therefore reducing resource depletion, energy consumption, emissions, and footprints associated with mining.

In general, although offering environmental benefits, second life lithium-ion stationary storage face great challenges in technicality and market price competitiveness. Although re-use is recommended before recycling as outlined in the Waste Framework, the Battery Directive does not impose any mandatory actions for disposers of large-scale batteries to consider the second life option. Industry actors are increasing efforts to make this idea viable and more attractive.

3.3 Recycling operations

Being re-used or not, lithium-ion batteries will eventually reach their ultimate end-of-life. The Battery Directive makes a ban on disposing large-scale batteries directly into landfills, so most used industrial batteries will be recycled. The Waste Framework defines recovery is also retrieve the valuable components of a product so that it can be re-introduced back into the industry. Among components in a Li-ion battery, the metal constituents in the cathode are the recovery targets for most recycling operators

An generalized frame for recycling Li-ion battery scrap is shown in figure 3. It usually starts with the preliminary treatment phase; battery systems are dismantled into a manageable unit, removing the housing, electronic peripherals, welding components, and only battery at the module level is left. This step is greatly alleviated if battery packs are already dismantled at operation site before shipped to recyclers.

Some recycling operators include a distinct phase for discharging leftover electricity to reduce thermal runaway risk. This is achieved by submerging the battery unit into a brine solution or using a discharge device. Some operators, however, opt to combine deactivation while crushing or burning battery waste to reduce operational complexity and costs. The deactivation method can be crushing the battery in an inactive environment, for example with water spray, nitrogen air (**Retrieve**), supercritical carbon dioxide, a combination of carbon dioxide and argon (**Recupyl**), or drying the comminuted unit for electrolyte removal (**Duesenfeld)**. While deactivation of battery while crushing requires less step, in the case of electric vehicles and large-scale energy storage, the voltage is high, and maybe it is recommended to discharge the battery to a safe level beforehand. For example, **Accurec** stated that in its recycling operation automotive Li-ion battery will be discharged below 60V.

3.3.1 Physical separation – the use of mechanics

Recycling operations usually implement the comminution technique; the incoming pre-treated battery waste enters a shredding and cutting machine, in which it is ground to fragments. The fine particles are often known as the black mass, consisting of materials in the anode and cathode; the coarse fractions consist of other components like plastic casings and current collectors.

Then, different separation techniques are employed to physically separate components from the mainstream (process [1] in figure 3). Sieves and screens are deployed to divide fine and coarse particles. Shaking tables aid the gravitational separation with its vibration. A magnet is used to sort out steel casing and ferrous metal. Zig zag gravity separators are utilized to separate current collectors from the stream. Froth flotation may be used to segregate anode component from the cathode in the back mass due to differences in the hydrophilic property.

Some industrial recycling plants, for example, **uRecycle** in Sweden and **Akkuser** in Finland, focus entirely on the mechanical treatment for battery waste. The output black mass is later sent to their external partners for metallurgical treatments and recovery of valuable components.

3.3.2 Pyrometallurgy - the use of heat

Smelting is the usual reference to pyrometallurgy. Battery waste is sent to an ultra-high temperature furnace at 1200-1450°C, without discharge or mechanical separation (process [2] in figure 3). The battery waste unit is reduced to (i) metal alloy, containing cobalt, nickel, copper, traces of iron, and (ii) slag, containing lithium, manganese, iron, aluminum. The smelting process consumes aluminum from the foils and current collectors, graphite from the anode, lithium-salt and organic compounds in the electrolyte, and plastic as the burning fuel; in other words, those mentioned components are expected as lost in this process. **Umicore** and **Sony-Sumitomo** recycling operations are often mentioned in literature as examples for pyrometallurgical smelting.

The metal alloy is further treated with hydrometallurgy (chaining process [2] to process [3] in figure 3) to selectively recover valuable metal. The slag is reported to be sold for use in construction materials. Because slag contains lithium, there is interest in the recovery of lithium from slag if the economic value of recovered lithium is attractive in the future. The by-product gas containing volatile organics or hydrogen fluoride, as consequences of combustion of fluoride and plastic, must be addressed by the additional gas treatment process.

Another technique followed under the pyrometallurgy category is pyrolysis. **Accurec** uses a rotary kiln at 800°C after pre-treatment (chaining process [1] then [2] in figure 3), or **Umicore** also has a 700°C pyrolysis chamber before smelting. This operation is to reduce undesirable contents such as plastic or graphite and output a more desirable compound downstream. The uses of heat at moderately high temperature 300°C or lower temperature max.80°C for evaporation of the electrolyte are also reported at several recycling operations.

3.3.3 Hydrometallurgy – the use of chemicals

The hydrometallurgical leaching process employs strong acid, usually HCl or H_2SO_4, with an assistive oxidizing agent such as H_2O_2. The black mass or metal alloy exposed to strong acid will leach into a liquid solution that contains metallic ion. In the leach solution, cobalt, nickel, and lithium are considered desirable, while copper, iron, and aluminum are considered impurities.

From the leachate containing a mixture of metal ions, a different set of chemical applications are used onto the leachate to selectively recover the metals ("Metal extraction" box in figure 3). Solvent Cyanex 301 may be used to extract a mixed solution of manganese and lithium from a solution of cobalt and nickel; then, solvent Cyanex 272 may be applied to separate cobalt and nickel (Hanisch *et al.* 2018). The precipitation of Lithium often occurs lastly in the material recovery chain. This is achieved, for example, by the addition of sodium carbonate to form precipitation of lithium as lithium carbonate, or the addition of sodium phosphate to form lithium phosphate precipitate. (Hanisch *et al.* 2018.) Figure 4 illustrates these techniques in operations at **Duesenfeld**'s recycling plant.

3.3.4 Innovative recycling operations and experiments

Apart from commercial processes, laboratory researches and pilot-scale operations have made significant progress in innovative recycling solutions. Experiments in hydrometallurgy using organic acid or alkaline compounds to replace industrial choice of strong HCl acid will reduce human hazards and environmental toxicity. The bio-metallurgical process exploits certain bacteria and fungi that can metabolize metals and extract in nano-particles (process [4] in figure 3).

