UNIVERSITY
OF OULU

FACULTY OF TECHNOLOGY

HYDROGEN-ASSISTED RECYCLING OF LITHIUM-ION BATTERIES

Pekka Oinas-Panuma

DEGREE PROGRAMME IN PROCESS AND ENVIRONMENTAL ENGINEERING
Bachelor’s thesis
April 2024

TIIVISTELMÄ

Hydrogen-assisted recycling of lithium-ion batteries

Pekka Oinas-Panuma

Oulun yliopisto, Prosessi- ja ympäristötekniikan tutkinto-ohjelma

Kandidaatin tutkielma 2024, 24 s

Ohjaajat yliopistolla: Aidin Heidari, Susanna Airaksinen, Eetu-Pekka Heikkinen

Litiumakut ovat yhä kasvavampi ja merkittävämpi osa nykypäivää. Syinä tähän on niiden loistava energian varastointikyky niiden suhteellisen matalaan painoon nähden. Yhteiskunta on siirtymässä yhä enemmän ja enemmän sähköistymiseen niin liikenteessä, kuin jokapäiväisessä elämässäkin. Tämän takia litiumakkujen arvokkaiden maametallien, kuten litium, mangaani ja koboltti, kysyntä kasvaa. Litiumakkujen kierrätys nykyään on hyvin ympäristölle haitallista prosessien vaatiman energian, haitallisten kemikaalien ja korkeiden päästöjen takia. Siksi on siirrytty tutkimaan vetypelkistystä näiden kierrätysmetodien korvaajaksi.

Tämän kirjallisuusselvityksen tavoitteena on selittää litiumakun rakenne sekä toimintaperiaate, yleisimmät nykyään käytössä olevat litiumakkujen kierrätysmetodit. Lisäksi selvittää kuinka vetyavusteinen litiumakkujen kierrätys toimii, ja millä tavalla se eroaa nykyisistä kierrätysmetodeista.

Tutkimuksessa selvisi, että vetyavusteinen kierrätys on huomattavasti päästöystävällisempi kierrätysmetodi nykyisiin kierrätysmetodeihin verrattuna, ja vetyavusteisen akkukierrätyksen tulevaisuus teollisuudessa on kirkas.

Asiasanat: Litiumioniakut, kierrätys, vety, pelkistys

ABSTRACT

Hydrogen-assisted recycling of lithium-ion batteries

Pekka Oinas-Panuma

University of Oulu, Degree Programme of Process and Environmental engineering

Bachelor’s thesis 2024, 24 pp.

Supervisors at the university: Aidin Heidari, Susanna Airaksinen, Eetu-Pekka Heikkinen

Lithium-ion batteries (LIBs) are a central part of modern society, since they have a high energy storage capacity in comparison to their weight and therefore, the demand for more and more LIBs is ever higher. The more batteries are produced, the more battery materials are needed for production. Also, more battery waste is produced. The production of LIBs requires valuable earth metals such as lithium, cobalt, nickel, and manganese. These metals can be obtained from discarded and spent lithium-ion batteries. Recycling of LIBs environmental challenges like high energy consumption, the harmful chemicals required, and high GHG emissions. Therefore hydrogen-assisted recycling is being researched as a new method to reduce the mentioned environmental impacts.

The goal of this literacy research is to explain the basic structure and function of lithium-ion batteries, the currently used methods of recycling and to research how hydrogen can be used in recycling LIBs.

The research found that hydrogen-assisted recycling is noticeably more environmentally friendly than the current methods and the future for hydrogen in the industry is bright.

Keywords: Lithium-ion batteries, Battery Recycling, Hydrogen-reduction

TABLE OF CONTENTS

1 INTRODUCTION

The global demand for lithium-ion batteries (LIBs) is surging ever higher. This is mainly due to the electrification of the transport industry, for example electronic vehicles (EVs). This raises ever higher need for battery recycling and more efficient methods for recycling of LIBs and the materials in them. (Yu et al., 2024)

Lithium-ion batteries have become the industry standard for energy storage due to their high energy storage capacity, low weight, low self-discharge, extended operational lifespan and comparably low environmental footprint (Zhao et al., 2024). No wonder then why, as we are moving away from fossil fuels, we, as a society, are attracted towards LIBs as energy sources in the most of our day-to-day applications such as EVs, mobile devices, portable speakers, wireless headphones, etc.

