

Review

Review of Achieved Purities after Li-ion Batteries Hydrometallurgical Treatment and Impurities Effects on the Cathode Performance

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Abstract: This paper is a product purity study of recycled Li-ion batteries with a focus on hydrometallurgical recycling processes. Firstly, a brief description of the current recycling status was presented based on the research data. Moreover, this work presented the influence of impurities such as Cu, Fe and Mg on recovered cathode materials performance. The impact of the impurities was described depending on their form (metallic or ionic) and concentration. This work also reviewed hydrometallurgical recycling processes depending on the recovered material, obtained purity and recovery methods. This purity data were obtained from both research and battery industry actors. Finally, the purity study was completed by collecting data regarding commercial battery-grade chemical compounds and active lithium cathode materials, including required purity levels and allowed impurity limitations.

Keywords: LiB (lithium-ion battery); hydrometallurgy; purity; recovery; impurity effect; cathode performance



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1. Introduction

The urgency of the ongoing climate change forces transition from fossil fuels to sustainable energy solutions, including bioenergy as well as advanced battery technologies with Li-ion batteries (LiBs) in focus [1]. Thus, the popularity of LiBs has grown significantly in the automobile sector as it is used in both hybrid and electric vehicles but even in modern-life portable equipment, i.e., mobile phones, tablets, cameras, etc. [2]. The transition from fossil fuels and growing energy demand together with lithium's high energy density, low atomic weight, good electrochemical performance, a broad range of operating temperature and low rates of self-discharge have contributed to the continuous expansion of the LiBs market [1,3]. The growing battery demand creates opportunities but as well concerns related to the product's lifetime as significant amounts of solid LiBs waste are being generated yearly. In 2019 it was approximately 100,000 tons of spent LiBs, but it is expected to increase up to 700,000 tons in 2025 [3,4]. This, combined with irresponsible discarding, results in environmental contaminations and health issues due to high concentrations of heavy metals and the presence of toxic electrolytes; in the deposited batteries, lithium generated during the discharge cycles is likely to react with water, which is risky due to the violent reaction [2–5]. However, within the European Union, landfilling is forbidden, and recycling of spent batteries is compulsory [6]. The battery industry is the largest end-use market for lithium, and it is predicted that the need for lithium carbonate will almost double between 2015 and 2025 from 265,000 tons to 498,000 tons, which puts pressure on the raw resource supplies; thus, it is approximated that the commodity market will come across lithium shortages already by 2023, and so, the spent LiBs can be considered as lithium reserve [5]. As a result, recycling has gained more attention in the past years. The

development of recycling processes creates opportunity not only from the environmental perspective but also from the economical point of view as it helps to recover and reuse valuable metals of limited supplies [3,4]. Waste management within the battery sector, in general, has been a known problem, and therefore, there are actions taken on both national and international levels. Extended Producer Responsibility (EPR) policy has been introduced in European Union, United States of America and Canada in order to raise customer awareness and develop battery-based waste collection infrastructure. Moreover, European Union has introduced the 2006 Battery Directive regulation that gives minimum target rates of collection and recycling for each member country [7].

The impurities are problematic for the recycling processes of LiBs as they challenge different technologies in different ways. Impurities influence production processes, product quality and performance, and what is most important, safety [8]. In the case of LiBs, safety is of great concern as they have a high energy density comparing to the other rechargeable materials. This can cause a temperature increase resulting in the ignition of the flammable organic solvents and an even further temperature increase [8]. Most of the LiB product faults have their origin in the internal faults that get triggered by the impurity presence. This, together with the production process and safety aspects, puts pressure on the material purity with a focus on trace elements characteristics and their content. According to Ardia et al. [9], the total impurity can be defined as a sum of impurities (metals in the ionic form) and contaminants (elements in a metallic form) [8,9]. In the hydrometallurgical recovery process, the final recovered product contains impurities mostly in ionic forms [9].

This work mainly focuses on reviewing current hydrometallurgical recycling processes with a focus on the process steps that are directly related to the final purity levels. Moreover, the influence of the impurities in the spent LiB cells are studied.

2. Recycling

The most interesting battery part from the financial perspective is the cathode material, and it is worth mentioning that as the technology development progresses, the changes in the cathode materials occur to achieve a high energy density for lower price. This makes the cathode material account for around 90% of a battery's total value [3]. The first-generation cathode material used in LiBs is LiCoO_2 , and therefore, the active materials of this cathode are studied the most. The newer generation cathodes are becoming more common and materials, such as LiMn_2O_4 , $\text{LiNi}_x\text{Mn}_y\text{Co}_{(1-x-y)}\text{O}_2$ and LiFePO_4 , have gained popularity, which creates new demands on the recycling procedures [3]. The majority of the current recycling approaches focus on the recovery of highly valuable metals—Co. In the majority of the current hydrometallurgical processes, Co recovery is prioritized in leaching and precipitation, meanwhile Li tends to remain in the leaching solution together with the impurities. When it comes to the trending pyrometallurgy processes, the main focus is put into Co and Ni recovery, meanwhile Li is lost in the slag [5]. Trending recycling technologies, the processes that are used to recover metals of high value from the spent LiBs, involve electrochemical methods and hydrometallurgy [3]. As pyrometallurgy and hydrometallurgy are being developed, the processes have been applied in the industrial scale within the battery industry. Umicore, SONY, OnTo, Accurec, Inmetco and Xstrata are example companies that have applied pyrometallurgy. In order to recover valuable metals, pyrometallurgy (the method of applying high temperature processes, such as smelting) is often combined with hydrometallurgy (aqueous chemistry, including steps, such as leaching, precipitation, solvent extraction and ion exchange). Such a combined process for recycling LiBs is applied by Umicore [10–12]. Example of the companies that, instead of combined process, only use hydrometallurgy to recover metals from spent LiBs are Fortum [13] and Northvolt [14]. According to the review commissioned by The Swedish Energy Agency [15], by 2019, there were approximately 50 companies worldwide that implemented a recycling process on spent LiBs. The recycle scale was everything from laboratory to industrial scales. The majority of the companies exist in China, and the second biggest market is located in South Korea, followed by EU, Japan, Canada and the