Copper and nickel, because of similar precipitation pH ranges, require extra solvent extraction step in hydrometallurgy as illustrated in figure 4 (Valio 2017, Zou *et al.* 2013). To address that issue, experimental designs have been proposed to directly refurbish the leachate (process [5]), adjusting the leachate to the correct ratio of manganese-nickel-cobalt by adding appropriate chemicals, and sintering with additional lithium carbonate to adjust lithium content (process [7] and [8]). The output is the cathode precursor for the NMC type (Worchester Polytechnic Institute, Zou *et al*. 2013).

Battery Resources in the U.S. and **Lithorec** project in Germany include a refinery stage within their recycling operations. The recovered compound of cobalt, nickel, and manganese from previous operations are thermally refined with previously recovered or new lithium carbonate (process [7] and [8]). The output is cathode precursor powder. Moreover, U.S based **OnTo** technology and Argon National Laboratory's **ReCell** research and development center promote a direct, old cathode to new cathode recycling pathway (process [6]).

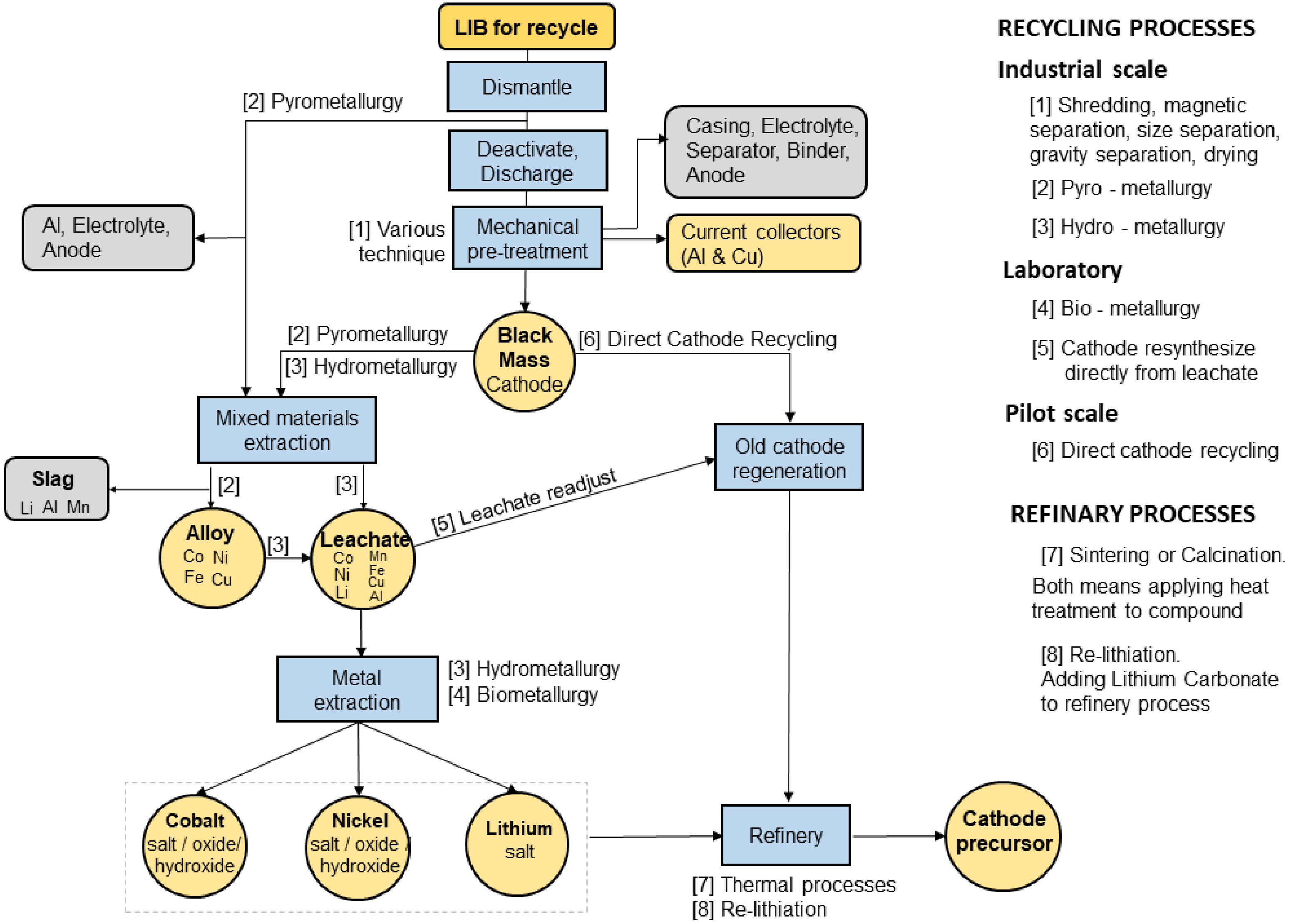
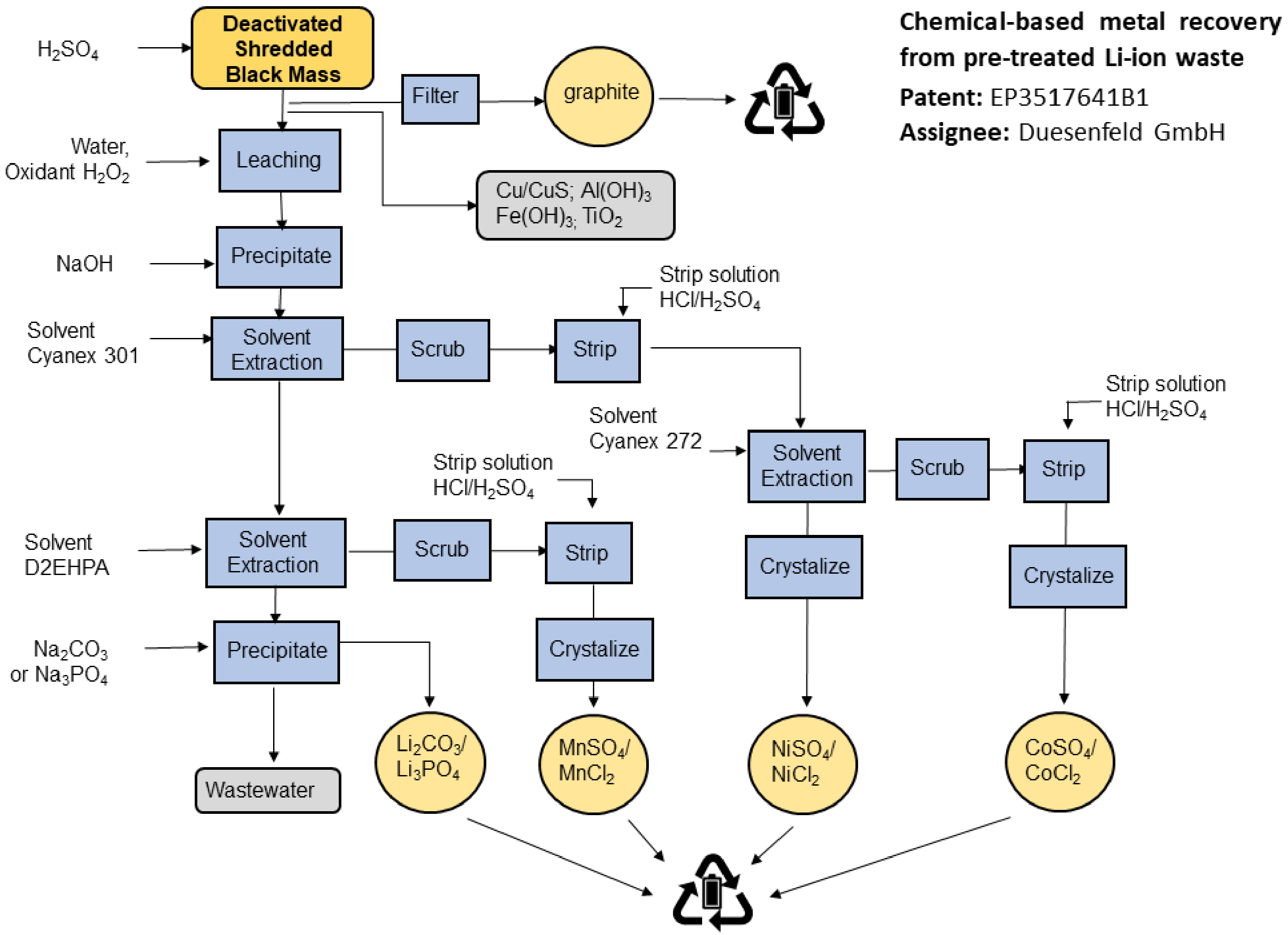