Currently the surge has caused concern with the gigantic increase in demand of battery materials such as graphite, in comparison to their limited availability in the world (Milian et al., 2024). By recycling both spent LIBs and byproduct LIBs, which are found to have manufacturing defects etc. and are discarded in production (Yu et al., 2024), we are able to reduce the demand of raw materials, which include valuable metals such as lithium, cobalt nickel and manganese used in battery-manufacturing. Also from a environmental point-of-view, recycling of batteries is important, since mining and refining new materials, which are limited in themselves, causes unnecessary strain on the environment. The topic is important for just these reasons. And with this rapid electrification of society, a rapid increase in recycling and recycling efficiency is needed.

In this thesis, the basic structure and function of lithium-ion batteries and explaining the different methods of recycling them is explained. These methods include both hydro- and pyrometallurgical methods in addition to physical methods. The goal of this research is to find out the advantages and disadvantages of using hydrogen for different recycling methods and to reflect the possibilities for the future of LIB recycling using hydrogen.

2 LITHIUM-ION BATTERIES

2.1 Structure of LIBs

Lithium-ion batteries are quite simple devices in principle. As it can be seen in Figure 1, LIBs conventionally consist of a cathode, anode, gasket, separator, gas release valve, and sealing plate. (Joo et al., 2015)

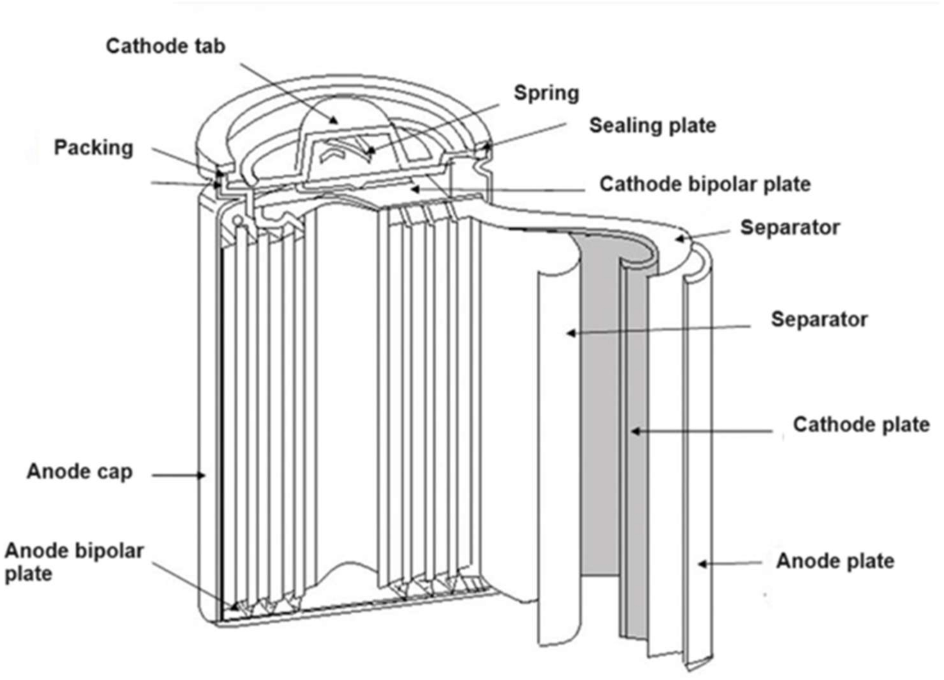

Figure 1. Structure of layered lithium-ion battery (Modified, Joo et al., 2015).