United States of America. It is expected that the number of LiBs that reach the end of life globally is expected to be around 700,000 tons by 2025; at the same time, worldwide recycling is expected to take care of around 400,000 tons of spent LiBs [15]. By 2019, the LiBs recycling market in China was corresponding to around 30 companies. The companies achieved a different recovery degree, but almost all of the companies applied hydrometallurgy. The main recovery products in this market are chemical products, such as Ni and CoSO_4 or Li_2CO_3 . Another type of products are complete blends for precursor production—NCM and NCA cathodes [15]. According to the same data from 2019 [15], European LiBs recycling companies do not obtain as high recovery efficiency as China. The main reason behind it is lower spent batteries volumes in Europe, which makes it unprofitable to extract all the substances from the waste. In the European market, the most common recovery methods are hydrometallurgy and a combination of hydrometallurgy and pyrometallurgy [15].

3. Battery Composition and Structure

With time, the batteries have become larger and assembled rather than portable. The portable old type of batteries was challenging for the recycling processes from the sorting perspective. Industrial batteries or batteries in electric vehicles are, on the other hand, assembled in bigger modules; these modules are assembled in battery packs. This creates new challenges for recycling processes as preparation prior to recycling is more advanced. Moreover, LiBs are mostly built into the devices so that the user is not meant to disassemble the battery from electric vehicles, but this is also more and more common in devices, such as PCs and smart phones. This means that spent LiBs do not directly go to the battery recyclers but to the electric vehicle recycler, where the battery can be disassembled [15]. The first batteries that were used in computers and mobile phones were only LCO battery types (LiCoO_2), consisting of 17%–20% of Co, which was the most valuable material. Later on, along the development of various electric equipment, such as electric bikes and busses, the LEP batteries (LiFePO_4) gained popularity. These types of batteries do not contain Co, and this makes them not profitable to recycle. Another common battery type that is commonly used in, for example, electric scooters is LMO (LiMn_2O_4). The most common batteries in the western world are NMC ($\text{Li}(\text{NiMnCo})\text{O}_2$) and NCA ($\text{Li}(\text{NiCoAl})\text{O}_2$) as they are most commonly used in electric vehicles. NMC batteries contain 2%–6.5% Co and NCA batteries contain less than 3% Co [15]. The outer shell is, most often, made of plastic or stainless steel, and the materials that are usually used in separators are polymeric materials. The anode consists of graphite powder and polymer binder, both coated by a foil manufactured from Cu [16].

4. Hydrometallurgical Approach in Li-ion Batteries Recycling

The hydrometallurgical recycling process has been found as the more popular metal recycling method than pyrometallurgical processes since they allow achieving very high purities of recycled metal compounds and have a lower impact on health and environment. Moreover, hydrometallurgy is a popular recovery method used for LiB recycling as it results in high yield recovery, low energy demand, no air emissions and complete recycle of pricy metals with high purity levels [3]. The hydrometallurgical process is usually conducted with a pretreatment step that is followed by leaching, separation and recovery of valuable metals from the leachate [12]. The complex composition of LiBs makes the pretreatment step a helpful part of the recovery process. Pretreatment makes it easier to separate different battery components in order to get active materials from the cathode, which is further used in the leaching step. The pretreatment process usually consists of discharging, dismantling, mechanical separation and active materials (black mass) recovery [12].

Spent LiB discharging is conducted to increase security in the further steps of the recycling process as it decreases the hazard of short-circuiting and self-ignition. Separating and dismantling are conducted after the spent battery is discharged. The battery parts that are separated and dismantled are, most often, anodes, cathodes, steel or plastic cases

and organic separators [12]. This step can be conducted manually on a small scale or mechanically on a large industrial scale. Afterwards, in the final pretreatment step, the active materials found in cathodes are separated from the cathode current collectors by using solvents or thermal treatments [12,17].

After pretreatments, leaching is applied as the next step in the hydrometallurgy process. This step is considered to be the most significant one when it comes to valuable metals recovery as its purpose is to convert the metals present in the cathode material procured in the pretreatment process into ionic solutions [12]. The leaching media is, most often, mineral acids (H_2SO_4 , HCl and HNO_3), alkali and organic acids. The ions are furtherly recovered in series of chemical treatments: precipitation, solvent extraction, and electrolytic deposition. Studies show that inorganic acids are highly effective for the recovery of Co and Li (99%) in the leaching procedure when applied in optimal conditions [1,12,18].

As the leaching solution has complicated composition, the separation step is necessary to accomplish recovery of the valuable metals (Li, Co, Ni, Mn, Cu, Al, Fe). This can be done by adding a selective leaching step prior to the separation to make the separation process simple. Another approach is proceeding with separation directly after the leaching procedure. The most common separation methods are solvent extraction, chemical precipitation, electrochemical deposition. Due to the leaching solution's complex composition, it is complicated to recover all the valuable components from the leachate by applying only one separation method. The separation procedure is usually a combination of two or more of the mentioned methods [12]. Figure 1 shows a flowchart of the generalized hydrometallurgical process.

