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A review on sustainable technologies for lithium ion batteries

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Abstract

The commercial success of Li-ion batteries (LIBs) in the 1990s permanently changed the landscape of energy storage today, yet the batteries' disposal poses an increasing environmental risk. To fulfil the demands of developing technologies like electric vehicles, decarbonized power, and electrochemical energy storage, enormous efforts are being made to create electrode materials, electrolytes, and separators for energy storage devices. Unfortunately, there hasn't been much discussion of the sustainability issues with lithium-ion batteries (LIBs) and the newest rechargeable batteries. Recycling is crucial to the long-term viability of batteries and is influenced by a number of factors, including environmental risks and the worth of the minerals that make up a battery. Consequently, when creating battery systems, recycling should be taken into account. Despite the fact that there are numerous technologies that are addressed for recycling of LIBs, It's still unclear if the most recent cutting-edge LIBs recycling technology qualify as green. Despite the potential for high overall efficiency, duncycling and waste reduction are typically the foundations of industrially applied recycling technology. On the other hand, circular procedures ensuring upcycling of all elements in the direction of zero waste and minimal energy consumption should underpin sustainable recycling of LIBs. This review aims to clarify why the safety issue brought up by lithium batteries needs to be taken into account. It compares and analyses how well various battery chemistries perform. This could provide one a general notion of how environmentally friendly LIBs recycling systems are.

Keywords: Li-ion batteries (LIBs), sustainability, recycling, electrochemical energy, environmental friendly

Introduction

The first rechargeable Lithium Ion Batteries (LIBs) arrived in 1990s, but the lithium development actually started in California in 1912 ^[1]. The market for lithium-ion batteries is growing rapidly, from \$12 billion in 2011 to \$50 billion in 2020, by 2024, estimates predict a rise of \$77 billion USD ^[2]. According to data from the International Energy Agency, production of lithium-ion batteries would expand six fold between 2016 and 2022 ^[3]. As a result, estimations by Winslow *et al.* and other sources suggest that 5.9 Mt of lithium-ion batteries will be produced in 2022 alone ^[4]. This remarkable rise will carry on in the future due in large part to the quick uptake of electric vehicles ^[5]. Due to the depletion of natural resources and the contamination of land and groundwater as a result of the non-recycling of used lithium-ion batteries, this growth has severe negative effects on the environment. To build a circular economy for the lithium-ion battery sector, sustainable recycling technologies must be used. Global recycling rates are currently less than 4% ^[6]. Since a very long time, experts have recognized the need for the creation of so-called green energy. Considering that there are more people on the planet now and that each person uses more energy as a result of rising nominal GDP per capita on a global scale, there will unavoidably be a significant rise in energy consumption in the years to come. Electrochemical energy storage has grown to be a significant problem in this situation.

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In particular, LIBs can tame the intermittent issues with renewable energy by balancing the electricity generated by solar or wind power. Electric vehicles are regarded as clean because they don't emit any gas [7]. Many review articles summarizing the current state of lithium-ion batteries and established solutions in pre-treatment and metallurgical processes have been published over the last few years to address the environmental and social challenges caused by the market's explosive growth [4-6]. The difficulties and restrictions of recycling lithium-ion batteries are also examined and discussed. A review of the legislation and policy governing lithium-ion battery disposal is also conducted. This review's objective is to offer advice on the possibilities and restrictions of sustainable lithium-ion battery recycling technology.

Structure and composition of lithium ion batteries

Lithium-ion batteries are used in a variety of applications, including electric vehicles, electronic devices, and electric

micro mobility. With the largest market share for lithium-ion batteries, electric vehicles give consumers a green choice mode of transport [8]. Although they are beneficial in meeting international climate requirements, electric vehicles are the greatest users of lithium-ion batteries in the world and provide a substantial difficulty in the generation of lithium-ion battery waste [5].

