

Overview of Recycling Techniques for Lithium-Ion Batteries

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Abstract

This paper presents a literature review on the processing of used lithium-ion batteries in both industry and research. On an industrial scale, lithium-ion batteries are primarily processed through pyrometallurgical methods, leading to incomplete utilisation of lithium cells. On the other hand, the hydrometallurgical route of recycling lithium-ion batteries poses challenges, such as large-scale discharging or inert gas pretreatment, largely due to explosion hazards. Modern methods of lithium-ion battery recycling are oriented toward refining the leach liquor through solvent extraction methods using D2EHPA and Cyanex 272, to recover Co, Mn, and Ni. The final Li product is obtained through Na₂CO₃ precipitation.

Keywords

Lithium batteries, recycling, lithium extraction, solvent extraction method, pyrometallurgical treatment, hydrometallurgical treatment

1 Introduction

In recent years, there has been a noticeable increase in lithium production driven by the growing interest in electric vehicles (EVs) and consumer electronics. Lithium, often referred to as “white gold,” finds application not only in lithium cells but also in ceramics and glass manufacturing, polymer production, lubricants, and air treatment. In 2022, global lithium production reached 134,000 t, marking a substantial 41 % growth compared to the previous year. It is expected that market trends moving towards decarbonisation will further drive the growth of battery production in the coming years. The largest lithium producers are Australia with 61,000 t, followed by Chile with 39,000 t, China with 19,000 t, Argentina with 6,200 t, Zimbabwe with 800 t, and Portugal with 600 t.^{1,2} Lithium is predominantly extracted from two sources: brines and hard rock mining (such as spodumene and petalite).³

For lithium production, the extraction of lithium from brine sources is a very efficient and cost-effective method. Lithium brines are sourced from Li deposits found in high-altitude salars, notably in regions of South America (e.g., Chile, Argentina, Bolivia), and China.⁴

In most lithium-ion batteries, the positive electrode contains a material based on Li_xMA₂, while the negative

electrode consists of graphite. Various materials, such as LiCoO₂, LiNiO₂, and LiMn₂O₄, are used on the cathode. Lithium-ion batteries contain flammable electrolytes and solutes like LiClO₄, LiBF₄, and LiPF₆. The batteries consist of metals, organic compounds, and plastics, with a content of 5–20 % cobalt, 5–10 % nickel, 5–7 % lithium, 15 % organic chemicals, and 7 % plastics. While these proportions may slightly differ among manufacturers, the presence of various components requires significant effort for the comprehensive processing of different types of lithium-ion batteries (LIBs) together.^{5–8}

2 Large scale

In general, lithium batteries can be processed by pyrometallurgy (PM), hydrometallurgy (HM), or biometallurgy. However, in practice, almost all recycling processes for lithium cells are hybrid processes, involving both mechanical and pyrometallurgical treatments before the final extraction of metals through hydrometallurgical processes. Electrolytes, plastic packaging, and binders can be removed by physical-mechanical processing or heat treatment.² Many industrial facilities do not fully utilise the material potential in lithium-ion batteries (LIBs) processing, as some lithium is lost in slag and fly dust, while metal Ni-Co-Mn-Cu alloy is recovered by pyrometallurgical processing.³ To recover all the metals from LIBs, a combination of hydrometallurgical and pyrometallurgical processes is typically necessary. In practice, various commercial methods have been employed for LIB processing, often tailored to specific cell types. As a result, a specific technology may not have universal applicability to all types of LIBs.⁴

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Note: The findings of this study were presented at the 4th International Convention of Scientists, Specialist Employees, and Students on the topic of Environmental Protection in the Republic of Croatia (4th ZORH Convention), held on April 20–21, 2023, at the Faculty of Chemistry and Technology University of Split, Croatia.

