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RECYCLING AND DISPOSAL OF LITHIUM-ION BATTERIES

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The number of lithium-ion batteries (LIBs) is steadily increasing to meet the ever-increasing demand for sustainable energy and improved human quality of life. However, this growth is accompanied by a significant amount of LIB waste, which, if not properly disposed of, poses a safety and environmental threat due to its toxic content. In addition, the high consumption of scarce resources of precious metals, which are essential for the production of batteries, is also a serious problem. Due to the serious environmental, resource, safety and recycling issues, the recycling of used LIBs is extremely important to achieve sustainable development. Therefore, this review analyzes the importance and necessity of recycling spent LIBs from different resource and environmental perspectives. Next, a general overview of existing technologies for the recovery and recycling of spent LIBs, such as pyrometallurgy, hydrometallurgy, and mechanical method, is presented. In addition, the advantages and disadvantages of the described methods are analyzed. The factories of the world in which LIB processing is carried out are considered separately.
Keywords: lithium-ion batteries, recycling, disposal, methods of recycling, cobalt

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Introduction

The development of green energy, electric vehicles, and comprehensive recycling of resources are critical aspects on the way to sustainable development, considering that the energy crisis and environmental security are two problems that need immediate solutions (Liang et al., 2021). The transition to renewable energy sources can ensure the end of the fossil fuel era. Electrochemical storage systems, particularly LIBs, are key technologies to successfully implement this transition (Brückner et al., 2020). First introduced to the commercial market in the 1990s, lithium-ion batteries have gained rapid adoption and have become one of the fastest-growing technologies in energy storage.

The main advantages of these batteries are high energy and power density, high reliability and long service life (Hua et al., 2020). By 2030, a total of approximately 10.5 trillion watt-hours of lithium-ion batteries (LIBs) are projected to be produced, of which electric vehicles (EVs) will account for 77%. This is approximately 8.1 trillion watt-hours, and LIB development will continue to grow (Liang et al., 2021). The service life of lithium-ion batteries (LIBs) depends on their application in different areas. Typically, in consumer electronics LIBs have a lifetime of 1-3 years, while for more powerful applications such as energy storage, this lifetime is typically 8-10 years. The volume of spent LIBs is projected to reach 1.08 million tons by 2023, including 200,000 tons of consumer LIBs and 880,000 tons of power LIBs. Consequently, the global LIB recycling market was valued at USD 1.78 billion in 2017 and is expected to reach USD 23.72 billion by 2030, with a CAGR of 22.1% from 2017 to 2030 (Du et al., 2021).

Used LIBs contain significant amounts of valuable metals such as lithium (Li), cobalt (Co), nickel (Ni), manganese (Mn), iron (Fe), copper (Cu) and aluminum (Al). Due to the rapid growth of LIB production, prices for these metals are increasing sharply, in particular for strategic cobalt, the price of which has quadrupled in the last two years - from $22 per kilogram to $81. Therefore, used LIBs should be considered as waste containing valuable strategic materials, considering the principles of sustainable development. Current approaches to the use of old batteries mainly involve disposal and recycling. Recycling used LIBs can not only help alleviate the shortage of raw materials, but also have significant economic benefits. However, recycling must consider economic, environmental, technical and various market perspectives (Lima, et al., 2022, Hua et al., 2020).

With the increasing demand for LIBs, there is an urgent need for their recovery and recycling, particularly due to the potential environmental and health hazards posed by used batteries. However, recycling these metals to match battery quality levels can have significant adverse environmental impacts. Despite the extensive efforts involved in LIBs production, valuable used materials are often discarded into landfills and rendered unusable after just a few years. This contributes to further environmental degradation, as electronic waste, including spent lithium-ion batteries, is one of the most polluting components across all waste types. Hence, battery recycling is not merely a financial imperative but also an environmental necessity. Therefore, the utilization and recycling of spent lithium-ion batteries is receiving more and more attention from many researchers. This article reviews the latest developments in technologies for the utilization and recycling of spent lithium-ion batteries (Du et al., 2021).

Recycling of lithium-ion batteries

One of the main areas of recycling of LIB is the economic value of the metals contained in the cathode active layer. In terms of recyclability and bottom line, currently only cobalt (Co), copper (Cu), iron (Fe), nickel (Ni) and aluminum (Al) are recyclable, while plastic is usually incinerated for recovery energy and lithium (Li), manganese (Mn) and graphite are rarely considered for recycling (Kirti, et al., 2024). LIBs mainly consist of metal shells, a cathode, an anode, an organic electrolyte, and a separator. The cathode mainly contains organic bonding (such as polyvinylidene fluoride), aluminum foil, and cathode materials such as $LiCoO_2$, $LiMn_2O_4$, $LiFePO_4$, and so on. In turn, the anode consists mainly of copper foil, organic bonding, and anode materials (eg, graphite) (Makuza, et al., 2021). The electrolyte is assumed mainly from some organic solvents and lithium salts, such as $LiPF_6$, $LiBF_4$ and so on. As a result, LIB composition can be relatively complex and contain valuable metals, often referred to as a "metal mine" (Costa et al., 2021).