FIGURE 3. Recycling for Li-ion battery waste with two possible pathways. Commercial operators aim at the recovery of constituent metals, displayed on the left of the black mass. Other paths, still in the laboratory or pilot stage, aim at more efficient ways to create cathode precursor compounds, displayed on the right of the black mass.



Chemical-based metal recovery from pre-treated Li-ion waste

Patent: EP3517641B1

Assignee: Duesenfeld GmbH

FIGURE 4. A majorly simplified flow for Duesenfeld GmbH's patented process for chemical-based metal recovery from pre-treated li-ion battery waste. Hydro-metallurgical techniques include leaching, stripping, precipitation, and solvent extraction (Inventor: Hanisch, Elwer, & Brückner 2018)

4 DISCUSSION ON RECYCLING

Analyzing the above information about recycling operations, this section will be dedicated for evaluation. To be specific, there are discussions for (i) advantages and disadvantages for traditional pyrometallurgy, modern hydrometallurgy, and futuristic direct cathode recycling, (ii) recovered and loss elements, along with the possibility for closed-loop battery's material cycle, and (iii) recycling different cathode chemistries.

4.1 Conventional, modern, and futuristic recycling processes

Pyrometallurgy has matured technology because of long historic applications in recycling battery other than lithium-ion. That also means the process can receive an input of miscellaneous battery types and lithium-ion cathode types. The process, however, uses a lot of energy for smelting, emits substantial carbon dioxide, and needs to address hydro fluoride gas emission. The recycling efficiency is also low because only high-melting-point metals are recovered after the process.

Hydrometallurgy is considered state-of-the-art for battery recycling because of a very high recovery rate and large-scale feasibility. Claimed recycling efficiency rates by commercial operators are 60% by **Accurec GmbH** (appendix 1.2), more than 80% by **Fortum**, more than 90% by **Duesenfeld GmbH**, and 80-100% by **Li-Cycle®**. Thanks to hydrometallurgy, metal can be recovered from mixed-material alloy and black mass; current collectors, anode, and electrolyte are also successfully recovered in mechanical-hydrometallurgical path, hence explaining for such high recovery efficiency. The output can be of high purity and able to introduce back into battery supply chain. The process consumes much less energy than pyro process, but it also needs to address hydro fluoride gas formation during the pre-treatment step, as well as effluent wastewater that concentrates with chemicals. On the other hand, hydrometallurgical requires rigorous sorting according to correct battery type, or even cathode type, and depends on mechanical separation to produce a high yield output.

Direct Cathode Recycling is a promising innovation. The process bypasses the need to reduce the batteries to elementary building blocks, but instead, directly transforming the cathodes from used batteries so that those cathodes can be used in new battery manufacture. That technically means this method can reach a recycling efficiency of 100%. Direct cathode recycling is shown to offer the best environmental performance in terms of energy consumption and greenhouse gas emission for treating pouch cells of different cathode types (U.S. Energy Storage Association, Ciez & Whitacre 2019) (Appendix 2). Nonetheless, direct cathode recycling inflexibly addresses a specific type of cathode, and there is a chance of the treated cathode being impure with too many concentrations of unwanted metals like aluminum. Table 2 provides an advantage-disadvantage comparison between these three recycling pathways.

TABLE 2. Assessment on li-ion battery recycling methods (Mossali *et al.* 2020, Chen *et al.* 2019, Harper *et al.* 2019)

	Advantages	**Disadvantages**
Pyrometallurgy	o Mature technology readiness, o No need for sorting, able to receive input of mixed cathode type, o Outputs are elemental Co and Ni that can be used in synthesizing new cathode materials of different chemistries.	o CO2 emission, HF gas emission, high energy consumption o The output of mixed metal alloy requires additional treatment to obtain specific elements, o Lithium and several non-cathode materials in Li-ion batteries (e.g. plastics, graphite, aluminum) are not recovered, o Low profit for low- or non- Co cathode.
Hydrometallurgy	o Good technology readiness, o All materials in the cathode can be recovered with high efficiency and high purity, including Co, Ni, and Li, o Low-temperature operation and low CO_2 emissions than pyrometallurgical process.	o Need preliminary sorting and mechanical processing, o Difficulty in efficiently separating similar property elements in the solution (Co & Ni), o Chemically concentrated wastewater discharge.
Direct Cathode Recycling	o Active materials are directly re-usable after recycled, o Significantly lower emissions and less secondary pollution than hydro-and pyro-metallurgical processes, o Leverage recycling of low economic value of LFP and LMO cathode chemistries.	o Strong need for pre-sorting and pre-processing, o Work only with exact cathode chemistry, o Challenging to output consistently high purity and pristine crystal structure, o Inflexible process, the output cathode may be outdated for new battery chemistry in the future.