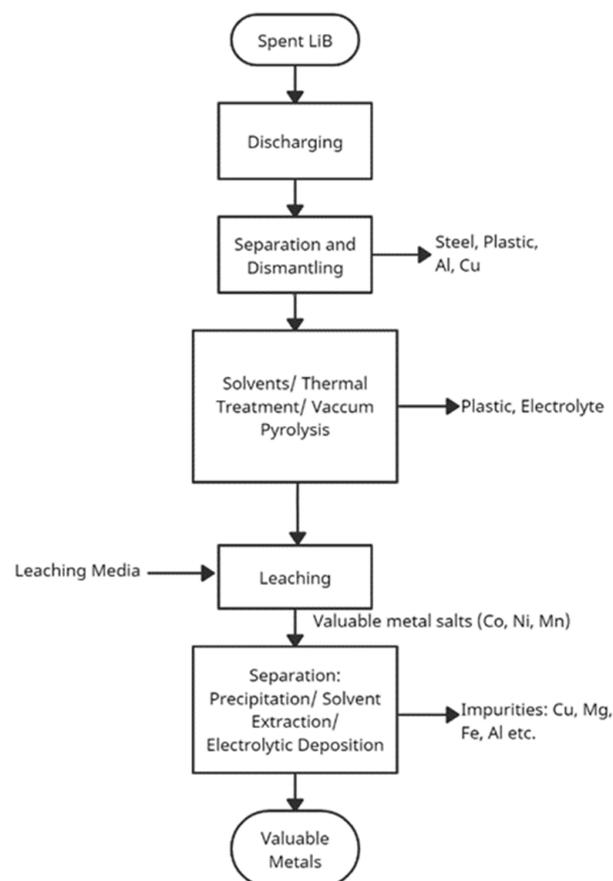


Figure 1. The general hydrometallurgical process [12].

5. Cu, Al and Fe as the Impurities in the Hydrometallurgical Process

When it comes to recovered LiBs, the impurities influence the electrochemical performance, lifetime and stability of the recovered material. Thus, understanding the impact that impurities have on recycling technologies is of great importance in order to develop the LiBs recycling technologies [8]. Cu, Al, Fe and Mg are common impurities that can be found in the leach liquor after acidic leaching. These impurities can be found in the leachate together with Li, Ni, Co, Mn that are obtained from the active cathode material of LiBs [19].

In the hydrometallurgical recovery process, metallic impurities can be, most often, removed based on their distinct solubility by applying chemical precipitation. In such a case, when chemical precipitation is combined with adjusted pH, metallic Al and Fe are considered easy to remove. However, Cu ions are more difficult to remove as their distinct solubility constant has values similar to Co, Ni and Mn ions [8].

On the other hand, the result of the study by Peng et al. [7] showed that increased levels of metallic Cu, Al and Fe result in increased leaching efficiency of Li and Co. Moreover, the overflow of battery waste that is rich in Cu and Al supports the dissolution of Co and Li from the underflow fractions, which is a very useful result for the recycling processes. In the paper [7], the overflow fraction of the battery waste is the fractions > 2 mm, mostly containing Al/Cu foils, as well as active materials. The underflow fractions are of a smaller size < 2 mm and comprise of high concentrations of Co- and Li-based active materials. This basically means that the fractions of current collectors should preferably not be deeply separated from the smaller fractions that are rich in Co. When it comes to Fe, it has been found that its presence in the solution is necessary in acid leaching as it catalyzes the reductive behavior of Al and Cu, which gives the leaching efficiency of Co and Li close to 100% [7].

Melin et al. [15] identified 80 studies that present recycling processes of LiBs where more than 90% of Li was recovered, and 86 studies with a Co recovery degree larger than 90%. Melin et al. [15] has also found 20 studies that bring up NCM cathode recycling and recovered more than 90% of the cathode material. It is reported that the majority of these research studies were conducted on a laboratory scale with optimal process conditions and proper control. Most of the mentioned studies report high purity levels. Even though there are plenty of industrial actors [15] dealing with hydrometallurgy of spent LiBs, there is not much data available about the exact purity degree provided by the recycling companies. Northvolt, for example, expresses the purity of the recovered material as high purity [14]. Fortum has recently reported [20] that they have developed a new LiBs recycling technology. The company patented a technology that is said to recover Li from electric vehicle batteries on an industrial scale. The quality (purity) of the recovered material is expressed as battery grade by Fortum. Due to a lack of information about the purity degrees of materials recovered from spent LiBs on the industrial scale, this study mainly focuses on reviewing research-based purity data rather than purity levels obtained on an industrial scale.

6. Obtained Purity Degrees—Research

Atia et al. [21] presented an enhanced hydrometallurgical process of black mass from heterogenous LiB waste. The work presented valuable products recovered with their purity degrees. The products were: Co_3O_4 (83% purity), CoC_2O_4 (96% purity), NiO (89% purity) and Li_2CO_3 (99.8% purity), which had the highest purity degree among the recovered products. NiO and Co_3O_4 did not reach market grade purity, while CoC_2O_4 and Li_2CO_3 did. However, Co_3O_4 obtained could still be used as mixed transition metal-oxide for cathode materials, such as NCM and NCA. When it comes to the efficiency of the recovery process, the recovery rates were reported as follows: between 80%–85% for graphite, 90% for Li and 90% for Ni. The process proposed included physical pre-treatment that was conducted to get electrode material in the form of powder. The obtained black mass was of heterogenous composition, containing C, Fe, P and F. Afterwards, the black mass was leached with a mixture of H_2O_2 and H_2SO_4 , and this step resulted in metal extractions of a