The cathode is the positive electrode of the battery. Cathode materials in layered cathodes are most often composed of different lithium compositions, such as $LiCoO_2$, but other metals such as nickel and manganese can be used instead of cobalt. Cobalt is predominantly used in lithium cathodes due to the ease of producing high-quality cathodes with it. The anode, the negative electrode of the battery, is made of layered graphite. (Joo et al., 2015)

Separator is used to prevent the anode and cathode short-circuiting themselves. The barrier, however, should be porous enough to allow for free flow electrolyte from the cathode to the anode and vice versa. Separators are most often made of polytetrafluoroethylene (PTFE) or other inert polymeric membranes with known and defined porosities and ionic conductivities. The electrolyte is most often a propylene carbonate and a mix of other solvents. (Joo et al., 2015)

2.2 Function of LIBs

Lithium-ion batteries, like all other batteries, function on the same electrochemical fundamentals, excluding a few distinctions. In most cases, there is no liquid lithium in a LIB, instead the lithium is intercalated into both electrode materials (Weicker, 2014). These lithium ions migrate between the anode and cathode, and it causes the charging and discharging of the battery cell (Joo et al., 2015).

During the charging lithium ions migrate towards the cathode and intercalate between the layers of the compound material. Vice versa, during the discharge, lithium ions migrate and intercalate between the layers of the carbon material in the anode. (Joo et al., 2015)

As seen from Reactions 1 and 2, the initial discharging reaction (Reaction 1) shows us the basic chemistry in the battery cell, lithium migrates from the cathode material, in this example, lithium cobalt oxide and binds with the carbon. The subsequent discharge reactions follow Reaction 2. This is also visible on Figure 2.

$$LiCoO_2 + C \rightarrow Li_{1-x}CoO_2 + Li_xC \tag{1}$$

$$Li_{1-x}CoO_2 + Li_xC \rightarrow Li_{1-x+d}\ CoO_2 + Li_{x-dx}C \tag{2}$$

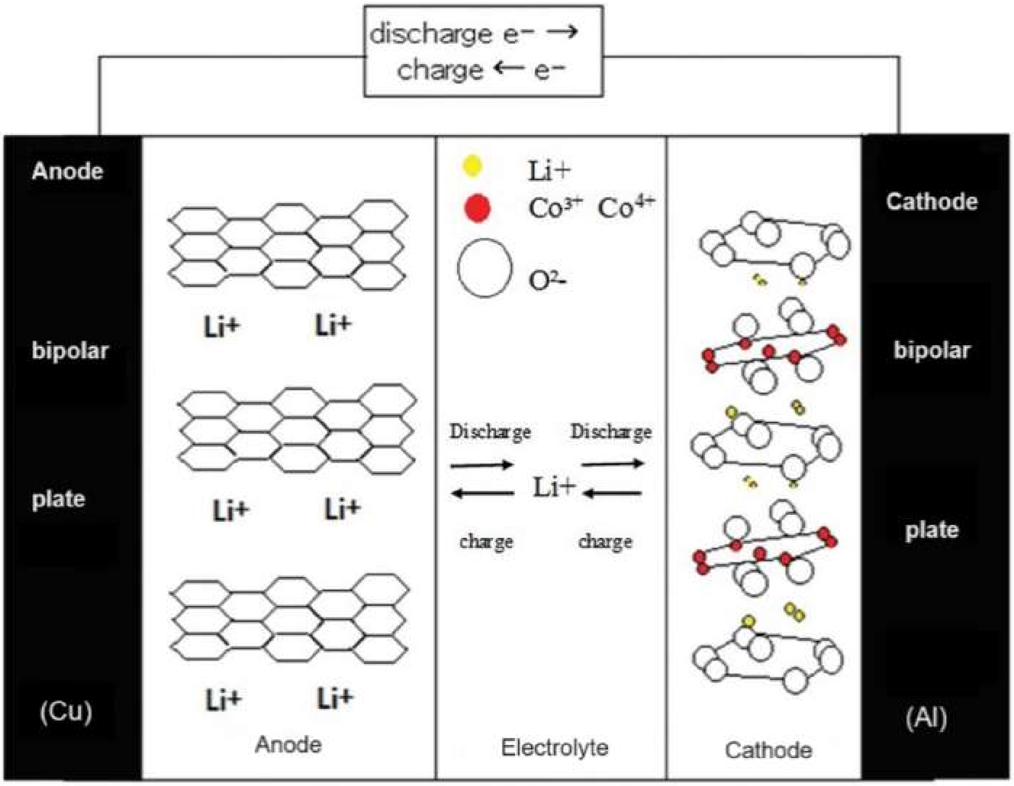

Figure 2. principle of operation of the LIB (modified, Joo et al., 2015).