4.2 Metal, graphite, electrolyte recovery

Cobalt metal is the main economic driver for recycling operations. Commercial operators aim to recover cobalt from the battery waste, and the cobalt output exists in varying compounds, including cobalt chlorine (**Umicore**, **Duesenfeld**), cobalt oxide (**Sony-Sumitomo)**, cobalt hydroxide (**Recupyl**), cobalt sulfate (**Duesenfeld**). Nickel and manganese are also critical extraction targets for recyclers; similar to cobalt, nickel and manganese are retrieved in a variety of salt, oxide, or hydroxide forms.

Some commercial operators proceed with the recovered cobalt compounds with additional refinery processes, such as **Umicore**, then using them in cathode precursor synthesis and later in new battery production. Some operators do not conduct a closed-loop material cycle but sell the recovered metal compound to the metal industry. In the future, operators are aiming that not only cobalt, nickel, and manganese are recovered but also refined them into active cathode powder, such as the LCO or NMC precursor, then re-introduce it to the battery supply chain.

In the case of lithium recovery, conducting a review on commercial and piloting recycling operators, Valio (2017) reported that lithium is often recovered from battery waste as lithium carbonate or lithium phosphate. The use for recovered lithium compound is either back to the battery supply chain, or to other industries such as glass manufacturing.

Battery-grade lithium compounds are extremely demanding, they must be 99.5% pure for portable li-ion battery, but 99.9% or even 99.999% for batteries purposed in electric vehicles and industrial use. Therefore, it is likely that recovered lithium compounds have to receive an additional purification process with additional virgin lithium, before being re-introduced back into the battery material chain. Alternatively, the recovered lithium compound can still provide value for industrial development as materials for glass or metal industry.

The recovery of cobalt and nickel is very efficient, from 94% to almost 99-100%, in either pyro-hydro-metallurgical or mechanical-hydrometallurgical process. Lithium recovery has high efficiency when treated by hydrometallurgy after mechanical separation, but the rate drops significantly if treated after pyrometallurgy and in mixed feed (Appendix 1.1). Meanwhile, Hans (2019) reported that laboratory experiments have been predominantly producing a cobalt and lithium recovery performance of 99 to 100% efficiency using state-of-the-art hydrometallurgical process.

With regards to the casing and current collectors, whose elemental metals are steel, copper, and aluminum, pyrometallurgical smelting consumes most aluminum as burning fuel while renders small amount to the slag, and reduces steel and copper to the metal alloy. On the other hand, those metals can be separated during the mechanical process. Steel is sorted out by magnet; aluminum and copper are separated by density separation techniques. Some traces of aluminum and copper exist in the fine powder, which are precipitated for separation because they are considered impurity in black mass. Recovered steel, copper, and aluminum can be introduced back into the battery supply chain, or for other industrial purposes.

Anode and electrolyte have also become interests for recovery, to address the requirements in the Battery Directive for recycling at least 50% of the battery weight. Froth flotation, dense liquid gravity separation, and porous filter are few recorded techniques for graphite separation. Additionally, the electrolyte may be recovered by condensation after being evaporated at the thermal-deactivation step. In fact, **Duesenfeld** states in its patent that recovered graphite are feasible to be used in lithium battery electrode manufacturing, as well as recovering most types of organic solvent in the electrolyte. Nonetheless, Gaines (2018) indicated there are valid concerns whether the anode and electrolyte can be incorporated back into the battery supply chain, due to their degraded performance and low market cost.

4.3 Recycling different cathode chemistries

For high cobalt type batteries such as portable LCO or NMC 1/1/1 (nickel/manganese/cobalt ratio is 1:1:1), both pyrometallurgy and hydrometallurgy offer effective recovery of that metal. However, with the trend that NMC 5/3/2, NMC 6/2/2, NMC 8/1/1 and NCA are lowering in cobalt content while increasing in market applications, recovery focusses may shift to nickel, manganese, and lithium, with the inclusion of more components such as anode, electrolyte, steel, copper and aluminum in the recovery scheme. Therefore, in response to the trend of lithium-ion battery development, pyrometallurgical smelting may become obsolete for li-ion recycling, and most recycling facilities will use mechanical and hydrometallurgy while using pyrometallurgical technique only for removal of binder and electrolyte.

The situation for LFP recycling is problematic. Constituent metals in LFP battery, which are lithium and iron, are intrinsically low in economic benefit due to cheaper mined counterparts, and the natural resources for lithium and iron are still abundant so there is no shortage in decades to come. Even in pyrometallurgical smelting, the removal of LFP batteries seems to yield higher value alloy (Gaines 2018). The most energy-consuming step in the battery lifecycle is in the cathode synthesis, a process mandatory both to regardless the metal is mined or recycled (Valio 2017). In general, there are little incentives for metal recovery of LFP in terms of profit and environment.

TABLE 3. Recycling cost estimates based on cathode type (EPRI 2017). Non-cobalt LMO and LFP/Graphite are more costly due to elements' low selling value.

Chemistry	Estimated Recycling Cost Price per Kilogram
NCA	$2.20
NMC	$2.20
LMO	$5.50
LFP/Graphite	$5.50
LFP/LTO	$2.20

Global efforts have been made for adapting to the situation of recycling LFP. In China where LFP is the most common li-ion type, **Gangfeng Lithium**'s has recycling plant focuses entirely on lithium recovery, so despite low lithium concentration in LFP, the large quantity and specialized operation may enable a more profitable solution.