high degree. Leach liquor was purified by iron-precipitation, liquid-liquid extraction and Li-Na separation. Moreover, the presented process is expected to handle various compositions of LiBs. It can be applied on spent cathodes made of layered oxides (LiNiO_2 , LiCoO_2), spinels (LiMn_2O_4) and Li-mixed transition metal oxides (LiNiMnCoO_2 , LiNiCoAlO_2) [21]. The study by Liu et al. [22] presented a recovery process based on hydrogen reduction. The obtained product from spent LiBs was battery-grade $\text{LiOH}\cdot\text{H}_2\text{O}$ with 99.92% purity. The recovery method of the process included reductive hydrogen roasting, water leaching and crystallization of $\text{LiOH}\cdot\text{H}_2\text{O}$ as a final step. The recovered product was used for cathode material $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ production, and its electrochemical performance was compared with $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ produced with commercial LiOH as Li-source. The overall electrochemical performance of the cathode material where the recovered material was just has been judged as good. This statement has been made based on the tests that measured the electrochemical performance of both cathode materials. The test results indicated that the initial coulomb efficiencies are almost identical—with a value equal to 90.7% for the cathode material made of the recovered Li-source and 90.5% for the commercially produced cathode material. A comparison of the electrochemical performance of NMC made from the recycled material is shown in Table 1.

Table 1. A comparison of the electrochemical performance of $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ synthesized with commercial and recovered Li-source, according to Liu et al. [22].

| Cathode Material | Charging and Discharging Capacity | Initial Coulomb Efficiencies | Substantial Polarization of the Electrode | Capacity through Cycle | Specific Capacity Retention at 10 C | Discharge Capacity Recovery at 0.1 C |
|---|-----------------------------------|------------------------------|---|--------------------------------------|-------------------------------------|--------------------------------------|
| $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ synthesized with commercial Li source | Higher | 90.5% | - | Worse | 75% | Close to initial value |
| $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ synthesized with recovered Li source | Lower | 90.7% | Occurs | Better due to stabilizing Al-O bonds | 63% | Close to initial value |

Research by Peng et al. [23] focused on MnO_2 recovery from an acid leaching solution. The purity of MnO_2 was over 99.5%. The recovery process is mainly based on solvent extraction conducted in a couple of steps. Firstly, obtained leaching liquor underwent solvent extraction, then scrubbing followed by selective stripping and, finally, oxidative precipitation. D2EHPA was used in the solvent extraction step, and solubilized MnSO_4 was used to conduct scrubbing. Jo et al. [24] have also studied the recycling process of LiBs. In the described process, the final recovery product was Li_2CO_3 with a purity degree of 99.48%. The recovery method used in the process was based on taking advantage of different material solubilities depending on solvent and temperature. The paper by Chen et al. [2] is a study that presented a recovery process of $\text{CoC}_2\text{O}_4\cdot\text{H}_2\text{O}$, Li_2CO_3 , MnSO_4 and Ni from leaching liquor obtained through H_2SO_4 acid leaching. The purity degree of Ni was reported to reach 98%; for Li_2CO_3 and $\text{CoC}_2\text{O}_4\cdot\text{H}_2\text{O}$, the reported purity levels were 99% and 98%, respectively. The obtained purity of MnSO_4 was expressed as relatively high. The valuable metals were recovered from the leaching liquor by applying precipitation in the presence of organic compound $\text{CH}_3\text{C}(\text{NOH})\text{C}(\text{NOH})\text{CH}_3$ (diethylglyoxime) to recover Ni. Afterwards, MnSO_4 and $\text{CoC}_2\text{O}_4\cdot\text{H}_2\text{O}$ were recovered in the solvent extraction procedure in presence of D2EHPA. NH_4OH was used in the precipitation procedure to recover $\text{CoC}_2\text{O}_4\cdot\text{H}_2\text{O}$, while Na_2CO_3 was used for Li_2CO_3 recovery. Another research article that focused on the recovery of valuable compounds from spent LiBs is Granata et al. [25]. The study investigated two different recovery routes of Li_2CO_3 and CoCO_3 . The purity of the recovered Li_2CO_3 reached above 98% in both of the recovery routes, while purity grades for CoCO_3 were different for the two processes. The simpler recovery process included leaching, precipitation, followed by carbonatation and crystallization. In the leaching

process, the powder is leached with H_2SO_4 . Afterwards, hydroxides of Al, Cu and Fe were removed from the obtained leaching liquor in the precipitation step by applying pH 5.5. $CoCO_3$ was extracted in the next step—carbonatation—followed by Li_2CO_3 recovery in the final crystallization step. In this process, the route purity of $CoCO_3$ was reported as 36%–37%. The more advanced route of the recovery process included additional solvent extraction steps prior to carbonatation. $CoCO_3$ purity obtained in this route was higher and reported as 47%. The research article by Peng et al. [26] has presented a recovery process where Co in its metallic was the final recovery product with a purity of 98.8%. The recycled material from spent NMC- $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ went through basic pre-treatment in the presence of NaOH to get rid of Al. The next step of the process was leaching in presence of H_2SO_4 with the addition of H_2O_2 . Leaching liquor was then treated with one selective extraction in the presence of P-204 to recover Mn^{2+} , followed by one more selective extraction in the presence of P-507 to recover Li^+ , Ni^{2+} and Co^{2+} . Afterwards, Co^{2+} obtained in second selective extraction went through electrowinning step where to obtain metallic Co.

The final purity data that were found in the research are recent data as the majority of cases are not older than 2019. The most common recovery product is Li_2CO_3 , just like in the case of industrial data that have been reported, but there are also other recovery products listed. The recovery methods are various, but precipitation and solvent extraction are the most common ones, although the chemicals used in these steps are very different. To understand if the obtained purity degrees are good enough to be directly used in battery production, the above literature sources have been used, but there are still some question marks left due to lacking information regarding acceptable levels for battery-grade materials. A summary of some research data is shown in Table 2.

