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Review

The Lithium-Ion Battery Recycling Process from a Circular Economy Perspective—A Review and Future Directions

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Abstract: Ever since the introduction of lithium-ion batteries (LIBs) in the 1970s, their demand has increased exponentially with their applications in electric vehicles, smartphones, and energy storage systems. To cope with the increase in demand and the ensuing environmental effects of excessive mining activities and waste production, it becomes crucial to explore ways of manufacturing LIBs from the resources that have already been extracted from nature. It is possible by promoting the re-usage, refurbishing, and recycling of the batteries and their constituent components, rethinking the fundamental design of devices using these batteries, and introducing the circular economy model in the battery industry. This paper through a literature review provides the current state of CE adoption in the lithium-ion battery industry. The review suggests that the focus is mostly on recycling at this moment in the battery industry, and a further understanding of the process is needed to better adapt to other CE practices such as reuse, remanufacture, refurbishment, etc. The paper also provides the steps involved in the recycling process and, through secondary case studies, shows how some of the industries are currently approaching battery recycling. Thus, this paper, through review and secondary cases, helps us to understand the current state of LIB recycling and CE adoption.

Keywords: lithium-ion batteries; circular economy; battery recycling; battery reuse; sustainability; environment

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1. Introduction

Lithium is a highly versatile element with various applications in industry, such as ceramics, batteries, and lubricants [1] (However, lithium-ion batteries are one of its most important applications, owing to their use in electric vehicles, smartphones, and other devices. Considering the policies of various countries in Europe, Asia-Pacific, and other parts of the world favoring the sale of electric vehicles (EVs), it is estimated that there will be an EV sales penetration of 30% for private cars and 70% for commercial vehicles by 2030, which might take the global demand for lithium-ion batteries to 2035 GWh [2,3]. This has led to an increase in the requirement for raw materials to manufacture LIBs. However, we might not be able to meet the demand for many critical minerals such as cobalt, whose market will grow to up to 1.6 times today's capacity in the next decade [4].

Furthermore, there is a considerable gap in the production and recycling of LIBs. Globally, less than 5% of LIBs are recycled, while in India alone, over 50,000 tons of LIB waste is generated annually [5,6]. Some unrecycled batteries may be used in second-life applications such as energy storage systems. Still, most unrecycled batteries generally end up in landfills, contaminating the land and groundwater reserves. This wastage of batteries also indirectly contributes to an increasing dependency on virgin raw materials, further harming the environment. One of the most effective ways of tackling the lack of minerals and waste generation is introducing into the LIB market the circular economy (CE) model, which believes in repairing, reusing, and recycling used and damaged products [7]. Here, the concept of a CE would mainly be applied to improving the capacity and volume of batteries recycled. Doing this could help reduce the load on the environment and promote greener production methods.

Generally, the recycling process consists of four stages—pretreatment, pyrometallurgy, mechanical processing, and hydrometallurgy. Recycling processes may or may not require all these stages. The recycling process depends on the type of batteries and the technology to be used for recycling.

While battery recycling is not a new concept, an in-depth investigation on this topic remains unexplored and under-researched, especially with regard to focusing on CE adoption and the application of its practices [8,9]. The extant literature also explores neither lithium-ion battery (LIB) recycling processes nor the companies that are actually working on the recycling process of LIBs [6,10]. We use secondary case studies to highlight the process of LIB recycling and also explore CE adoption possibilities in the battery industry. The concept of CE, the recycling process, and its applications in the industry are explained in the subsequent sections of the paper.

2. Theoretical Background

2.1. Circular Economy

The linear economy model, characterized by the "take, make, and waste" [11] plan, has dominated the industrial revolution for over 150 years [12]. In this system, raw materials are collected, converted into products, and then used until they are disposed of, owing to their design. However, this kind of model has been deemed unsustainable due to its impact on the environment and the consumption of natural resources [7,13]. The search for a model better than the existing linear model in resource consumption has led businesses to find ways of reusing products and reducing waste outputs.

In contrast to the linear economy model, the CE model relies more on leasing, sharing, reusing, recycling, repairing, and refurbishing existing products and raw materials as far as possible. According to the Ellen McArthur Foundation, the definition of a circular economy is "looking beyond the contemporary take-make-dispose extractive industrial model; a circular economy intends to redefine growth, concentrating on positive society-wide benefits. It entails gradually decoupling economic activity from consuming limited resources and scheming waste out of the system. Reinforced by a transition to renewable energy sources, the circular model builds economic, natural, and social capital. It is based on three principles, i.e., project out waste and pollution; keep products and materials in use; and regenerate natural systems" [14]. Thus, it helps to reduce the output of waste and pollutants and the load on natural resources. The CE model aims to increase the periodic productivity of equipment and other manufactured goods [6] and use waste material and energy as inputs for other processes [15]. Implementing a CE model (Figure 1), eliminating waste and pollution, circulating products and materials, and regenerating nature [16] are of the utmost importance.