In the European context, the French Alternative Energies and Atomic Energy Commission **CEA** patents a mechanical-hydrometallurgical process specifically designed for LFP battery with both graphite and LTO anode (Laucornet, Barthelemy, & Diaferia 2017). The recovered compounds include lithium and $FePO_4$ solution can be used in synthesis of new electrodes. Additionally, graphite, titanate oxide, electrolyte, and binder are recoverable during the process. Also implementing mechanics and hydrometallurgy, **Umicore** patents a process for selective recovery of lithium salt and iron phosphate from LFP (Schurmans & Thijs 2012). From the process, lithium is recovered with high efficiency, low iron impurity; meanwhile, recovered $FePO_4$ can be used in agriculture or gardening, or even introduce back into the battery material chain. Then, although not specifically addressing LFP, **Akkuser Oy** is developing a mechanical process for the treatment of low- to non- cobalt content; the output is likely to be further treated by the company's partners for value recovery.

Finally. the most economically and environmentally attractive effort for LFP is Direct Cathode Recycling (Harper *et al.* 2019, Chen *et al.* 2019, Gaines 2018). Because new LFP cathodes can be synthesized from old ones, the problem of low economic value for recovered components and associated cost when synthesizing material into cathode will be avoided.

5 CASE STUDY: PRELIMINARY WASTE MANAGEMENT PLAN FOR LITHIUM-ION BATTERIES USED IN ENERVERA PROJECT

The ENERVARA project is a collaboration work between Tampere University of Applied Sciences, with TAKK Vocational School, and Tampere small-and-medium enterprises, while being funded by the Pirkanmaa Association. This research and development project aims to leverage the application of lithium-ion batteries as mobile large-scale energy-storage in the electric vehicle charging stations and renewable energy integrated systems.

This section was written as a separate document and delivered to ENERVARA's project manager and included in this Thesis as a case study. It was to provide preliminary considerations for the future management of the Li-ion batteries used in the project, when the project is decommissioned, or when the batteries eventually reach end-of-use. Its intention is to help the project manager and management board take appropriate thinking and actions to responsibly manage their retired Li-ion batteries.

5.1. Planning for decommissioning

New batteries are expected to serve in the large-scale application for 10 to 20 years (Electric Power Research Institute 2017). If the batteries are in the second-use phase from first-use electric vehicles, lifespan is expected to be 7 years. If there is a Battery Management System (BMS) in ENERVARA's system, the lifespan can be more accurately predicted with computer modeling. ENERVARA project may possibly end earlier than the batteries' lifespan, which renders the batteries' immediate end-of-life stage.

There are legal obligations put on the battery supplier of the project, namely Waste Framework Directive 2008/98/EC, Battery Directive 2006/66/EC, and the Finnish equivalent of Waste Act 646/2011 and Government Decree on batteries and accumulators 3.7.2014/520. Therefore, the project management board and battery supplier should communicate, scheme, and finance for the waste management plan for 108 cells used in the project.

Apart from necessary paperwork, it is advisable for ENERVARA's management board to carefully assess risk issues, safety plans, disturbance to the surrounding environment, or even consider writing a Project Decommissioning Report.

5.2. Dismantle and Testing

When the project decommissions, workforce needs to dismantle the battery pack from connecting cable and electrical system, then discharges battery modules to a safe state of charge (SOC), recommended at 30% full capacity, and around 3.10 V according to the voltage discharge curve in the specification document. Then, electro-impedance testing on each battery cell should be done to obtain capacity at the time of end-use, its relation to the original specification's capacity, and therefore knowing the state of health (SOH). Data from BMS are analyzed to graph the aging performance of each battery cell or module and predict if the batteries are still capacity functional or close to the point of accelerated aging, also known as the technical term "aging knee".

5.3. Packaging and Transportation

The inactive battery cells must have the electrodes covered with insulating tape or terminal covers to avoid short circuits or accidental connections. In terms of packaging, it is convenient that the batteries are designed as a mobile large-scale energy storage, so the metal casing surrounding battery modules may sufficiently be safe.

The management board may take advantage of the mobility trailer to deliver batteries to the recycling site. However, a long-distance journey may render the decision for a transportation service; in that case, an outer packaging encasing metal boxes and cushioning material may be required.

Packaging for batteries intended for recycling are advised to follow ADR regulations when transporting by road to the waste management destination, which includes logos of (i) Class 9A, (ii) UN3480, and (iii) "batteries for recycling" in capital text. (ADR Special Provision 377, Packaging Instruction P909.)

5.4. Reuse

It is wasteful if functional end-of-use batteries have to face recycling treatment for metal recovery. After diagnostic tests for the SOC and aging profile of ENERVARA's batteries, the management board should let those batteries be used in future R&D energy storage project with SMEs. Or, the board can collaborate with an actor within the BatCircle network for a used-battery-refurbishment R&D project to restore the batteries' SOH to original specifications, because this idea is a highly interesting area for global battery research. These initiatives will strengthen reuse and second-life applications for Li-ion batteries, as well as strengthening collaboration between universities, research centers, and small-medium-big companies in Finland.

5.5. Recycle

Recycling in the commercial pathway

If the batteries are too deteriorated in SOH or face the accelerated aging problem, the project management board can consider a Finnish commercial recycling service as a waste disposal strategy. A potential operator Fortum promises a highly efficient of more than 80% recovery rate, battery-grade quality output, low energy requirement, chemical-based metallurgical process. However, the company seems to interest in the recovery of Cobalt and Nickel because of their high selling value (Fortum n.d.). ENERVARA's batteries, on the other hand, are none in Cobalt or Nickel. Therefore, the management board can consider choosing operator AkkuSer, which is currently piloting mechanical processes for low Cobalt value batteries (Akkuser n.d., Business Finland Batteries 2019).

The management board is encouraged to communicate with the recycling operator about the Watt-hour rating, mass, SOH, SOC, and cathode chemistry; this information greatly facilitates disassembly, sorting, and classification stages. In addition, the board should request the operator about clarification on the recycling process and a comprehensive report on recovery steps and efficiency. For example, if AkkuSer Oy is chosen for service, what will happen when it delegates the black mass, which is the output from its mechanical process on batteries, to its metallurgical partner, what are the main recovery products, and whether they can be implemented as secondary materials for battery supply chain or other industrial uses.

Recycling in R&D pathway

Alternatively, should the management board want to contribute to research and development, the batteries can be transferred to a recycling research group, possibly a university department or an institute in the BatCircle network. The batteries will enable the research group to experiment with an efficient, and hopefully high battery-grade purity, metal recovery process. Moreover, because ENERVARA's battery type has low economical value for elemental metals, it is encouraged that the research group will innovate a direct recycling technique to recycle and synthesize cathodes from spent-batteries into new-cathode, ready to introduce into the battery supply chain. If such technology is successful, it will be a technological breakthrough for the Finnish battery supply chain.

