

COMPREHENSIVE APPROACHES TO LITHIUM BATTERY RECYCLING IN THE ERA OF CIRCULAR ECONOMY

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Abstract

This paper aims to provide a comprehensive overview of lithium battery recycling approaches, highlighting its ecological and socio-economic importance. Lithium batteries have become indispensable energy storage devices and have a wide range of applications. The increasing consumption and disposal of lithium batteries have caused significant responses to concerns about environmental sustainability. To solve these challenges, lithium battery recycling has emerged as an adequate solution for efficient use and sustainability. The proliferation of lithium batteries in various industries has led to the need for efficient recycling strategies that minimize environmental impact, conserve resources, and promote circular economy. The recycling approaches include mechanical separation, hydrometallurgical techniques, and pyrometallurgy. Establishing a strong recycling industry encourages the creation of new jobs, promotes technological innovation, and facilitates the transition to a more sustainable energy mechanism. Around that, recycling reduces reliance on limited sources of raw materials, increases energy efficiency, and contributes to a more self-sufficient and resilient economy. Reviewing current trends, challenges, and best practices in battery recycling, this paper contributes to the body of knowledge in this area and encourages further research and development efforts. Successful implementation of efficient and environmentally friendly acceptable lithium battery recycling practices plays a vital role in achieving a sustainable and circular approach to energy storage and resource management.

Key words: lithium battery, recycling, circular economy.

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1. Introduction

Lithium batteries became thus reliable and handy sources of energy, which can be used in a great variety of applications, for example, portable electronics, electric vehicles, renewal energy systems etc. However, an increasing apathy towards the catastrophic overconsumption and the disposal of lithium batteries has sparked unease over environmental sustainability, resource limitations, and hazardous waste management. Lithium batteries have become pervasive in many sectors, thereby raising problems of their efficient recycling, avoiding damage to the environment, saving resources, and contributing to the circular economy requirements. Battery recycling processes include the recovery of several metal materials from batteries through a series of disassembly, sorting, and battery materials processing which consists of lithium, cobalt, nickel, copper etc. The recycling of lithium batteries is a term that encapsulates several areas of science and technology. It includes development and optimization of processes, mechanical separation, hydrometallurgical, pyrometallurgical processes and other direct processes.

This paper aims to provide an overview of lithium battery recycling, highlighting its scientific, environmental and socio-economic importance. By reviewing the current trends, challenges and best practices in battery recycling, this paper contributes to the body of knowledge in the field and encourage further research and development efforts. The successful implementation of efficient and environmentally friendly lithium battery recycling practices plays a significant role in achieving a sustainable and circular approach to energy storage and resource management.

2. Recycling of used lithium batteries

Pyrometallurgy, hydrometallurgy and direct recycling are the three main mechanisms among recycling spent lithium batteries. Pyrometallurgy refers to the treatment of batteries at elevated temperatures with a separation process. Hydrometallurgy involves the use of aqueous solutions to dissolve significant metals from spent lithium batteries and extract the metals through a purification step. Direct recycling means that the structure of the battery's active materials is directly renewed. Currently, pyrometallurgical and hydrometallurgical processes are widely applied in industry, while the direct recycling technique is mostly at the laboratory level. Zhou and colleagues (2020) compared the advantages, disadvantages and challenges of mentioned lithium battery recycling processes. When it comes to the hydrometallurgical process, the main advantages are – a high recovery rate, a product of high purity, low energy consumption, less waste gases and high selectivity, the main disadvantages include – more waste water and a long process, while the main challenges include the treatment of waste water and process optimization. The main advantages of the pyrometallurgical process are simple operation, short flow, no requirements for input categories and sizes, high efficiency, disadvantages include the facts that Li and Mn are not recovered, high energy

consumption, low recovery efficiency, more waste gases and waste gas treatment costs, while the main challenges include reducing energy consumption and emission pollution, reducing the negative impact on the environment. The advantages of the direct recycling process refer to a short recovery path, low energy consumption, environmental friendliness, high recovery rate, the disadvantages include high operational requirements of the equipment, incomplete recovery, while the main challenges include reducing recovery costs, reducing requirements and additional performance optimization products.

3. Pyrometallurgy

Pyrometallurgy uses high temperatures to recover (Lv, 2018) and purify valuable metals through physical and chemical transformations (Liu et al., 2019). Structural changes and phase transitions occur at lower temperatures, and chemical reactions occur at higher temperatures. One of the effective pyrometallurgical options for recovering valuable elements from end-of-life lithium batteries is melting. Due to its high productivity and simplicity, the application of melting prevails in the industry. During this processing, lithium battery substances are heated above their melting points, facilitating the segregation of metal elements in the liquid phase through reductions and preventing the resulting formations from mixing layers (Makuza et al., 2021). Melting takes place in two stages. At the very beginning, the battery materials are heated at low temperatures in order to evaporate the electrolyte and prevent the battery from exploding due to pressure. This is followed by heating at higher temperatures where the battery materials melt.