Table 1. Lithium-ion battery components and their weight ratio (Du et al., 2021)

Components	The percent composition in LIBs	Materials
Cathode	15–41	Aluminum current collector with $LiCoO_2$ coating, $LiMn_2O_4$, $LiFePO_4$, $LiNi_xCo_yMn_zO_2$, $LiNiO_2$, etc.
Anode	2–26	The current receiver is copper, coated with graphite
Electrolyte	2–11	$LiPF_6$, $LiBF_4$, $LiAsF_6$, $LiClO_4$, PC, EC, DMC, DEC
Separator	2–14	Polyethylene, polypropylene
Outer shell	12–24	Stainless steel, aluminum, plastic and polymers

Methods of recycling

Due to management issues, it is important that the recycling method is not only environmentally friendly, but also economically beneficial. As a result, the LIBs processing method should have the following characteristics: environmental safety, efficient energy consumption, absence of secondary pollution, high reaction speed, selectivity and high efficiency. Today, the recovery process can be divided into three main methods: pyrometallurgical, hydrometallurgical and mechanical processes. Pyro- and hydrometallurgical processes aim to recover the raw materials used in batteries, including metals, for their further use in the production of new batteries or other applications requiring these metals. Most modern processing methods are based on pyrometallurgy, which is characterized by high energy consumption and emissions of toxic flue gases, which occurs during the production of low-quality metal alloys. Therefore, pyro- and hydrometallurgical methods are evaluated from the point of view of recovery speed, and the mechanical method - from the point of view of battery performance. (Dalini, et al., 2020).

Pyrometallurgy. Pyrometallurgy uses melting in a high-temperature process, typically above 1400 C, which usually involves combustion and subsequent separation to produce a mixed metallic alloy of Co, Cu, Fe and Ni (Kader, et al., 2021), which can be separated mainly by leaching. This is an established technology that is commonly applied to small and large LIBs. Usually, this method does not require preliminary sorting of battery types, but the resulting alloy requires further processing. Pyrometallurgical processes are often associated with high gas emissions and require strict gas filtration standards.

Hydrometallurgy. Hydrometallurgy encompasses leaching, solvent extraction and precipitation using a wide range of reagents. Hydrometallurgical recycling processes are attractive because they can be applied to a range of lithium-ion battery chemistries (Costa, et al., 2021). However, the separation of metals from each other requires additional stages of purification: in addition, unlike pyrometallurgy, sorting is necessary. Aqueous solutions are used to leach metals from the cathode. Water and various acids are used to separate the active substances, which most often form compounds of cobalt, lithium and nickel. In addition, the emission of gases in this process is minimal.

Mechanical/physical process. The mechanical or physical process includes a physical treatment step that aims to reduce the volume of scrap by separating valuable materials and removing the outer casing of the battery. Recovered materials can be subjected to various physical separation processes such as crushing, screening, filtration and magnetic separation. Flotation is sometimes used to separate shells and plastic. Physical separation can be classified into four types: (1) size separation, (2) magnetic separation, (3) density separation, and (4) flotation separation. Machining is a necessary process for each element to be separated. Given the volume of material in the recovery process, mechanical processing is more recommended than manual processing (Dalini, et al., 2020).

Processes, advantages, disadvantages, efficiency are shown in Table 2.

Looking at Table 2, it can be noted that the hydrometallurgical metal recovery method from LIB has several advantages, such as high metal recovery efficiency with high purity, limited energy consumption, and minimal gas emissions. On the other hand, pyrometallurgical processes are used at high temperatures and are usually accompanied by large emissions into the atmosphere.

Table 2. Comparison of different LIB recycling methods (Liu et al., 2019)

Method	Description	Advantages	Disadvantages
Pyrometallurgy	Lithium-ion batteries are recycled at high temperatures	Various chemicals can be processed simultaneously with limited disassembly and remain inexpensive on an industrial scale	The process is very energy intensive with low metal recovery and harmful gas emissions
Hydrometallurgy	It is leached in acids, metals are removed using solution cleaning methods	High metal recovery rate with low energy consumption	Disassembly and pretreatment of lithium-ion batteries is necessary if large volumes of reagents are used
Mechanical	Involves crushing and grinding, which is often the beginning of further recovery methods	No chemicals are needed	An oxygen-free environment or passivation is required before milling

Current recycling capacities in different countries of the world

The main processing facilities are based in China, South Korea, Japan, the European Union and the United States of America (Werner, et al., 2020).