6 CONCLUSION

Battery disposers, who are this thesis' target audience, after reading this document will have adequate understanding to execute a waste management strategy for their retired large-scale energy storage. They will know how to plan for a safe, minimized risk decommission, to package battery waste according to guidelines. Then after diagnostic tests, if batteries are still serviceable, the disposers are recommended give the batteries a second-life opportunity. Perhaps, that can be an R&D energy storage project which the intended lifetime is under five years.

When the batteries are determined for recycling, the choice for an operator is determined by its offered service. The service includes package and transport, high recovery efficiency by using a combination of mechanical separation and hydrometallurgy, and possibly pyrometallurgical techniques, report, reasonable fees which table 3 may be a good baseline. In addition, the possibility of recovered materials to return to the battery material cycle is a good selling point.

Meanwhile, the disposers should communicate with recyclers the specifications of the batteries, including watt-hour rating, mass, and volume, so that recyclers can estimate if they can handle such large bulk. For an estimate, 7,000 ton/year Umicore and 4,500 ton/year Retrieve recycling plant could accept treatment of 1GWh large-scale energy storage (EPRI 2017). The batteries' type, state of health, and state of charge are some specifications that enable recyclers to conduct easier recycling processes and better safety management. Alternatively, the batteries can be supplied to an R&D research group, perhaps to facilitate researches towards the direct recycling pathway for lithium-ion batteries.

For the thesis' secondary audience who are battery industrial and academic actors, these followings are recommended for capacity building competent recycling in the battery supply chain. Firstly, technological improvements will have to be made for more efficient recycling; that includes automated disassembling, mechanical separation, metallurgical techniques especially hydrometallurgy, and direct component recycling. Secondly, the second life opportunity should be enabled with more technical possibilities and price competitiveness. Therefore, lithium-ion batteries will maximize potential and facilitate sustainable development goals. Finally, not only technology for reusing or recycling but also the collection and logistics scheme for batteries must be addressed. By increasing collaborations and networks, a viable, profitable Circular Economy model that encompasses holistic operations for waste management will be realized. The battery supply chain will greatly be secured when the recovered components can be incorporated back into the material cycle.

REFERENCES

ADR Book. 2017. 3480. Adrbook.com Read on 08.07. 2020. Available at: https://adrbook.com/en/2017/TabloA/3480

Akkuser. N.d. Recycling of low-grade cobalt Li-ion batteries. Access on 02.09.2020. https://www.akkuser.fi/en/process-descriptions/low-grade-cobalt-li-ion-battery/

Andolfsson-Tallqvist, J., Ek, S., Forstén, E., Heino, M., Holm, E., Jonsson, H., Lankiniemi, S., Pitkämäki, A., Pokela, P., Riikonen, J., Rinkkala, M., Ropponen, T., Roschier, S. 2019. Batteries from Finland Final Report. Business Finland. Read on 04.2020. https://www.businessfinland.fi/49cbd0/globalassets/finnish-customers/02-build-your-network/bioeconomy--cleantech/batteries-from-finland/batteries-from-finland-report_final_62019.pdf

Azevedo, M., Campagnol, N., Hagenbruch, T., Hoffman, K., Lala, A., Ramsbottom, O. 2018. Lithium and cobalt – a tale of two commodities. Metals and Mining. https://www.mckinsey.com/industries/metals-and-mining/our-insights/lithium-and-cobalt-a-tale-of-two-commodities#

BATCircle. Finland based circular ecosystem of battery metals consortium. Batcircle.fi

Battery University. 2019. BU-301a: Types of Battery Cells. Updated on 24.04.2019. Read on 08.07.2020. https://batteryuniversity.com/learn/article/types_of_battery_cells

Chen, M., Ma, X., Chen, B., Arsenault, R., Karlson, P., Simon, N., Wang, Y. 2019. Recycling End-of-Life Electric Vehicle Lithium-Ion Batteries. Joule. DOI: 10.1016/j.joule.2019.09.014

Ciez, R. E., Whitacre, J. F. 2019. Examining different recycling processes for lithium-ion batteries. Nature Sustainability, 2, 148-156.DOI: 10.1038/s41893-019-0222-5

Directive 2006/66/EC on batteries and accumulators and waste batteries and accumulators. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:02006L0066-20131230&rid=1

Directive 2008/98/EC on waste. https://ec.europa.eu/environment/waste/framework/

EESE/EERA. 2016. European Energy Storage Technology Development Roadmap Towards 2030.

Electric Power Research Institute. 2016. Energy Storage Integration Council (ESIC) Energy Storage Commissioning Guide 2016. Access on 02.09.2020. https://www.epri.com/research/products/3002009250

Electric Power Research Institute. 2017. Recycling and Disposal of Battery-Based Grid Energy Storage Systems: A Preliminary Investigation. Access on 02.09.2020. https://www.epri.com/research/products/000000003002006911

Energy Storage Association. 2020. End-of-life Management of Lithium-ion Energy Storage Systems. Access on 08.08.2020. https://energystorage.org/thought-leadership/end-of-life-management-of-lithium-ion-energy-storage-systems/

Fortum. N.d. Lithium-ion Battery Recycling Solution. Access on 02.09. 2020. https://www.fortum.com/products-and-services/fortum-battery-solutions/recycling/lithium-ion-battery-recycling-solution

Gaines, L. 2018, Lithium-ion battery recycling processes: Research towards a sustainable course. Sustainable Materials and Technologies. Vo 17. DOI: 10.1016/j.susmat.2018.e00068

Decree on batteries and accumulators 502/2014. Valtioneuvoston asetus paristoista ja akuista.
https://www.finlex.fi/fi/laki/ajantasa/2014/20140520

Hanisch, C., Elwer T., Brückner, L. 2018. Method for the utilization of lithium batteries. European Patent Office EP351764B1

Harper, G., Sommerville, R., Kendrick, E., Driscoll, L., Slater, P., Stolkin, R., Walton, A., Christensen, P., Heidrich, O., Lambert, S., Abbott, A., Ryder, K., Gaines, L., Anderson, P. 2019. Recycling lithium-ion batteries from electric vehicles. Nature 575. 75-86. DOI: 10.1038/s41586-019-1682-5

Hart, P., Kollmeyer, P. Modelling of second-life batteries for use in a CERTS microgrid. In: Proceedings of IEEE power energy conference III, PECI; 2014 p. 1-8. DOI: 10.1109/PECI.2014.6804554

Hesse, H. C. Schimpe, M., Kucevic, D., Jossen, A, 2017. Lithium-Ion Battery Storage for the grid – A Review of Stationary Battery Storage System design tailored for Applications in Modern Power Grids. Energyies. DOI: 10/3390/en10122107

Korpela, A. 2020. Personal email exchange.