Negative and positive electrodes, electrolytes, and a separator coiled in a variety of shapes and sizes make up lithium-ion battery cells [5, 9-12]. The aluminium current collectors that make up the positive electrode are where the environmentally hazardous and lucrative metal oxides are discovered deposited (cathode during discharge). Several types of cathode compositions—which are summarised in Table 1 below. Graphite is placed on a copper current collector to form the negative electrode (anode during discharge) [4, 14-15].

Table 1: Primary differences in lithium-ion battery cell chemistry [13]

Cathode substance	Voltage of The nominal cell (V)	Specific Energy (Wh/Kg)	Life span (Cycles)
Lithium cobalt oxide	3.6	150-200	500-1000
Lithium iron phosphate	3.2-3.3	90-120	1000-2000
Lithium manganese oxide	3.7	100-150	300-700
Lithium nickel cobalt aluminium oxide	3.6	200-260	500
Lithium nickel manganese cobalt oxide	3.6-3.7	150-220	1000-2000

Lithium salt is dissolved in an organic solvent, often a combination of ethylene carbonate and dimethyl carbonate, to create the normally liquid electrolyte [15]. High electrochemical stability and operability across a larger voltage range are made possible by the solvent's strong permittivity and low viscosity [16, 17]. The separator is a porous, semi-permeable polypropylene membrane that gives the electrolyte a channel for ionic conduction and inhibits short-circuiting [15, 16, 18]. The casing makes up the remaining portion of the cell.

Policies and rules for disposing of lithium-ion batteries

The introduction of environmental rules is a key motivator for the growth of lithium-ion recycling. These regulations will aid in directing the lithium-ion battery market towards a circular economy through the use of environmentally friendly recycling techniques [19].

In India, The regulations are based on the Extended Producer Responsibility (EPR) principle, according to which battery producers (including importers) are accountable for the collection, recycling, and refurbishment of used batteries as well as the utilization of recovered materials from wastes in the production of new batteries. EPR prohibits the burning of used batteries and their disposal in landfills and mandates that all used batteries be collected and transported for recycling or refurbishment. In order to fulfil their EPR obligations, producers can either organize for their own collection, recycling, or refurbishment of used batteries or grant permission to any other organization.

The Resource Management Act and Waste Minimization Act of New Zealand, which also encourage a reduction in the amount of rubbish created and disposed of, contain definitions of sustainable management [20].

The United States, China, and the European Union have all developed policies for the environmentally friendly recycling of lithium ion batteries.

Recycling methods for lithium-ion batteries

By implementing sustainable recycling technologies and creating a circular economy where new lithium-ion batteries can be produced from recycled components, the previously discussed environmental effects of lithium-ion batteries can be significantly minimized. Reduce, reuse, and recycle are the three R's that lithium-ion battery recycling must follow. Lithium-ion batteries' number can be reduced by using repurposed alternatives, such as enhancing renewable infrastructure to handle peak loads [15]. Lithium-ion batteries that have been reused for their intended usage fall under the definition of reuse. This includes direct recycling processes, which recover cathode materials as reusable cathode mixes rather than as separate metals, hence minimising the requirement for downstream processing. The recycling of lithium-ion batteries is described as the recovery of material and extraction of metal elements. Presently, there are four basic categories of recovery procedures: pre-treatment, pyrometallurgy, hydrometallurgy, and biometallurgy processes. Often, recovery methods combine these processes. Disassembly is a developing field that intends to use automation in the pre-treatment stage; physical approaches involve physically separating the battery components using techniques like crushing and heating. Separation of metal ions from the electrode materials is a part of the chemical processes. The field of biological techniques is developing quickly [21].

Dismantling and Pre-Treatment

For subsequent steps, lithium-ion battery sorting with intelligence is essential [15]. In order to put this into practice, lithium-ion batteries need to be classified by chemistry

before being treated. Similar to this, different lithium-ion battery designs from manufacturer's present significant challenges for pre-treatment before recycling [5]. There is currently no industry standardization for lithium-ion batteries, and this is anticipated to continue to be a problem in the future.