The German company Accurec uses a hybrid approach for recycling LIBs. The process begins with the mechanical treatment of LIBs, followed by low temperature pyrolysis (250 °C) which removes organic compounds, like electrolytes and solvents. The subsequent steps include size reduction under 200 µm, screening, as well as magnetic and gravitational separation to recover metals. The black mass is then mixed with coke and slag formers, reduced in a shaft furnace, resulting in the production of a Co-Mn alloy and lithium-enriched slag. The slag is then subjected to leaching and precipitation processes to obtain LiCl.^{3,5-7}

The OnTo recycling process consists of several steps. It begins with the cleaning and disassembly of discharged LIBs. Subsequently, the extracted materials are placed into a reactor and submerged in a mixture of liquid CO₂, ammonia, and other additives. This controlled environment leads to an increase in temperature and pressure, causing the cells to fail and rendering the cells inert. LIBs undergo size reduction, and are then subjected to separation processes involving wet shaking tables, gravity, and magnetic separation to obtain active materials.^{3,5}

The Retrieval process uses cryogenic physical-mechanical pretreatment and hydrometallurgical treatment for recycling LIBs. The process begins by cooling the LIBs using liquid nitrogen, bringing the temperature down to -196 °C. The LIBs are crushed with the addition of a lithium-containing solution. The next step is gravity separation on shaking tables, which produces three fractions: a plastic fraction, concentrate with high copper and cobalt content, and a slurry. The produced slurry is then filtered and the obtained solution is used to produce Li₂CO₃ by precipitation with Na₂CO₃. The next marketable product is the filter cake containing Ni, Mn, Co, and Cu.^{2,5,8-10}

The Umicore ValÉas process combines both pyrometallurgical and hydrometallurgical methods for recycling LIBs. LIBs are fed directly into furnaces without prior treatment. Plastics and graphite act as reducing agents and fuel. Pyrometallurgical processing results in an alloy comprising Co, Cu, Fe, and Ni. This alloy is treated by leaching in H₂SO₄. Solvent extraction is then employed to separate the individual metals. The final processing products obtained are CoCl₂, Ni(OH)₂ and NiSO₄. However, a drawback of this process is its incomplete utilisation of the entire material potential of the used LIBs, as the lithium ends up in the slag without further processing.^{2,5,11,12}

Akkuser recycling technology relies on physical-mechanical pretreatment for the processing of LIBs. The process initiates with size reduction using a knife mill to achieve fractions within the size range from 12.5 to 25 mm. Any emerging gases, primarily H₂ and O₂, are captured and then passed through filtration cyclones. Following this, the material undergoes additional size reduction to achieve fractions below 6 mm. Subsequently, magnetic and gravity separation is employed to create a marketable product suitable for subsequent metallurgical processing.^{5,13-15}

The LithoRec process begins by discharging the LIB to eliminate the risk of cell explosion. Accordingly, the lithium cells, especially cellblocks like those from EVs, are manually disassembled, and crushed. The crushed material, with

a size below -20 mm, is heated to temperatures between 100–140 °C in an inert atmosphere. The heating process facilitates the removal of organic compounds like solvents and electrolytes. The crushed lithium cells are then subjected to separation using a zigzag separator, which effectively separates aluminium, iron/steel, copper, and plastics. The electrode material is further sieved to obtain a fraction below -500 µm, which is leached. The leachate is then filtered to separate the graphite. Subsequently, the solution is subjected to the precipitation process to recover Ni, Co, and Mn compounds, or they can be obtained through electrochemical methods. The refined solution is precipitated or crystallised to recover lithium salts as Li₂CO₃ or LiOH.¹⁶⁻²⁰

The Sumitomo process commences with calcination at 1000 °C to remove organic solvents and electrolytes. After this stage, the remaining fractions undergo pyrometallurgical processing, resulting in the production of a Co-Ni-Fe alloy. Unfortunately, the lithium is oxidised and forms slag. Following this, a leaching process is employed to extract the desired metals from the metal alloy. The primary product of Sumitomo-Sony is high-quality cobalt oxide (CoO), which meets the necessary standards for use in the field of LIB production.^{16,21-23}

The primary challenge in LIB recycling stems from the rapid diversification of electrode materials. Even two vehicles that are the same model from the same manufacturer may incorporate batteries with different technologies and compositions.²⁴ This variability poses a significant challenge for hydrometallurgical methods, which need to be suited to electrode materials. In contrast, pyrometallurgical processing offers broader applicability, accommodating a wider range of inputs. The differences in the chemical composition of LIBs can negatively influence both the process of metal extraction into solution, and their subsequent recovery through extraction methods, as well as the purity of the final product. One key reason for the prevalence of pyrometallurgical processes is the inadequate labelling and disposal of batteries. With proper labelling, it would be easier to identify the battery's composition. This, in turn, would facilitate more precise sorting, for the best suitable hydrometallurgical recycling method tailored to the specific active materials, reducing losses in the recycling process.²⁵⁻²⁷