3 DIFFERENT METHODS OF RECYCLING LI-ION BATTERIES

3.1 Pyrometallurgical methods

Recycling lithium-ion batteries via pyrometallurgical methods involves high temperature smelting at temperatures of 1000-1700 °C and carboreduction in temperatures of 700-900 °C (Velpoor et al., 2023). Smelting with these high temperatures, metals such as nickel, iron, cobalt, and copper can be reclaimed from alloys. However, the process has a major downside as the smelting process loses most of the lithium, and manganese and other lighter metals that form a slag that needs to be treated further to be able to be claimed. (Velpoor et al., 2023) Also, the high temperature environment raises the cost of recycling (Huang et al., 2022b).

In recent years, reduction roasting has been explored as an option of processing of the batteries. Reduction roasting has offered benefits with higher yields and possibility for recovery of selected metals. Reduction roasting has been explored with multiple solid reductants for example with carbon (carboreduction), or aluminium (thermite reduction). (Huang et al., 2022b)

However, reduction roasting has its flaws. The product is often highly impure, and different cleaning processes are needed to remove the impurities and trace amounts of reductants. For example, carbothermic reaction needs an additional calcination step, to remove the residual carbon from the product. Thermite reduction needs multiple steps of leaching with both alkaline and acids to leach out the aluminium. (Huang et al., 2022b)

In conclusion, pyrometallurgical methods of recycling include smelting and reduction roasting. These processes require high temperatures and lots of energy. Pyrometallurgy has trouble in reclaiming lighter metals and requires further treatment to purify the product.

3.2 Hydrometallurgical methods

Hydrometallurgy conventionally includes leaching using inorganic and organic acids and alkali solutions. Also, more environmentally friendly methods, using deep eutectic chemicals such as urea, diketone – alkyl phosphate oxide-based eutectic solvents, ethylene glycol-based solvents and inorganic solvents like ferrous sulphate are being researched and developed. (Milian et al., 2024)

Methods of leaching are categorized by the nature of solvent used. In these methods, the disassembled batteries are through solvents dissolved and then precipitated into an insoluble material (Milian et al., 2024). Most common hydrometallurgical method of recycling battery scraps via leaching is using an inorganic acid with hydrogen peroxide as an assistant and sodium carbonate, ammonium salts and sodium hydroxide as an additional material for the separation and precipitation (Yu et al., 2024). The precipitation is done using salts, such as $KMnO_4$, Na_3PO_4, NaOH, H_3PO_4, or through electrochemical precipitation (Milian et al., 2024).

With the previously mentioned precipitation agents' lithium, nickel, manganese, and cobalt can be recovered. However, recovery of pure metals can cause difficulties due to the different ions and the overlap of the desired pH range for precipitation. Therefore, hydrometallurgical methods are often used in addition to pyrometallurgical methods in the modern industrial application of the recycling process and thus purely hydrometallurgical recycling is rare. (Milian et al., 2024)

Emerging methods of leaching are mostly still in the development. One method developed for recovering lithium and cobalt consists of two step reduction process using sulphuric acid with methanol being used as an eco-friendly reducing agent (Kong et al., 2023). Other methods include microwave assisted leaching and ultrasound assisted leaching. In microwave assisted leaching, a thermal pre-treatment via microwaves increases the surface area of the battery materials and separates the solid black mass from the possible carbon residues, electrolyte solvents and binders and therefore, increasing cathode leaching. (Milian et al., 2024)

Normally heating increases the valence of the valuable transition metals and causes stronger bonds between the metal and oxide ions. Microwave radiation causes the crystal structure of the batteries to disorganize and eases the otherwise difficult leaching process. Microwave irradiation is considered to be a green process of recycling, mainly due to the reduced amount of work and material required.(Milian et al., 2024)