Use of salt-saturated solutions, such as immersion in sodium chloride solution, is a frequent technique to discharge cells before cell opening [22], despite the fact that this causes structural damage to the cells before complete discharge [23]. Although this process takes less time than other ways [24], if the cell casing is broken, enabling the reaction of electrolyte in water, hydrogen fluoride may be released. Liquid nitrogen cooling under passivation is also a method of disassembly that minimizes the reactivity of lithium-ions and lessens the risk of fires, explosions, and poisonous emissions [15]. There has been electrolyte recovery from lithium-ion batteries both on a lab and an industrial scale, established. It frequently occurs in despite electrolyte recovery being economical, other organic compounds and the electrolyte are wasted during recycling procedures. Due to its high boiling point in comparison to water, one technique of recovering electrolyte involves recovering organic solvent through distillation [25]. In order to stop the release of harmful fluoride-based gases, the electrolyte and separator are frequently submerged in an alkaline solution after being disassembled [26, 27].

Electrode materials are found in pre-treated lithium-ion battery powder; these materials can be floated or magnetically separated to separate the metals from the graphite [15]. Separating the solid electrolyte interface layer, binder, and additives from the graphite is the main obstacle encountered in graphite recovery [15, 28, 29]. Pre-treatment methods like mechanical separation use processing methods to concentrate the metallic fraction in accordance with characteristics including density, conductivity, and magnetic behavior [30]. Not all the chemicals in lithium-ion batteries can be separated mechanically, which is a drawback [19].

Pyrometallurgy

In pyrometallurgy, metals and other materials are recovered by high-temperature processing. Plastic, electrolyte, and other components from used lithium-ion batteries are broken down in high-temperature, oxygen-rich furnaces at temperatures above 1200 °C [31], resulting in the formation of metal oxide. Before being employed in the production of lithium-ion batteries, the metal oxide needs to go through additional processing. While being commonly employed in industry, this process produces hazardous flue gas. It needs downstream processing to comply with environmental laws. Pyrometallurgy, on the other hand, is flexible, straightforward, and has a high processing capacity. Moreover, it can enable a mixed lithium-ion battery feed with the least amount of pre-treatment required. It has been established that, in comparison to the extraction of raw materials, pyrometallurgical recycling produces net increases in greenhouse gas emissions and energy consumption, which are mostly caused by the combustion of nonmetallic-based lithium-ion battery componentry [15].

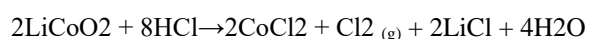
Insoluble organic additives and adhesives can be separated with the use of a lower temperature treatment. Heating the mixture to about 150 °C in a regulated environment. In order to remove lithium-ion battery contaminants before further processing, low-temperature treatment is attractive.

Thermal treatment has, however, been connected to substantial dioxin emissions and chloride compounds, much like pyrometallurgical procedures. Hence, downstream air purification is important [19].

Hydrometallurgy

Using a wide variety of chemicals, hydrometallurgy includes leaching, solvent extraction, and precipitation. In comparison to pyrometallurgy and biometallurgy methods, hydrometallurgy can produce metals with higher purity while using less energy and emitting fewer gases [32, 33].

An acidic solution can leach the remaining metallic dust and mechanical pre-treatment residues, transferring the metals into the bulk aqueous solution [34]. Among of the evaluated inorganic acids, hydrochloric acid was shown to be the most effective leaching agent by Zhang *et al.* [35]. The proposed process for the LCO residue reaction with hydrochloric acid is presented in –



Due to its low energy consumption and flexibility to handle a variety of cathode chemistries while allowing the metals to be recovered in high purity, hydrometallurgical acid leaching seems promising for large-scale applications [15].