Table 2. An overview of summarized research data.

| Study Reference | Product | Purity Grade | Battery Grade | Recovery Method | Year |
|------------------------|-----------------------|--------------|---------------|---|------|
| (Atia et al., 2019) | Co_3O_4 | 83% | — | 1. Fe-precipitation 2. liquor-liquor extraction 3. Li-Na precipitation | 2019 |
| | CoC_2O_4 | 96% | — | | |
| | NiO | 89% | — | | |
| | Li_2CO_3 | 99.8% | + | | |
| (Liu et al., 2021) | $LiOH \cdot H_2O$ | 99.9% | + | 1. Reductive hydrogen roasting 2. Water leaching 3. Crystallization | 2021 |
| (Peng et al., 2019) | MnO_2 | >99.5% | ? | 1. Solvent extraction (D2EHPA) 2. Scrubbing ($MnSO_4$) 3. Stripping 4. Oxidative precipitation ($KMnO_4$ and MnO_2) | 2019 |
| (Jo and Myung, 2019) | Li_2CO_3 | 99.5% | - | Based on different material solubilities depending on solvent and temperature | 2019 |
| (Chen et al., 2015) | $CoC_2O_4 \cdot H_2O$ | 97.9% | ? | 1. Ni—precipitation ($CH_3C(NO_2)C(NO_2)CH_3$) 2. Mn, Co—solvent extraction (D2EHPA) 3. Co-precipitation (NH_4OH) and Li-precipitation (Na_2CO_3) | 2015 |
| | Li_2CO_3 | 99.2% | — | | |
| | $MnSO_4$ | rel.pure | — | | |
| | Ni^+ | 97.8% | ? | | |
| (Wang et al., 2019) | Metallic Co | 98.8% | ? | 1. Extraction with (I)P204 (II)P507 2. Electrowinning | 2019 |
| (Granata et al., 2012) | Li_2CO_3 | >98% | — | 1. Precipitation in pH 5.5 2. Carbonatation | 2012 |
| | $CoCO_3$ | 36–37% | — | | |
| (Granata et al., 2012) | Li_2CO_3 | >98% | — | 1. Precipitation in pH 5,5 2. Solvent extraction 3. Carbonatation | 2012 |
| | $CoCO_3$ | 47% | — | | |

7. Obtained Purity Degrees—Hydrometallurgical Industrial Actors

Fortum (Finland) is a company that applies hydrometallurgy as the recycling process of spent LiBs. In their process, the spent battery goes through a mechanical treatment where

plastics, Al and Cu are separated. Then the remaining chemical and mineral components that are present in the black mass (Li, Mg, Co, Ni) are recovered by applying chemical precipitation. The recovery rate for Fortum's process is 80% [13]. Northvolt (Sweden) is another example of a company that applies hydrometallurgy to recover valuable metals (Li, Ni, Mn, Co) from spent LiBs [14]. The batteries first undergo a discharging step, followed by dismantling where steel cases, Al current collectors and Cu wiring are removed. The next step is crushing the cells and modules, as well as the evaporation of the electrolyte solvent. The remaining material is then sorted, and the obtained black mass undergoes hydrometallurgy. In this step, metals are dissolved in sulfuric acid in optimized conditions. Impurities (Cu, Fe, Al) are removed from the solution by applying precipitation, solvent extraction and ionic exchange. Metals (Mn, Co, Ni) are recovered from the purified solution by applying solvent extraction. Li is extracted from the NCM solution at the very end, after the concentration levels and ratios of the solutions are adjusted to the production requirements. The final step in Northvolt's process is battery-grade metal compounds [14]. Another company Li-Cycle (Canada) claims a 95% recovery rate after hydrometallurgical processing of LiBs. The company recovers cobalt and nickel in a form of sulfates. Lithium and manganese are recovered as carbonates [27].

Recupyl [28] and OnTo [29] are examples of companies that also use or used a hydrometallurgy in their recycling processes.

The Recupyl process had an annual LiB recycling capacity of 110 tons (Figure 2). The main Li products recovered from this process were Li_2CO_3 and Li_3PO_4 . The recovery method used by Recupyl was a precipitation with CO_2 . There is no efficiency or purity data for this process, but it is known that their recovery products were meant to be used for cathode production [29].

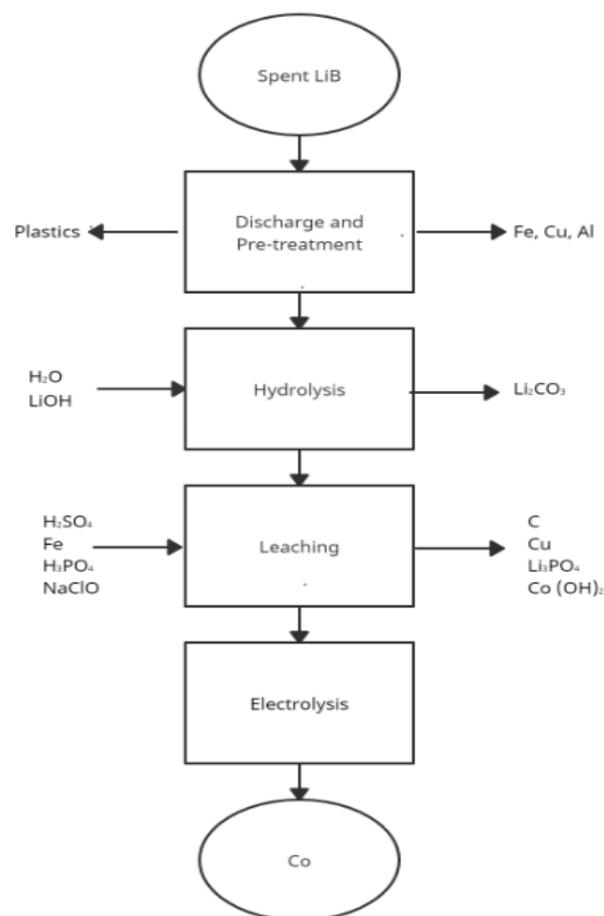


Figure 2. A simplified flowchart of Recupyl's process.