Ultrasound assisted leaching is also a method of enhancing leaching. Ultrasound causes the breakdown of the insolvable binder material and facilitates the release of active electrode materials from the foil. The ultrasonic effect helps dispersing solids and enhances the solid-liquid material exchange. When used with certain organic acids or bio-organisms, ultrasounds decrease the leaching time and enhance metal extraction. Ultrasound also increases leach recovery, making pyrometallurgical pretreatment and the use of reducing agents unnecessary. Thus, saving time and energy. (Milian et al., 2024)

To summarize, hydrometallurgical methods are most often used with pyrometallurgical processes as a secondary process with the goal of purifying the product. The lithium recovered with hydrometallurgical methods is environmentally advantageous due to it being recovered as hydroxides. Currently, the main issues in the hydrometallurgical and pyrometallurgical recycling processes is the high amount saline toxic waste of energy consumed. Though the chemicals used in the recycling process are also hazardous to the environment, both more eco-friendly processes and alternative substances are being researched. (Milian et al., 2024)

3.3 Other methods

Direct recycling methods often tackle the issue that older batteries experience lithium degradation. This causes the batteries performance to suffer. The most common methods of directly recycling battery materials are electrochemical lithiation, eutectic molten salt application, hydrothermal methods, solid state sintering and ionothermal synthesis (Milian et al., 2024). These methods do differ in reaction parameters and in chemicals used, but product is the same re-lithiated cathode materials.

In electrochemical lithiation, the spent cathode is restored in an electrode station at room temperature. A lithiated pyrene reagent is used as an electron and lithium donor (Wu et al., 2023). This causes the degraded cathode to be flooded with new lithium and electrons and therefore, restoring its energy storage capacity. (Milian et al., 2024)

Eutectic molten salt application is a way of relithiation by mixing eutectic salts such as lithium hydroxide or lithium nitrate with the cathode material and aluminium impurities as a doping agent and heating the solution to the melting point of the salt. After the reaction between the cathode material, the solution is thermally annealed at temperatures of 300-900 °C to recover the cathodes structure. Hydrothermal methods are based on the dissolution and recrystallization in reactors by controlling, for example, temperature and pressure. (Milian et al., 2024)

The final two methods in literature are hydrothermal methods and solid-state sintering. Ionothermal synthesis is conducted at lower temperatures and in the process, lithium salts are mixed with ionic liquids and with lithium halide are used as a precursor. The process is firstly heated to approximately 150-250 °C and finally heated to 500 °C. Solid-state sintering simply mixes used cathodes with salts and bonds them together with heat treatment. (Milian et al., 2024)

Advantages to directly recycling lithium-ion batteries are the decreased need for chemicals and energy that currently used hydrometallurgical and pyrometallurgical methods require. However, direct recycling has the downside of needing manual peeling and separation of materials which is currently hard to automate. (Milian et al., 2024) If the pretreatment can be automated, direct recycling is possibly a better method of recycling than the more common hydrometallurgical and pyrometallurgical methods mentioned previously.

4 HYDROGEN-ASSISTED RECYCLING OF LI-ION BATTERIES

4.1 Use of hydrogen in recycling LIBs

One of the currently highly researched industrial processes in multiple different sides of the industry is hydrogen reduction. Lithium ions in the cathode materials can be recovered via hydrogen reduction in much lower temperatures than normal carboreduction, only at temperature of approximately 450 °C (Xie et al., 2023).

Hydrogen as a reducing agent is a valuable research topic in LIB recycling, because it is considered as a green gas since the process forms water as a byproduct instead of carbon dioxide in carboreduction. The advantage of hydrogen use is that fewer process steps are needed for the separation and recovery of materials and less lithium is lost during the recovery. (Huang et al., 2022b)

In regular reduction roasting, lithium falls susceptible to be absorbed into the graphite. When the hydrogen reduction product compromises the lithium oxides, it forms a highly water-soluble lithium hydroxide. Other products formed are water-insoluble manganese oxide and water-soluble magnetic nickel and cobalt that can be removed later in the process. Huang et al., 2022b)

In conclusion, hydrogen is being researched as an environmentally friendly option to currently used reducing agents. Hydrogen has a potential to decrease the environmental impact of lithium-ion battery recycling and decrease the release of greenhouse gases that the reduction processes produce as a byproduct.