2.1 Physical-mechanical treatment

The physical-mechanical treatment of spent LIBs carries risks, due to the highly reactive nature of lithium and organic compounds, which can lead to fires or explosions. To mitigate these risks during LIB crushing, several approaches have been proposed. These include cryogenic crushing, processing LIBs in an inert atmosphere (under vacuum or with inert gases). However, these methods are often challenging and expensive.⁹

An alternative approach is to discharge end-of-life lithium cells using an external device, although this requires labour-intensive manual work when performed on a large scale. To ensure the economic feasibility of the discharge phase, it should have the capability to bulk-discharge LIBs from various manufacturers. One proposed method for

large-volume battery discharge involves immersing them in a brine solution. This is because the combined effect of a short circuit and electrolysis of the brine solution is expected to effectively discharge the battery.¹⁰

Ojanen *et al.* conducted research on the discharging of LIBs, comparing various electrolytes, including NaCl, Na₂SO₄, ZnSO₄, FeSO₄, and variations in their concentrations, mixing conditions, and the addition of metal powders, to assess the duration and efficiency of the discharge process. In *ex-situ* experiments, NaCl proved to be the most efficient electrolyte for discharging LIBs, with an increased concentration of up to 20 %, resulting in a reduced discharge time of 4.4 h. However, safety concerns arose due to the formation of chlorine gas. Consequently, the authors also investigated the use of sulphate salts. The drawback of using sulphate salts was the formation of a precipitate and passivation of Pt wires, which hindered the discharge reaction. However, when stirring was introduced into the Na₂SO₄ solution, the discharge rate accelerated, and the battery was fully discharged within 3.1 h.

Xiao *et al.*¹² introduced a method for treating used lithium cells by discharging them in a 5 % sodium chloride solution for 24 h, followed by crushing and desulphurising of black mass. Subsequently, the active mass underwent roasting in an inert nitrogen atmosphere, and leaching in water for 45 min, to achieve lithium conversion in solution. The lithium was then precipitated as Li₂CO₃. The filtration cake with high Mn and graphite content was oxidised to form high-purity Mn₃O₄ (95.11 %). Saeki *et al.*¹³ introduced a mechano-chemical process for LIB treatment, involving the grinding of LIBs with the addition of PVC. PVC served as the chlorine source, which formed water-soluble Co and Li chlorides. Ninety percent of the chloride from PVC reacted with Co and Li after 30 h of milling. The neutral leaching process resulted in a yield of nearly 100 % Li, and 90 % Co.

2.2 Leaching

The initial stage in hydrometallurgical processing is known as leaching, and the rate of this process is influenced by several factors, including the concentration of leaching reagents, mixing speed, temperature, leaching duration, and the ratio of solid to liquid.¹⁴ Based on the information provided in Table 1, it can be inferred that the most favourable leaching agent is sulphuric acid when complemented with hydrogen peroxide as an oxidising agent. Sulphuric acid offers significant advantages due to its cost-effectiveness and the reduced investment required for equipment in this process.

3 Possibilities of refining solutions containing lithium

Precipitation is the most widely used method for recovering metals and refining solutions after LIB leaching. In terms of precipitation, Li losses (10–30 %) are problematic, especially when precipitating Al and Fe from the solutions. Therefore, it is necessary to investigate refining options to reduce Li losses.^{16,17}

An innovative approach for extracting lithium from solutions is the use of “ion-sieve” materials. These materials are molecular structures that remain stable even after the target ions have been removed from the crystalline matrix. The voids or spaces created within the crystalline structure can only accommodate ions with ionic radii that are equal to or smaller than the target ions. In the case of lithium-ion sieves (LIS), they specifically adsorb lithium ions into these vacancies, as lithium possesses the smallest ionic radius among all metals. LIS can be categorised into two types: lithium-manganese oxides (LMO type) and lithium-titanium oxides (LTO type).¹⁸ LIS exhibit significant potential for recovering lithium from solutions. However, one drawback of using LIS is the gradual acidification of the solution dur-