Another pyrometallurgical process used to recycle used lithium batteries is thermal reduction (baking/calcination). Pretreatments of lithium batteries are usually done in advance to obtain the cathode material for extractive processing (An, 2019). During thermal reduction, active cathode substances are heated with reducing coke, charcoal or C, leaving a C residue and a mixture of intermediate compounds for further processes. A carbonaceous material, waste graphite from reducers is particularly attractive - a low-cost process with high effects (Zhang et al., 2020). Compared to the high-temperature melting process, thermal reduction shows the superiority of high metal recovery rates and low energy requirements. Huang and colleagues (2019) developed a thermal reduction process using spent graphite as a reductant to recover nickel and cobalt from electrode powders of used lithium batteries. Salt roasting aims to convert metal compounds into water-soluble salts by salt roasting. This technique has been shown to be effective for the separation of metals in Li transition metal oxides (Shi et al., 2019). A chlorination roasting approach combined with aqueous leaching has been proposed to recover manganese, cobalt, nickel and lithium from NCM cathode substances (Jian et al., 2021).

4. Hydrometallurgy

After pretreatment, hydrometallurgy is used to regenerate high-value elements from spent lithium batteries such as manganese, lithium, nickel and cobalt. The technology of hydrometallurgical recycling consists of two stages, namely the leaching of lithium batteries and the recovery of high-value elements from the leachate (Chagnes & Pospiech, 2013). The leaching agent commonly used in the rinsing step includes a bacterial solution or an alkaline and acidic (including organic and inorganic acid) solution. High value elements are separated and recovered by various technologies such as solvent extraction, electrolysis, precipitation, ion exchange and so on.

Leaching is most important in the entire hydrometallurgy process to extract high value elements from used lithium batteries. The purpose of the leaching process is to dissolve and convert the valuable elements in the batteries into metal ions in the leaching solutions. The metal ions in the leachate are further separated and recovered by subsequent processes. Acid leaching includes inorganic and organic acid leaching, which is mainly used to dissolve metals in spent lithium batteries. Inorganic acid used for lithium battery leaching includes HCl (Xuan et al., 2019), H₂SO₄ (Fan et al., 2021), HNO₃ (Cognet et al., 2020), and H₃PO₄ (Chen et al., 2019). Reducing agents such as Na₂SO₃ or H₂O₂ can be used to improve the efficiency of the leaching process. Organic acid leaching is a new treatment method used for lithium batteries. The organic acids used include tricarboxylic acid (e.g. citric acid (Refly et al., 2021), dicarboxylic acid (Li et al., 2017), ascorbic acid (Refly et al., 2020), oxalic acid (Zeng et al., 2015) and monocarboxylic acid (Li et al., 2017), acetic acid (Li, 2018) and formic acid (Gao et al., 2017)). Organic acids used for battery leaching have many advantages compared to inorganic acids, such as recyclability, easy degradation, sufficient acidity for leaching, and less secondary emissions.

Alkaline leaching uses interactions between metallic elements and hydroxide ions in an alkaline environment. This method has attracted attention because it selectively dissolves metals and potentially avoids further expensive purification or separation steps. NaOH is commonly used to leach material from batteries for selective removal (Chen et al., 2011).

Bioleaching is another leaching method that uses acids produced by metabolizing microorganisms to leach end-of-life lithium batteries. Bioleaching is an attractive alternative to traditional acid leaching due to lower cost, environmental acceptability, and less demand in industrial implementations. However, this method takes a long time and is subject to pollution, these disadvantages limit its application. Horeh and colleagues (2016) proposed a process for leaching used lithium batteries using a fungus called *Aspergillus niger*.

The fungi used secrete citric acid, oxalic acid, gluconic acid and malic acid during the bioleaching process.

5. Direct recycling

Another way to recycle used lithium batteries is direct recycling. In this processing, the structures of the lithium battery are retained, while the active substances of the battery are directly recovered and collected (Shi et al., 2018). Direct recycling shows many advantages over pyrometallurgy and hydrometallurgy in terms of simplicity of operation, less amount of secondary waste and less hazardous emissions (Ciez & Whitacre, 2019). In addition, after regeneration, the electrodes can be reused directly, which is another advantage of direct recycling. Solid-phase sintering uses solid-phase Li agents as a Li resource for the regeneration of cathode materials. In this process, Li-I is integrated into available sites at elevated temperatures, and is used to replenish the loss of Li. Solid phase sintering is considered the most direct route for lithium recovery (Bai et al., 2020). LiOH (lithium hydroxide), LiNO₃ (lithium nitrate), or Li₂CO₃ (lithium carbonate) can be used as a Li resource in the solid state sintering process.