Recycling of lithium-ion batteries in the United States of America

The United States lacks a specific policy addressing the recycling of lithium-ion batteries. Under the Universal Waste Disposal Regulations of the U.S. Environmental Protection Agency, lithium-ion batteries are not classified as hazardous waste and thus are not covered by the Battery Act, despite being labeled as hazardous substances by the Department of Transportation due to their fire risk. While eight states have waste management regulations and battery recycling programs, only three of them explicitly incorporate lithium-ion batteries (Kader, et al., 2021).

Table 3. Current recycling plants in the United States of America (Mrozik et al., 2021; Min et al., 2021)

The company name	Country	Recycling method	Capacity (ton of battery per year)
Inmetco	USA	Pyrometallurgy	6000
Retriev Technologies	USA	Mechanical	4500

Recycling of lithium-ion batteries in the European Union

Within the European Union, Directive 2008/98/EC oversees the storage and treatment of used batteries and accumulators, encompassing both household and electric vehicle batteries. These directives outline responsibilities and procedures for all stakeholders involved in the battery lifecycle. They also categorize batteries based on their usage and establish waste management regulations. Notably, targets for collection and recycling are established to promote material reuse over disposal or incineration. Recycling, in this context, refers to the secondary processing of waste in a production process to recover materials or raw materials, or for alternative purposes, excluding energy recovery (Kader, et al., 2021).

Table 4. Current recycling plants in the European Union (Mrozik et al., 2021; Min et al., 2021)

The company name	Country	Recycling method	Capacity (ton of battery per year)
Accurec Recycling	Germany	Pyrometallurgy, Mechanical	4000
Aubermacher Redux	Germany	Mechanical	1000
Duesenfeld	Germany	Mechanical	3000
Nickelhu¨tte Aue	Germany	Pyrometallurgy, Hydrometallurgy	1000
Euro Dieuze	France	Mechanical, Hydrometallurgy	6000
SNAM	France	Pyrometallurgy, Hydrometallurgy, Mechanical	1500
TES (Recupyl)	France	Mechanical	1000
Sungeel Hi-tech	Hungary	Mechanical	3000
Umicore	Belgium	Pyrometallurgy, Hydrometallurgy	7000

Recycling of lithium-ion batteries in China

China has recently adopted regulatory measures regarding the recycling and reuse of electric batteries, including vehicles, particularly lithium-ion batteries. These measures entered into force in August 2018 and include strict requirements for maintenance, collection, transportation, as well as the use of recycling and recycling technologies. The progressive policy ensures that electric vehicle manufacturers are responsible for the collection, sorting, storage and transport of lithium-ion batteries, and requires that such batteries have a means of identification for further recycling. These provisions also facilitate initial design planning for disassembly and recycling, and provide access to open source information throughout the supply chain (Kader, et al., 2021).

Table 5. Current recycling plants in China (Mrozik et al., 2021; Min et al., 2021)

The company name	Country	Recycling method	Capacity (ton of battery per year)
Bangpu Ni\\&Co High Tech	China	Hydrometallurgy	3600
High Power International	China	Pyrometallurgy, Hydrometallurgy	10000
Huayou Cobalt	China	Hydrometallurgy	60000
Hunan Brunp Recycling Tech	China	Hydrometallurgy	30000
Jiangxi Ganfeng Lithium	China	Hydrometallurgy	5000
Tele Recycle	China	Hydrometallurgy	2000

Recycling of lithium-ion batteries in Japan

Japan does not have any policy regarding the recycling of lithium-ion batteries (Keskes et al., 2021)

Table 6. Current recycling plants in Japan (Mrozik et al., 2021; Min et al., 2021)

The company name	Country	Recycling method	Capacity (ton of battery per year)
Dowa Eco-System Co. Ltd	Japan	Pyrometallurgy, Hydrometallurgy	1000
JX Nippon Mining & Metals Corp	Japan	Pyrometallurgy, Hydrometallurgy	6000
Nippon Recycle Center Corp	Japan	Pyrometallurgy	2000

Recycling of lithium-ion batteries in South Korea

South Korea does not have any policy on recycling lithium-ion batteries (Keskes et al., 2021).

Table 7. Current recycling plants in South Korea (Mrozik, et al., 2021, Min et al., 2021)

The company name	Country	Recycling method	Capacity (ton of battery per year)
KOBAR	South Korea	Hydrometallurgy	1000
SungEel Hitech	South Korea	Hydrometallurgy	8000

Recycling of lithium-ion batteries in Ukraine

In Ukraine, there are still no regulatory documents regulating the procedure for dealing with lithium-ion accumulators that have failed. In 2011, the Lviv State Enterprise "Argentum" was opened in Ukraine, which is engaged in the processing of batteries. Due to the low level of organization of battery collection in Ukraine, recycling volumes are very small. The enterprise was ready to process up to a ton of batteries per day, but only 2.4 tons of batteries were collected in 1.5 years - while almost 10 tons of batteries and accumulators are imported to Ukraine every day (Kaluyan et al., 2021)

Income of recycling methods

A research study Yang et al., 2020 investigated the revenue generated by recycling 10,000 tons of used lithium-ion batteries using various recycling technologies. The simulation utilized the EverBatt software, developed by the Argonne National Laboratory in the USA and released in 2019, incorporating all relevant data into the model. It is worth noting that the prices of cobalt (Co), nickel (Ni), and manganese (Mn) were based on the average prices from the London Metal Exchange spanning from 2016 to 2018, while prices for other materials were sourced from external references linked within the model. The findings of this study are presented in Table 8.