Table 1 – Li and Co leaching yields^{3,15}

Tablica 1 – Prinosi izdvajanja Li i Co^{3,15}

Leaching agent	Leaching conditions	Yield/%
H ₂ SO ₄	25 °C and 75 °C, s : l = 1 : 10 (g ml ⁻¹) and 1 : 5 (g ml ⁻¹)	> 90 % Co, > 90 % Li
H ₂ SO ₄ + H ₂ O ₂	75 °C, s : l = 1 : 20 (g ml ⁻¹), 15 obj. % H ₂ O ₂	95 % Co, 100 % Li
H ₂ SO ₄ + H ₂ O ₂	65 °C, s : l = 1 : 30 (g ml ⁻¹), 1 obj. % H ₂ O ₂	75 % Co, 100 % Li
H ₂ SO ₄ + H ₂ O ₂	60 °C, s : l = 1 : 10 (g ml ⁻¹), 2 M H ₂ SO ₄ , 6 obj. % H ₂ O ₂	98 % Co, 97 % Li
H ₂ SO ₄ + H ₂ O ₂	75 °C, s : l = 1 : 10 (g ml ⁻¹), 2 M H ₂ SO ₄ , 5 obj. % H ₂ O ₂	93 % Co, 94 % Li
HNO ₃ + H ₂ O ₂	75 °C, s : l = 1 : 100 (g ml ⁻¹), 1,7 obj. % H ₂ O ₂	95 % Co, 95 % Li
HCl	80 °C, 4 M HCl	99 % Co, 97 % Li
HCl, NH ₂ OH·HCl	80 °C, s : l = 1 : 100 (g ml ⁻¹)	> 90 % Co, > 90 % Li
Malic acid + H ₂ O ₂	80 °C, 1,2 M malic acid, 1,5 obj. % H ₂ O ₂	94.3 % Co, 99 % Li
Citric acid + H ₂ O ₂	90 °C, s : l = 1 : 50 (g ml ⁻¹)	> 90 % Co, 100 % Li
H ₂ SO ₄ + Na ₂ S ₂ O ₃	90 °C, 0.25 M Na ₂ S ₂ O ₃ , 3.5 M H ₂ SO ₄	99.5 % Li

Table 2 – Overview of solvent extraction methods and optimal conditions for leachate refining

Tablica 2 – Pregled tekuće ekstrakcije

Leaching agent	Extractant	pH	Time/min	T/°C	O : A	SX yield	Ref.
H ₂ SO ₄	Saponified CYANEX 272	5.5–6	30	25	2 : 1	95–98 % Co	25
H ₂ SO ₄	CYANEX 272 + kerosene	7	10	25	2 : 1	90 % Co, 100 % Mn, 5 % Li	26
HCl	D2EHPA	1.5	120	32	1 : 1	75 % Li	27
H ₂ SO ₄	CYANEX 272 + Exxol D80	3	10	40	1 : 1	99.5 % Al	28
H ₂ SO ₄	10 % LIX84-IC (PC-88A) + kerosene	3	10	25	1 : 1	99.5 % Cu	29
H ₂ SO ₄	11 % LIX84-IC (PC-88A) + kerosene	4.6	10	25	1 : 1	99.1 % Ni	29
H ₂ SO ₄	10 % D2EHPA	3	10	25	1 : 1	98.9 % Mn	29
H ₂ SO ₄	Acorga M5640 + 5 % TBP	3.75	10	40	2 : 1	95 % Fe, 77 % Cu, 94 % Al	30
H ₂ SO ₄	15 % Cyanex 272 + Shellsol D70	5.5–6	10	22	2 : 1	90 % Co	31

ing the extraction process. This acidification can reduce the efficiency of LIS, especially when solution pH decreases. Therefore, it is typically more suitable to employ LIS for lithium extraction for primary production from brines and seawater.^{18–20} A forward-looking application of LIS technology might involve the development of ion-selective membranes. These membranes could enable the processing of acidic solutions, even those with higher lithium concentrations, expanding the range of applications for LIS technology.^{21,22}