4.2 Differences in methods

One proposed recovery process and flow chart by Huang et al. (2022a) can be seen in Figure 3. In the experiment, the powdered cathode material is first dried in an oven at the temperature of 100 °C. After drying, the material is heated using a horizontal tube furnace in an argon-hydrogen atmosphere with temperatures varying between 600-1000 °C and time varying between 15 and 90 minutes. After the reduction, the powder was grounded in a ball mill and then leached into ultrapure water with the help of ultrasound. After the water leaching, the products were filtrated through a quartz tube surrounded by magnets. From the filtration, the lithium compounds were separated from the Co-Ni and Mg alloys. (Huang et al., 2022b)

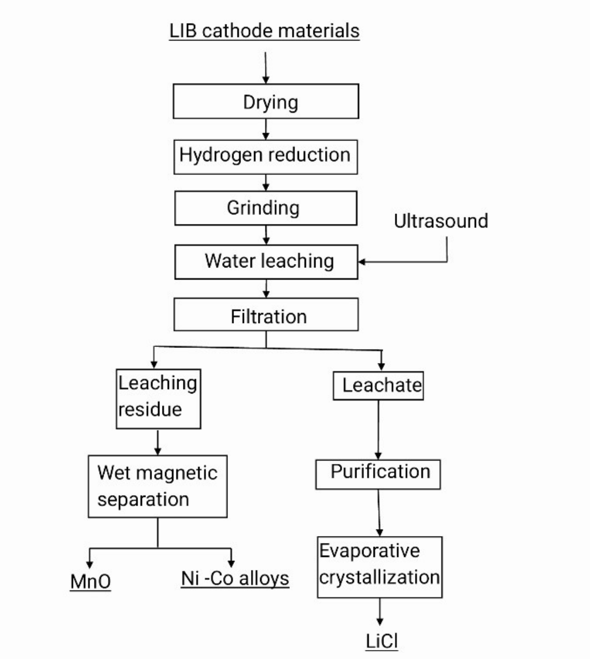

Figure 3. Proposed flow chart of recovery of valuable metals from LIBs. (Huang et al., 2022a)

Rather than the method used in the experiment by Huang et. al. (2022b), Zhou et. al. (2023) used a quartz tube inside of an electric oven. Heating was done in argon gas atmosphere, to ensure inert conditions with the heating rate 5 °C/min until the target temperature was reached. At the target temperature, the atmosphere was replaced by 5%- H_2 95%-Ar atmosphere, where the reduction process was done for a set time. After the roasting, the cooling process was done with a cooling rate of 5 °C/min again in an argon atmosphere to ensure protection of the product.

Second step also differs from Huang et. al. (2022b). Since instead of ultrasound assisted electrolysis, Zhou et. al. (2023) used neutral water electrolysis in two alumina boats as it is shown in Figure 4. In neutral water electrolysis, the anode chamber and cathode chamber are separated by filter papers that allow the materials to pass through.