Korpela, A. 2018. Suuren kokoluokan energianvarastointiteknologioiden teknistaloudelliset näkymät. Tampere University of Applied Sciences. ISBN 978-952-7266-32-8

Laucornet, R., Barthelemy, S., Diaferia, N. 2017. Method for recycling lithium batteries and/or electrodes of such batteries. European Patent Office EP2754201B1

Lebedeva. N., Persio, F, D., Boon-Brett, L. 2016. Lithium ion battery value chain and related opportunities for Europe. European Commission. https://ec.europa.eu/jrc/sites/jrcsh/files/jrc105010_161214_li-ion_battery_value_chain_jrc105010.pdf

Lithium Battery Service GbR. 2017. Module Recycling and Disposal. Lithium-battery-service.com. Read on 08.07.2020. https://www.lithium-batterie-service.de/templates/emotion_lbs/frontend/_resources/images/lbs/module_disposal/EN-Recycling-and-Disposal-Lithium-Battery-Service-2017.pdf

Melin, H. E. 2019. The lithium-ion battery end-of-life market – A baseline study. Access on 09.09.2020. Available at: http://www3.weforum.org/docs/GBA_EOL_baseline_Circular_Energy_Storage.pdf

Miao, Y., Hynan, P., von Jouanne, A., Yokochi, A. 2019. Current Li-ion Battery Technologies in Electric Vehicles and Opportunities for Advancements. Energies, 12, 1074. DOI: 10.3390/en12061074

Mossali, E., Picone, N., Gentilini, L., Rodriguez, O., Pé rez, J M., Colledani, M. 2020. Lithium-ion batteries towards circular economy: A literature review off opportunities and issues off recycling treatment. Journal of Environmental Management, 264. DOI: 10.1016/j.jenvman.2020.110500

Neubeuer, J., Pesaran, A., Williams, B., Ferry, M. A. Techno-Economic Analysis of PEV Battery Second Use: Repurpose -Battery Selling Price and Commercial and Industrial End-user Value. In: SAE 2012 Word Congress & Exhibition. http:// dx.doi.org/10.4271/2012-01-0349

Romare, M., Dahllöf, L. 2017. The Life Cycle Energy Consumption and Greenhouse Gas Emissions from Lithium-Ion Batteries: A study with focus on Current Technology and Batteries for light-duty vehicles. ILV Swedish Environment Research Institute. C243

Schurmans, M., Thijs, B. Process for the recovery of lithium and iron from LFP batteries. WO2012072619A1.

Statista. 2020. Lithium-ion battery pack costs worldwide between 2011 and 2020 (in U.S. dolalrs per kilowatt hour). Access on 17.09. 2020. https://www.statista.com/statistics/883118/global-lithium-ion-battery-pack-costs/

Valio. J. 2017. Critical review on Lithium ion battery recycling technologies. Master's Thesis. Aalto University. Helsinki, Finland.

Waste Act. 646/2011. https://www.finlex.fi/fi/laki/kaannokset/2011/en20110646_20140528.pdf

Zou, H., Gratz, E., Apelian, D., Wang, Y. 2013. A novel method to recycle mixed cathode materials for lithium ion batteries. Green Chemistry, 15, 1183-1191.

APPENDICES

Appendix 1. Date collected on recycling efficiency.

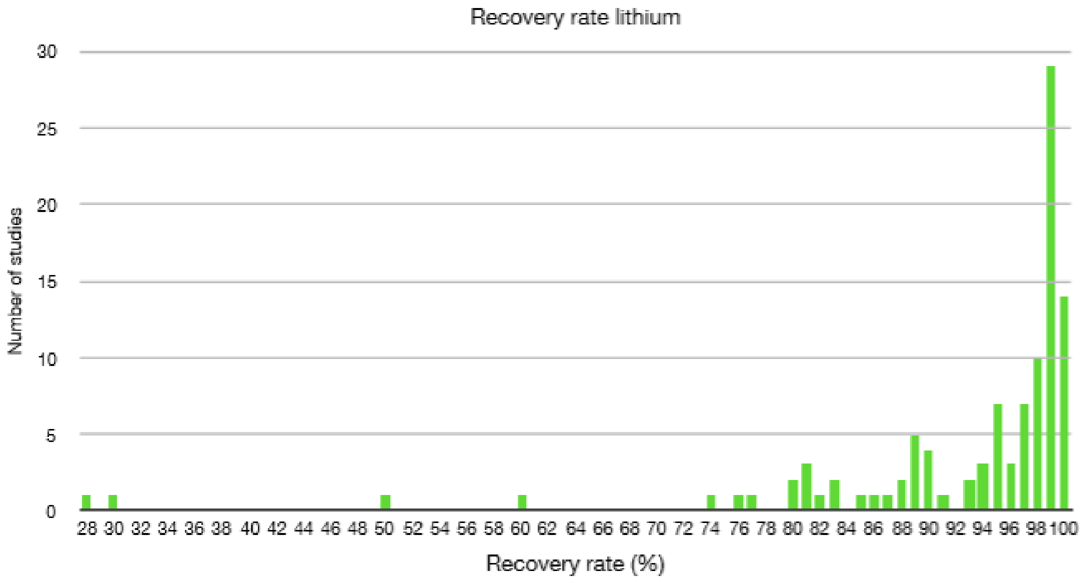
Appendix 1.1. Recycling efficiency based on processes and chemistry (Lebedeva, Persio, Boon-Brett 2016)