In the OnTo process (Figure 3), the anode and cathode purifications occur in two parallel hydrometallurgical processes. The anode is purified first at a low pH to remove the remaining cathode particles, then at a high pH so that Al and Cu are extracted. Cathode purification occurs in a basic solution to conduct anode particle extraction [29]. The purity of the recovered product Li_2CO_3 is 99%. Meanwhile, according to Linneen et al. [30], industrial-grade Li_2CO_3 for LiB applications require at least 99.5% purity. Thus, for the final recovered product to be used in battery production, the purity needs to be increased by, for example, blending with virgin materials [29].

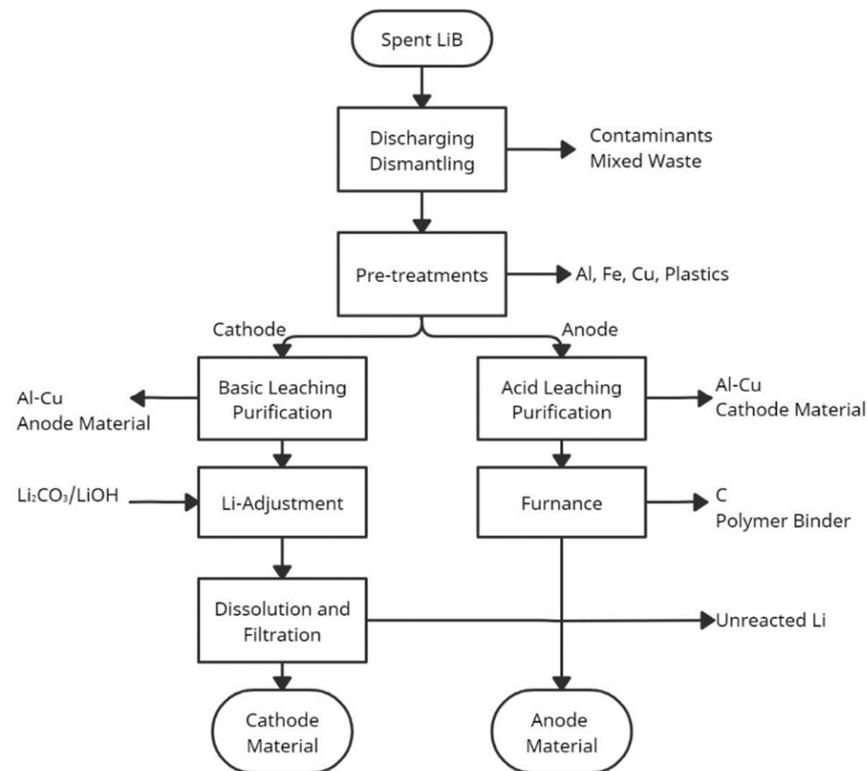


Figure 3. A simplified flowchart of OnTo's process.

Table 3 summarizes the few examples from the industry that were found. Recupyl recovers mainly Li_2CO_3 and Li_3PO_4 through precipitation with CO_2 . Previously, it has been mentioned that battery grade purity for Li_2CO_3 is above 99.5%. OnTo also recovers Li_2CO_3 , and the obtained purity is 99%, which is below the battery-grade requirement.

Table 3. Summary of industrial data.

| Company | Process Type | Product | Method | Purity Degree | Product Application |
|---------------------|------------------------------------|---|---|---------------|---------------------|
| Northvolt Fortum | hydrometallurgy hydrometallurgy | | Precipitation, SX Precipitation | Battery grade | |
| Li-Cycle | hydrometallurgy | CoSO_4 , NiSO_4 , Li_2CO_3 , MnCO_3 | Precipitation, SX | Battery grade | |
| Recupyl | hydrometallurgy | Li_2CO_3 Li_3PO_4 | precipitation with CO_2 | no data | cathode production |
| OnTo | hydrometallurgy | Li_2CO_3 | anode purification: low and high pH cathode purification: basic solution | 99% | cathode production |

8. The Effect of the Impurities on the Cathode Performance

8.1. Cu as the Impurity in Recycled Cathode Material

Research studying Cu-ion [31,32] impurity influence on the NCM-based cathode materials has found that this impurity in certain amounts might be beneficial. The impurity can improve electrochemical performance. Zhang et al. [8] presented more comprehensive research that studied Cu impurity in both metallic and ionic form and their influence on electrochemical and physical properties of recovered NCM622 cathode material. The presented results indicated that Cu in its metallic form has an undesired effect on the electrochemical performance of the NCM622 material, as it causes short circuits issues and therefore leads to Cu metal-based electrode failures. Fear et al. [33] and Zhao et al. [34] have also reported negative impacts of metallic Cu impurity as it has the ability to dissolve in organic electrolytes, which enables the metallic impurity to deposit on the surface of Li metal anode. This happens due to the migration of Cu dissolved in organic agents from the cathode, through the separator, to the surface of the Li anode. When the impurity deposits on the surface of Li anode, it causes Li dendrite growth which ultimately leads to internal short circuits and so cathodes failure [8].

Zhang et al. [8] showed that Cu impurity in its ionic form has a more complex effect on NCM622 cathode material than the metallic form of the impurity as it strongly depends on the concentration. The same impurity in the same form but different concentrations have different effects on recovered cathode material NCM622. The right amounts of the Cu-ions (0.2–1.0 at%) behave as dopants and can have a positive impact on the material as they can facilitate the nucleation that contributes to the increase in primary particles. Another positive impact is the reduction of cation mixing. Small amounts of Cu-ions enable cycling stability as well as rate capability, which directly improves the electrochemical performance of the recovered cathode. The optimum concentration presented in the study was 1.0 at% of Cu ions. Cu in this concentration works as a dopant rather than an impurity.