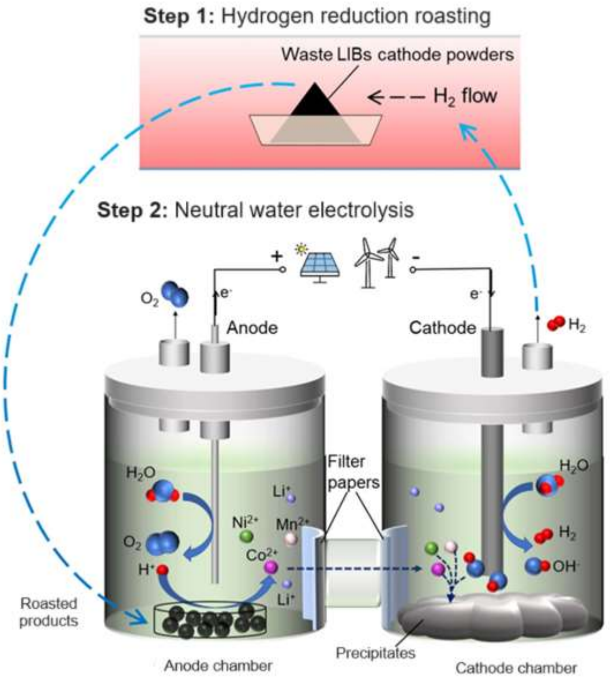

Figure 4. Schematic of the experimental steps of H_2 reduction roasting and neutral water electrolysis. (Modified, Zhou et al., 2023)

Table 1. Different reaction parameters in used hydrogen reduction experiments from literature. Data collected from various sources.

Research	Gas composition	Gas flowrate	Operating temperature	Heating rate	Reaction time
Zhou et al. (2023)	5% H_2-95% Ar	150 mL/ min	375–500 °C	5 °C min^{-1}	50 min
Nuraeni et al. (2023)	15%H_2-85% N_2	300 mL/min	600–1000 °C	N/A	30-180 min
Velpoor et al. (2023)	100% H_2	500 mL/ min	500–900 °C	20 °C min^{-1}	60 min
Huang, et al. (2022a) (1)	Varying H_2-Ar	100 mL/ min	25–1000 °C	20 °C min^{-1}	N/A(3)
Huang et al. (2022a) (2)	Varying H_2-Ar	100 mL/ min	500–900 °C	10 °C min^{-1}	3 h
Xie et al. (2023)	99,999% H	not mentioned	25–800 °C	10 °C min^{-1}	3h

(1) non-isothermal reaction, (2) isothermal reaction with 100 °C increments, (3) not available.

There are lot of experiments in literature using hydrogen reduction with varying parameters as seen in Table 1. Parameters changed are for example gas composition, gas flow rate, reduction process' operating temperature, reaction time, and ovens heating rate, etc. These parameters change the properties of the products, and these differences are analysed in depth.

As mentioned previously, Zhou et al. (2023) used a 5% H_2 - 95% Ar atmosphere. This gas mix was inserted with the flowrate of 150 ml/min and the heating rate of the oven was 5 °C/min until the desired operating temperature of 375-500 °C was reached. The samples were held at the operating temperature range and atmosphere for 50 minutes. Results of the experiment show that hydrogen can be utilized as an effective reducing agent when used in combination with neutral water electrolysis.

Nuraeni et al. (2023) researched the hydrogen reduction of $LiCoO_2$ with the temperature range of 600-1000 °C with the atmosphere of 15% H_2 - 85% N_2. The research showed that metallic cobalt was generated at 400 °C and was stable until 1200 °C. CoO was produced as the decomposition product. Research showed that the reduction of cobalt was strongly dependent on the amount of hydrogen present. Lithium hydroxide was stable until 1035 °C.

The reduction of lithium was more dependent on the temperature of the process than the amount of hydrogen present. The research results showed that the best temperatures for operation are between 800-900 °C. There the cobalt was able to be recovered in larger mass, since the lithium was generally present as a Li_2O. (Nuraeni et al., 2023)

Velpoor et al. (2023) compared regular carbothermal reduction to hydrogen reduction. Hydrogen reduction was done in 100% hydrogen atmosphere, in 500-900 °C temperature range. Hydrogen reduction at 500 °C managed to yield 93 % dissolution and 82.7 % recovery of cobalt. Higher temperatures increased the presence of metallic cobalt but decreased the yield of lithium. This was caused by the lithium salts vaporizing in higher temperatures.