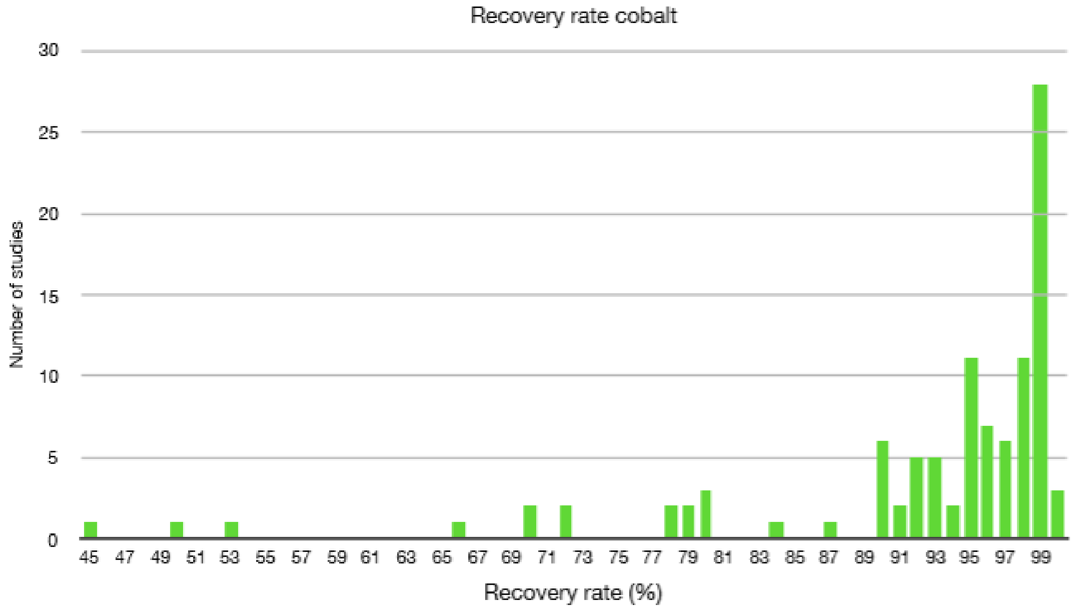
Elemental material	**Recycling Efficiency (%/w battery)** based on recycling processes and input stream of battery type		
	Pyro + Hydro NMC + LFP	Hydro NMC	Hydro LFP
Lithium	57	94	81
Nickel	95	97	-
Manganese	0	~100	-
Cobalt	94	~100	0
Iron	0	-	0
Aluminum	98	98	-
Phosphate	0	-	0
Graphite	0	0	0

Appendix 1.2. Accurec GmbH's recycling plant operation efficiency.
The operation uses a combination of mechanical – pyro – hydro metallurgy (Accurec 2018 https://accurec.de/publications-downloads)

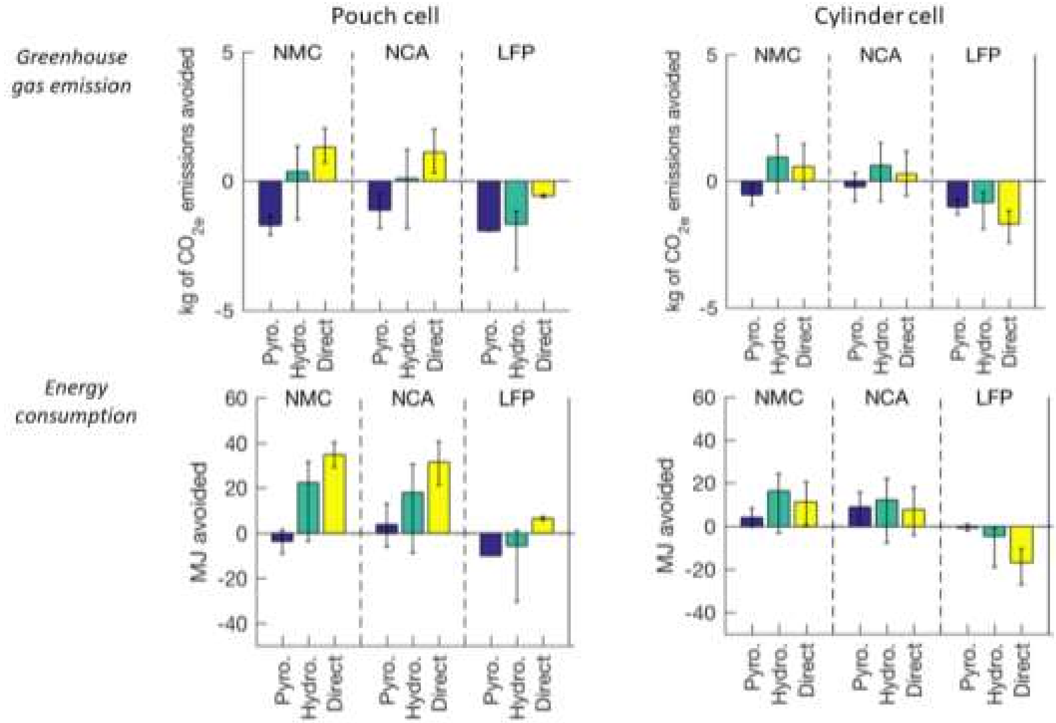
Component	**%w/w input**	**%w/w output**	**Component Recycling Efficiency (%)**
Steel casing	23.6	20.3	86.0
Copper foils	15.2	14.3	94.0
Aluminum foils	4.3	0.3	69.7
Cobalt	18.5	11.36	95.9
Nickel	12.1	6.5	97.0
Graphite	6.7	12.2	78.7
Plastics, electrolyte solvents	15.5	-	-
Others	4.1	-	
Recycling Efficiency		65.2% (Claimed)	59.3% (Audit)

Appendix 1.3. Melin 2019’s review synthesizes recovery rate from researches





Appendix 2. Environmental savings per kilogram battery of different recycling pathways



The above figures compare environmental performance for three recycling operations (pyro-metallurgical, hydro-metallurgical, and direct cathode recycling). The evaluated parameters are amount of greenhouse gas emission and energy consumption may be saved, when one kilogram of battery is manufactured with recycled inputs instead of mining ones. are compared on the y-axis. Negative value means the recycling process adds more footprint to the life cycle. Analysis also shows difference among different three cathode chemistries (NMC, NCA, and LFP) and two cell configurations (pouch and cylindrical). The bar chart delivers statistical median and 95% confidence interval of the data.

From the results, direct recycling is shown to offer environmental benefits compared with other recycling pathways in pouch cell. However, for cylindrical cell, hydrometallurgy has better environmental performance.

Calculation was based on the U.S. average data for electrical grid. Data and figures were obtained from the publication by Ciez & Whitacare 2019