On the other hand, excess of the same ions (0.5 at%) has shown undesired effects on the NCM622 cathode. The undesired effect is increased cation mixing degree in the cathode material, which has a negative impact on the recovered cathode's electrochemical performance. The large cation mixing degree prohibits Li-ions from transferring to the Li layer. Thus, decreased transport of Li-ions puts bad impacts on the specific capacity, cycling stability and rate performance of the recovered NCM622 cathode material [8].

8.2. Fe as the Impurity in Recycled Cathode Material

Cathode active material LiCoO_2 (LCO) has been in the center of attention in research and development within LiBs recycling [19]. A study by Contestabile et al. [35] and another one by Nan et al. [36] have shown that this cathode material can be resynthesized with a high efficiency degree by using acid leaching and chemical precipitation. The material obtained by applying acid leaching and chemical participation was not only recovered with high efficiency but also met performance demands. LiCoO_2 (LCO) cathode material has been replaced lately with NCM $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ material in the electrical vehicles industry due to NCM's better thermal stability, greater reversible capacity and lower price. Thus, the research focus has shifted towards NCM cathode material [19]. To understand the impacts of Fe, Park et al. [19] simulated the resynthesis of NCM ($\text{Li}[\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}]\text{O}_2$) and NCMF ($\text{Li}[\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}]\text{Fe}_x\text{O}_2$) active cathode materials with different amounts of Fe from the spent LiBs in the leaching liquor. It was reported that Fe impurity in recycling processes of LiBs can have a favorable impact on the resynthesized NCM cathode material. Fe, in small amounts, can contribute to higher power and cycling stability. The study has also reported that the crystal structure in NCMF becomes more and more disturbed with increasing concentrations of Fe due to the fact that the presence of Fe contributes to unwanted cation mixing on Li^+ sites. In NCMF, Fe concentrations at levels 0.05% and 0.25% start reducing overpotential leading, while Fe levels equal to 1% of Fe in NCMF result in the poorest overpotential leading ability. Another drawback of Fe impurity is that the ratio of Mn^{3+} ions is lowered with increasing Fe concentrations. However, the study

has reported that the increasing concentration of Fe in NCMF improves capacity retention and thus cycling stability.

8.3. Mg as the Impurity in Recycled Cathode Material

Mg is an additive that can be used in LiBs modifications to improve the cathode material. However, the drawback of using Mg from the perspective of hydrometallurgical recovery processes is removing it from the leaching solution. The impurity removal is usually related to the largest part of the expenses of the whole recovery process [37]. As previous research works have stated that Mg in small amounts behaves like doping and has the ability to improve the electrochemical performance of the cathode material, Weng et al. [37] has investigated Mg impurity in a recovered cathode material $\text{Li}[(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})_{1-x}\text{Mg}_x]\text{O}_2$ and the leachate itself. Through a series of pretreatments, the leaching procedure, purification and crystallization-solid state synthesis, the active cathode material of $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ was recovered from the leaching liquor, which, in its turn, was obtained from the spent LiBs. The recovered material $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ showed good electrochemical performance and that it can be used as a cathode material for rechargeable LiBs. Moreover, it has shown desirable properties, such as a layered structure and uniform spherical morphology [37]. The electrochemical analysis has shown that with increasing concentrations up to 5% of Mg^{2+} ions, the cell capacity of the $\text{Li}[(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})_{1-x}\text{Mg}_x]\text{O}_2$ decreases. However, low metallic Mg concentrations (up to 1%) turned out to have a positive impact on the cycling performance, while the higher content of metallic magnesium (with concentrations larger than 2.5%) cause a loss of the capacity and declined cyclability as at these levels; the Mg^{2+} ions cannot be homogeneously distributed anymore. Mg levels up to 360mg/L in the leachate are acceptable. This level is higher than what is usually required in the impurity removal process. This result can be useful for recycling companies and can contribute to lowering the purification cost and saving time in the purification procedure.

Besides the research, the industry sector also shows interest in a deeper understanding of the impurities. Volvo Cars and Northvolt have just started a new research project called NyBat [38]. This project will examine the performance and quality of Northvolt's lithium-ion NMC battery cells, made of materials obtained from spent batteries used in Volvo Cars. This project will focus on studying the impact of non-metallic contaminants and their influence on battery performance, which has not been published before.

9. Producer Purity/Impurity Data of Battery Grade Chemical Compounds

In this study, the purity research has been partly conducted by gathering information about single impurities and their influence on the recovered battery material. However, the industrial data regarding the purity subject has turned out to be hard to obtain. Thus, to get more understanding of the chemical compounds that are found in LiBs, producer data of battery-grade chemical compounds and active lithium cathode materials have been summarized below. Presented data has been brought from product specification sheets and material safety data sheets. The data is not consistent, as the impurities and purity levels are presented in different forms depending on producer and compound. In many cases, there is no direct information regarding the overall purity level of each compound, and instead, the impurities and their maximum concentrations are listed. The impurity limits in some commercial precursors are summarized in Table 4 and a content limits of commercial cathode materials is shown in Table 5.