In comparison, the carbothermal reduction was done at 900 °C with 30 % in-situ graphite which resulted in lithium being able to be dissolved with 61 % solution rate and 76.5 % recovery rate of cobalt. These results show that hydrogen reduction produced higher yield rates and lower needed reduction temperatures in comparison to carbothermal reduction. (Velpoor et al., 2023)

Huang, et al. (2022a) did two experiments with hydrogen reduction, both isothermal and non-isothermal reactions. Both experiments' parameters can be read from Table 1. Results of the non-isometric experiment show that the reaction can be split into three stages. Initial decomposition of the cathode material under the reducing atmosphere happened at the 195-475 °C temperature range. Following the initial stage, the decomposition of nickel and cobalt oxides mainly occurred at the 475-685 °C temperature range. Finally, the reduction of $LiMnO_2$ and the cobalt and nickel oxides remaining occurred at the temperature range of 685-880 °C. Kinetics analysis of the reaction showed increasing activation energy with the progression of the reduction process.

The isothermal reduction experiment showed that the lowest oxide presence was at 800 °C and the content of nickel and cobalt oxides gradually decreased with increasing the temperature. The activation energy also showed a decreasing trend with the increase in temperature. (Huang, et al., 2022a)

Xie et al. (2023) researched the reduction thermodynamics and phase transformation of LIBs in hydrogen reduction process. The experiment used 2 g cathode powder in a nickel crucible in a tube furnace and heated it in a hydrogen atmosphere. The experiment was followed with a thermodynamic analysis with XRD as a tool utilized.

The XRD showed minor peaks of CoO and LiOH at 300 °C. This showed a minor reduction in $LiCoO_2$ as it starts to break down in the increasing temperature. At 350 °C the minor peaks of $LiCoO_2$ completely disappear, and small peaks of Co appear. At 450 °C the CoO almost completely disappeared as the CoO is transformed into metallic cobalt. By further increasing the temperature to 650 °C, the peaks of LiO_2 started to appear in the graph, pointing to the dissolution of LiOH. (Xie et al., 2023)

LNCM ($LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$) also shows similar results when reduced via hydrogen. LNCM completely disappears at 400 °C, while still remained unchanged at 350 °C. At 450 °C peaks of LiOH and MnO appear and NiO is reduced to Ni completely. MnO is not reduced at all temperatures. In both LNCM and $LiCoO_2$ temperatures higher than 650 °C caused a loss of LiO_2. At 450 °C, both compounds were able to form soluble LiOH and the transition metals were lowered to their lower valence forms. This resulted the leaching rates of 98.03 % and 99.36 %, for $LiCoO_2$ and LNCM respectively. (Xie et al., 2023)

5 SUMMARY

Lithium-ion batteries are a valued method of energy storage with multiple valuable earth metals in them. In a world, where society is moving towards the electrification of transportation, the demand for LIBs is increasing and the supply of these valuable metals is limited. Therefore, a clean, environmentally friendly method of recycling is needed.

Currently LIBs are recycled in two main categories of methods. These are pyrometallurgical and hydrometallurgical methods. In pyrometallurgical methods, the spent LIBs are subjected to high temperatures, to either smelt the materials or reduce them via reduction roasting. In smelting, battery materials are smelted, and these valuable metals are reclaimed as alloys. In reduction roasting, battery materials are reduced via carbon or by aluminium to a form that can be reclaimed in hydrometallurgical methods.

In hydrometallurgical methods, such as leaching, are used in combination with pyrometallurgical methods to purify the recycled cathode materials. These methods have the disadvantage of needing lots of energy and lots of environmentally harmful chemicals in order to operate. Less common methods of recycling are direct recycling methods. In them, the spent LIB is disassembled and then directly with certain processes re-lithiated with fresh lithium to be reusable.

Hydrogen-assisted recycling of LIBs is currently a highly researched topic as it is a greener alternative to the currently used recycling methods. Hydrogen is used in the reduction process as a replacement to carbothermal reduction. The main advantage of hydrogen reduction in comparison to carbothermal reduction is the production of water as a byproduct instead of carbon dioxide like in carbothermal reduction. Other advantages are faster reduction time, lower required operating temperatures, and therefore lower energy consumption. In hydrogen reduction, hydrogen molecules react with the battery materials and as a result lithium is transformed into a soluble form, and the higher valence metals are transformed into lower valence forms of themselves.

Using hydrogen for recycling LIB is being researched to find the effective parameters for optimization of process. The current research shows that it can significantly improve the process environmentally and economically, when compared to currently used recycling methods.

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