Another promising method for lithium extraction is liquid-liquid extraction, often referred to as solvent extraction (SX), which has a broad range of applications. Table 2 provides an overview of the reagents and conditions employed in the extraction of metals from LIB leachates using SX. From the information presented in the table, it can be deduced that selectively extracting lithium *via* SX from acidic leachates using H₂SO₄ is not a feasible option. Therefore, solution refining and subsequent Li recovery is required. Cyanex 272 appears to be the most versatile extractant, which can be used to selectively recover Al and Co at different pH values. D2EHPA can also be used for Mn recovery and PC-88A for Cu and Ni recovery depending on the type of electrode material.

The most widely used compound is Li₂CO₃, acquired industrially through precipitation with Na₂CO₃. In terms of the precipitation of Li₂CO₃ from sulphate solutions, the efficiency and purity of the resultant product fall within the range of 70–90 % and 90–99.5 %. Optimal conditions are pH = 10 at 80 °C. To enhance precipitation efficiency, it is necessary to consider the use of gaseous CO₂ or ultrasound, which can increase both the yield and purity of the final product.^{23,24}

4 Conclusion

Lithium-ion batteries are typically processed using both hydro- and pyrometallurgical routes, with pyrometallurgical

methods being more commonly employed in large-scale processing. Pyrometallurgical methods are suitable for mixed lithium-ion battery input. However, it is important to note that pyrometallurgical processing can result in lithium losses to slag without further slag processing. On the other hand, hydrometallurgical approaches exhibit higher selectivity and yield.

In the context of leaching, sulphuric acid and hydrogen peroxide emerge as the most promising reagents. For solution refining, precipitation and solvent extraction are chosen methods. The use of NH₄OH as a reagent for pH adjustment and precipitation of impurities is particularly suitable, considering that impurities in the form of alkali metals, such as Na or K, are not introduced into the solution. However, a notable loss of Li is expected when precipitating impurities (Al, Cu...), prompting exploration of other options for solution refining, such as solvent extraction. Cyanex 272 can be considered the most suitable SX reagent for the recovery of Al (pH = 3), Co and Mn (pH = 5.5–7). Similarly, PC-88A can selectively recover Cu (pH = 3) and Ni (pH = 4.6) from the solution.

Alternative methods such as LIS are not commercially available products. LIS proves suitable for selective lithium extraction only in alkaline solutions, as the extraction process leads to a decrease in solution pH due to the LIS changing H⁺ ions for Li⁺. However, LIS is a prospective method for Li recovery from brines.

Lithium precipitation is the final step in lithium recovery from raw materials and end-of-life products such as lithium-ion batteries. The most suitable conditions for lithium carbonate precipitation are a pH of 10 at 80 °C.

ACKNOWLEDGEMENTS

This work was developed within the framework of the VEGA grant of the Ministry of Education of the Slovak Republic 1/0678/23 and 1/0759/21 with its financial support.

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SAŽETAK

Pregled tehnika recikliranja litij-ionskih baterija

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Ovaj rad predstavlja pregled literature o obradi rabljenih litij-ionskih baterija u industriji i istraživanju. U industrijskoj razini, litij-ionske baterije ponajprije se obrađuju pirometalurškim metodama, što dovodi do nepotpunog iskorištenja litijevih ćelija. S druge strane, hidrometalurški put recikliranja litij-ionskih baterija predstavlja izazove kao što su veliko pražnjenje ili predobrada inertnim plinom, uglavnom zbog opasnosti od eksplozije. Suvremene metode recikliranja litij-ionskih baterija usmjerene su na rafiniranje tekućine za ispiranje metodama ekstrakcije pomoću D2EHPA i Cyanex 272, da bi se dobili Co, Mn i Ni. Konačni produkt, Li, dobiva se taloženjem s Na₂CO₃.

Ključne riječi

Litijeve baterije, recikliranje, ekstrakcija litija, metoda ekstrakcije otapalom, pirometalurška obrada, hidrometalurška obrada

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Pregledni rad
Prispjelo 23. kolovoza 2023.
Prihvaćeno 7. studenoga 2023.