Hydrothermal relitiation is another potential method of direct recycling of spent lithium-ion batteries. Initially, the excess Li source solution is used to treat spent battery materials, and the added lithium is incorporated into spent cathode structures. It is usually followed by a short annealing treatment, with the aim of reconstructing the crystallinity of the material and improving the restoration of the structure. In the further process, technologies are incorporated to accelerate Li ion transfer including water pulsed plasma discharge (Zhu, 2016) and ultrasound (Zhang et al., 2014).

Stoichiometric additions of lithium are not required because the hydrothermal approach can limit lithium restorations, in contrast to solid-phase rhelithium (Bai et al., 2020). Degraded cathodes with any level of defects could be compensated by the hydrothermal technique. Hydrothermal relithia can work effectively with many cathode chemistries such as NMC 111, NMC 523, NMC 622 and LCO (Sloop et al., 2020).

6. Conclusion

The recycling of lithium batteries is of utmost importance due to the significant growth of their use in various industries. With the ever-increasing demand for lithium batteries and their limited resources, it is crucial to implement efficient recycling techniques to recover valuable materials and reduce environmental impact. The recycling of lithium batteries is in accordance with the principles of the circular economy and contributes to a more sustainable and responsible approach to resource management. It helps to minimize waste generation and associated environmental hazards from improper disposal. The EU Battery Directive sets specific requirements for the collection, treatment and recycling of batteries, including lithium batteries. The directive aims to increase the recycling rate of batteries and reduce the impact of their disposal on the environment. It also places the responsibility on battery manufacturers to take over and properly manage end-of-life batteries. Compliance with EU regulations ensures

that lithium battery recycling is done in a safe and environmentally friendly way. It encourages the development of infrastructure and technologies for efficient battery recycling, promotes the use of recycled materials and encourages a circular economy approach in the battery industry.

REFERENCES

- [1] An, L. (2019). *Recycling of Spent Lithium-Ion Batteries*. Springer.
- [2] Bai, Y., Muralidharan, N., Sun, Y.-K., Passerini, S., Whittingham, S. M., & Belharouak, I. (2020). Energy and environmental aspects in recycling lithium-ion batteries: Concept of battery identity global passport. *Materials Today*, 41, 304–315. <http://dx.doi.org/10.1016/j.mattod.2020.09.001>.
- [3] Chagnes, A., & Pospiech, B. (2013). A brief review on hydrometallurgical technologies for recycling spent lithium-ion batteries. *Journal of Chemical Technology and Biotechnology*, 88(7), 1191–1199. <http://dx.doi.org/10.1002/jctb.4053>.
- [4] Chen, L., Tang, X., Zhang, Y., Li, L., Zeng, Z., & Zhang, Y. (2011). Process for the recovery of cobalt oxalate from spent lithium-ion batteries. *Hydrometallurgy*, 108(1–2), 80–86. <http://dx.doi.org/10.1016/j.hydromet.2011.02.010>.
- [5] Ciez, R. E., & Whitacre, J. F. (2019). Examining different recycling processes for lithium-ion batteries. *Nature Sustainability*, 2(2), 148–156. <https://doi.org/10.1038/s41893-019-0222-5>
- [6] Cognet, M., Condomines, J., Cambedouzou, J., Madhavi, S., Carboni, M., & Meyer, D. (2020). An original recycling method for li-ion batteries through large scale production of metal organic frameworks. *Journal of Hazardous Materials*, 385, 121603. <http://dx.doi.org/10.1016/j.jhazmat.2019.121603>.
- [7] Fan, X., Song, C., Lu, X., Shi, Y., Yang, S., Zheng, F., Huang, Y., Liu, K., Wang, H., & Li, Q. (2021). Separation and recovery of valuable metals from spent lithium-ion batteries via concentrated sulfuric acid leaching and regeneration of LiNi₁/3Co₁/3Mn₁/3O₂. *Journal of Alloys and Compounds*, 863, 158775. <http://dx.doi.org/10.1016/j.jallcom.2021.158775>.
- [8] Gao, W., Zhang, X., Hong, Z., Cao, H., Zhang, Y., & Sun, Z. (2017). Lithium carbonate recovery from cathode scrap of spent lithium-ion battery: A closed-loop process. *Environmental Science and Technology*, 51(3), 1662–1669. <http://dx.doi.org/10.1021/acs.est.6b03320>.



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