Table 4. Impurity limits in commercial precursors (if not stated differently the purities below are given in ppm).

| Precursor | Purity Level | H ₂ O | Na | Ca | SO ₄ | Fe | Al | Cu | Ni | Mg | Co | Zn | Cl | Mn | Pb | Cr | Cd | Si | K | LiOH | CO ₂ | HF | LiPF ₄ | Insolubles |
|--|--------------|------------------|----------|----------|-----------------|----|----|----|--------|----|---------|----|-----------|------|----|----|----|----|----|-----------|-----------------|----|-------------------|------------|
| Li ₂ CO ₃ [39] | ≥ 99.5 wt% | 0.5 wt% | 0.05 wt% | 0.04 wt% | 0.1 wt% | 5 | 10 | 5 | 6 | | | 5 | 0.01 wt% | | | | | | | | | | | 0.02 wt% |
| LiOH·H ₂ O [40] | | | 20 | 15 | 0.01 wt% | 5 | 10 | 5 | 10 | | | 10 | 0.002 wt% | | 10 | 5 | | 30 | 10 | ≥56.5 wt% | ≤0.35 wt% | | | 0.01 wt% |
| NiSO ₄ [41] | | | 15 | 4 | | 3 | 5 | 1 | 22 wt% | 10 | 100 | 3 | 10 | 2 | 5 | 5 | 1 | 10 | 10 | | | | | 50 |
| CoSO ₄ [42] | | | 12 | 12 | | 5 | 12 | 3 | 100 | 10 | ≥21 wt% | 5 | 12 | 5 | 5 | 5 | 5 | 12 | 5 | | | | | 50 |
| MnSO ₄ ·H ₂ O [43] | | | 50 | 50 | | 10 | | 10 | | 50 | | 10 | | ≥32% | 10 | | 10 | | | | | | | |
| LiPF ₆ Solution in EMC [44] | | 15 | 1 | 1 | 2 | 1 | | | | | | | 1 | | 1 | | | | 1 | | | 50 | 0.95–1.10 M | |
| LiPF ₆ solution in PC [45] | | 15 | 1 | 1 | 2 | 2 | | | | | | | 1 | | 1 | | | | 1 | | | 50 | 0.95–1.10 M | |
| LiPF ₆ solution in DMC [46] | | 15 | 1 | 1 | 2 | 1 | | | | | | | 1 | | 1 | | | | 1 | | | 50 | 0.95–1.10 M | |

Table 5. Content limits of commercial cathode materials.

| Cathode Material [47–51] | Li+Mn+Co (wt%) | Li (wt%) | Ni (wt%) | Co (wt%) | Mn (%) | Fe (wt%) | Cu (wt%) | Zn | Ca (wt%) | Na (wt%) | S | Moisture | Li ₂ CO ₃ |
|--------------------------|----------------|------------|------------|------------|------------|----------|----------|-----------|----------|----------|-----------|-----------|---------------------------------|
| NMC 622 | 58.0 | 7.0~8.0 | | | | 0.01 | 0.005 | | | 0.03 | | 0.05 wt% | 0.2 wt% |
| NMC 532 | 57.0~62.0 | 7.0~7.6 | 50.0 ± 1.0 | 20.0 ± 1.0 | 30.0 ± 1.0 | ≤0.005 | ≤0.002 | | ≤0.01 | ≤0.03 | | | |
| NMC 811 | 58.0~60.5 | 7.1~7.6 | | | | <0.005 | <0.002 | <0.02 wt% | <0.01 | <0.03 | | | |
| NMC 111 | | 7.60 ± 1.0 | 20.0 ± 1.5 | 20.0 ± 1.5 | 16.0 ± 1.5 | ≤100 ppm | ≤20 ppm | ≤20 ppm | ≤300 ppm | ≤100 ppm | ≤4000 ppm | ≤1500 ppm | |
| NMC Ni83 | 58.5~60.5 | 7.0~7.6 | 83.0 ± 0.7 | 11.0 ± 0.5 | 6.0 ± 0.5 | ≤0.005 | ≤0.002 | | ≤0.01 | ≤0.005 | | | |

10. Conclusions

This work is a literature study that analyzes the purity in recycled LiBs and reviews hydrometallurgical recycling technologies. This study has given a brief overview of the ongoing electrification development of the battery industry and related challenges. Impurities have been presented as one of the curtail challenges of recycling processes. The common impurities (Cu, Fe, Mg) in LiBs were reported together with their influence on the hydrometallurgical process and the recycled cathode active material depending on their form (metallic or ionic) and concentrations. It has turned out that impurities have a complex influence on cathode materials and, in some cases, can contribute to an increase in the electrochemical performance meanwhile in others ruin the cell. When it comes to recycling technologies, there is plenty of processes presented by the research. They are mainly conducted on a lab scale, but many of the studies report that the processes should be easy to scale up for industrial production. The purity degree of products recovered from spent LiBs is usually reported to be high; however, it does not often reach battery grade. Thus, the recovered products need to be furtherly treated by, for example, mixing with virgin material before being reintroduced to battery production.

Moreover, the purity data of recovered materials from active cathode materials have been gathered from both research and battery industries. The research data have been reported with purity degree, recovery product and recovery method. The purity data obtained from battery industries are not as informative as research data due to low accessibility to information or intellectual properties. This work also presents purity requirements for commercial products. The data has been gathered from diverse product specification sheets and material safety data sheets from producers within the battery industry. Collected information specifies purity levels for both precursors and cathode materials as well as commercial limitations of common impurities.

Based on this paper, it can be concluded that impurities are a complex subject. There are plenty of different cathode active materials, and each impurity can influence a cathode material in a very different way depending on its form and concentration. It has also been found that the tolerance margin for impurities is not large as a small change in impurity concentration easily makes doping properties turn into the cell destructive properties. This creates challenges for the battery industry, both recycling and manufacturing processes.

Impurities, their limitations and their influence on materials recovered by hydrometallurgical methods from LiBs is still a relatively new subject. Majority of the studies that this paper refers to have been published in the past couple of years. As the battery sector and the recycling processes are continuously being developed, the main subject of this paper is still a relatively new field that is at the beginning of its development. In order to get a deeper understanding of impurities, more research data should be published.

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