Following the leaching procedure, each metal is typically selectively removed from leachate into an organic phase using solvent extraction, and then recovered by precipitation, crystallization, or reduction [36, 37]. Direct precipitation of cobalt oxide from an aqueous leachate was made achievable employing an electrochemical-driven approach in addition to the more traditional recovery techniques [38]. Whilst many studies focus on separate separation of the metals via solvent extraction [39], others have examined co-extraction followed by mixed metal oxide precipitation as a means to directly replenish the cathode material [40].

Others have looked into using deep eutectic solvents to remove Li and Co from used lithium-ion batteries, and they've reported high extraction rates and straightforward post-leaching precipitation procedures [41-43].

Biometallurgy

The removal of metals from used lithium-ion batteries via biometallurgy is a successful process. In order to create soluble metals from insoluble trash, the metabolites excrete organic acids [44]. In biometallurgy, fungi are utilised instead of bacteria because they can grow across a larger pH range and leach more quickly. In their research, Bahaloo-Horeh *et al.* used mixed lithium-ion battery trash and biometallurgy processes. *Aspergillus Niger*, a fungus known to secrete citric, gluconic, malic, and oxalic acids, was used to accomplish this [45]. These procedures need lengthy incubation times (up to two weeks), high liquid-solid ratios, and are still being tested on a wide scale [15].

Existing procedures

Lithium-ion batteries are being recycled on an international basis. In addition to being mostly a result of environmental constraints, this can also be linked to the financial advantages of recovering the metallic lithium-ion battery components. Numerous businesses devised strategies to manage the inflow of lithium-ion batteries entering the waste stream at the end of their useful lives. Currently, a

variety of businesses from numerous nations are engaged in recycling lithium-ion batteries on a variety of scales.

Although this list is not exhaustive, the majority of these businesses continue to use old-fashioned, energy-consuming techniques like pyrometallurgy^[16]. Recent efforts to lessen this reliance are the result of growing public awareness of the damaging environmental effects of such recycling processes^[15]. It should be noted that for the data gathered, it is unclear whether the reported processing capacity include other types of batteries, ores, or industrial trash.

An important step towards a circular lithium-ion battery economy is the development and integration of the 3-R model. To depart from the prevalent, environmentally harmful pyrometallurgy techniques of today, however, a change in attitude is required.

Conclusions

Due to the increasing popularity of electric vehicles, the lithium-ion battery business is expanding quickly. While using electric vehicles has numerous advantages for the environment, the creation of lithium-ion batteries, landfilling, and even existing recycling techniques have a major negative impact on the environment.

Due to the lithium-ion battery market's explosive growth, there will also be a significant increase of lithium-ion batteries that are nearing the end of their useful lives and need to be controlled. New sustainable recycling methods must be adopted in order to create a circular economy for the lithium-ion battery business because the rate at which new lithium-ion batteries are produced far outpaces present recycling efforts.

The bulk of recycling techniques used today involve pyrometallurgy, which produces low-quality metal alloys while consuming a lot of energy and emitting hazardous flue gases.

Hydrometallurgy methods, in contrast, present a competitive alternative with encouraging developments in lab-scale research, indicating there is a road towards later industrial-scale recycling procedures that can rival lithium-ion battery manufacturing. These procedures must be flexible enough to adapt as new mixed cathode chemistries emerge, like ones that use less or no cobalt at all. According to the study discussed in this article, a combination of pre-treatment and hydrometallurgical processes has the potential to recover important metals, minimize the energy needed for recycling, and ultimately lessen the dependency on raw material extraction.

In addition to the recovery of the electrolyte and graphite from depleted lithium-ion batteries, effective solvent recovery of leached metals from cathode scrap was noted as a restriction in the literature. Other advantages of using automation in the pretreatment process include lower costs and better material recovery effectiveness.

It will be necessary to apply political pressure to the recycling of lithium-ion batteries through financial incentives, statutory regulations, and public awareness campaigns.

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