Results, in the table 8 show that the revenues of the mechanical method are higher than those of the pyrometallurgical and hydrometallurgical methods.

Table 8. Income received per kg of processed raw materials

Processing method	Revenue per kg cell recycled ($)			
	Lithium cobalt oxide	Lithium Nickel Manganese Cobalt Oxide	Lithium Nickel Manganese Cobalt Oxide Powder	Lithium iron phosphate
Pyrometallurgy metod	11.66	5.28	3.54	0.83
Hydrometallurgy metod	11.8	5.61	3.74	0.96
Mechanical/physical process	17.8	7.33	5.54	5.09

Environmental pollution by recycling methods

Processing methods do not always meet environmental standards. For example, pyrometallurgy is a highly energy-efficient process that results in greenhouse gas emissions and the generation of toxic gases or hazardous slag that requires proper disposal. In addition, the "black mass" (a mixture of lithium, manganese and cobalt) produced during this process may contain other harmful substances, such as alkyl fluorophosphates, which are a serious health hazard. On the other hand, hydrometallurgy, although it produces less greenhouse gases, requires additional treatment of wastewater to prevent additional pollution, for example, by acid solutions. It is also noted that hydrometallurgical processes can create environmental risks, such as acidification of fresh water and soil. Regarding mechanical processing, there is a lack of sufficient data on its potential environmental impact, as this method is still in the early stages of development (Mrozik et al., 2021).

Disposal of lithium-ion batteries

If recycling facilities are unavailable or if the recovery of materials is not financially viable, batteries are classified as waste and must be disposed of accordingly. The management of used lithium-ion battery disposal follows standards that mandate waste handlers to segregate hazardous materials for proper disposal, while adhering to state and local laws regarding the disposal of other hazardous wastes. Local disposal methods are contingent upon national regulations, the presence of recycling facilities, the effectiveness of collection systems, consumer behaviors, and the dynamics of battery retail markets. Customers typically return small LIB through designated battery collection points or include them in waste electrical and electronic equipment if they are unable to detach them from the device. These batteries are then transported to sorting facilities for processing or disposal. However, due to lack of awareness about the collection system, customers may inadvertently dispose of small spent LIBs in regular waste or landfills (Mrozik, et al., 2021).

Environmental impact of LIBs

Environmental issues arise from both the extraction and processing of lithium sources and the improper disposal of lithium batteries, which contain hazardous materials like nickel and cobalt (Mrozik, et al., 2021). The repercussions of mishandling used lithium-ion batteries can be severe. Discarding lithium-ion batteries in landfills can lead to the corrosion of the aluminum casing by acid-producing microorganisms, resulting in the release of toxic metals such as cobalt, nickel, and manganese into water sources. This contamination poses a significant threat to groundwater and soil. Moreover, the corrosion of lithium-ion battery casings can trigger reactions between the electrolyte and water, releasing harmful gases like hydrogen fluoride into the atmosphere. Furthermore, the disposal of lithium-ion batteries with high lithium content in landfills can cause dangerous explosions and fires due to the lithium reacting with water (Kader, et al., 2021).

Conclusions

One of the key challenges is the need for technological advancements to address the complexity LIBs, which include a variety of chemistries and designs. In addition, there are policy gaps that prevent the development of an efficient and environmentally friendly recycling system. Balancing the economic benefit of recycling and the environmental impact of spent LIBs is also a major challenge, as more complete and accurate assessments of the environmental impact of batteries are needed.

In modern times, developed countries are actively demonstrating successful experience in solving the problem of processing spent sources of electricity by implementing a state policy aimed at the principles of a circular economy. However, there are currently no regulatory acts in Ukraine that would regulate the process of handling spent lithium-ion batteries. Ukraine should develop a regulatory and legal framework that provides for the responsible attitude of both producers and consumers to used LIB. It is necessary to organize the collection and logistics of the used LIB to the places of their further processing and to raise the environmental awareness of citizens, emphasizing that the separate collection of waste is the only way to achieve a clean environment. It is also important to convey information to citizens about the harmful effects on health from the discharge of batteries to landfills together with household waste.

Conflict of interest

The authors state no conflict of interest.

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