With a growing population, the demand for raw materials has increased manyfold. However, the supply of such critical raw materials is limited. Hence, to make the most out of the already extracted minerals, the introduction of CE principles is essential to promote the cradle-to-cradle model instead of the cradle-to-grave model [13]. Implementing CE can help in the following ways:

1. It minimizes the harm caused to the environment by waste disposal and the extraction of raw materials, as it aims at improving manufacturing processes to make them more sustainable by promoting the usage of biodegradable materials and reusing raw materials and disposed products [17].
2. An increasing population would lead to an increase in consumption. Implementing CE would reduce the pressure on primary raw materials in the future and promote the reuse and recycling of such resources [12].

3. It can help improve economic growth, since overall production costs reduce as the raw materials are more easily obtained and reused. The quality of the products thus developed will also be superior since they will be more durable and easily reusable. It will also ensure an increase in the efficiency of raw materials and energy usage [18,19].
4. It can also improve the quality of human and animal health and preserve biodiversity through the judicious usage of natural resources, which reduces emissions and pollution [20].

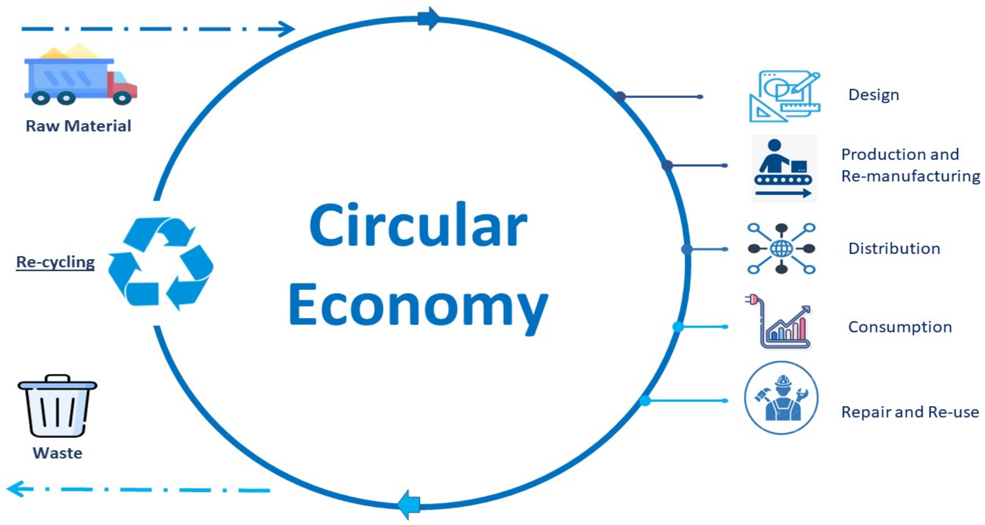

Figure 1. Circular Economy Model.

2.2. Batteries

The most basic definition of a battery is that it is a device that converts chemical energy stored between its electrodes to electrical energy by a redox (reduction–oxidation) reaction [20]. A battery is a combination of two or more cells connected in series or parallel, storing energy in the form of chemical energy until it is connected to a live circuit. Batteries are classified as:

1. Primary Batteries—These batteries are non-rechargeable, i.e., disposable (reflecting the linear economy model). Different varieties of such batteries are alkaline, mercury, and silver–zinc. They are primarily used in portable electronic devices such as flashlights.
2. Secondary Batteries—These are rechargeable and thus can be used multiple times before discarding (reflecting the circular economy model). Most of these batteries have a fixed number of charge–discharge cycles, after which their capacity decreases, making them unfit for usage. These include lead–acid, lithium-ion, nickel–cadmium, and nickel–metal hydride batteries. These batteries can find applications in mobile phones, laptops, and electric vehicles [21]. This paper mainly focuses on the recycling process of lithium-ion secondary batteries and the industrial application of that process in various companies.

2.2.1. Lead–Acid Batteries

Lead–acid batteries, invented by a French physicist, Gaston Plante, in 1859, were the first of their kind of secondary batteries in the world. The battery consists of two electrodes: the negative terminal, made of porous lead, and the positive terminal, made of lead oxide, and dilute sulphuric acid as the electrolyte. During discharging, both electrodes are converted to lead sulphate. Despite having a relatively lower energy density than other types of batteries, they have a high power-to-weight ratio. This benefit, along with their low cost, makes them ideal for use in automobiles such as cars, boats, and scooters and for backup power supply in places such as hospitals and phone towers [22]. They are also rugged and can endure excessive usage. However, lead–acid batteries are not environmentally friendly since lead is a toxic material. Lead recycling is a well-established industry and manages to recycle a vast amount of lead, irrespective of the fact that over 40,000 metric tons of lead end up in landfills every year [23].

2.2.2. Nickel–Cadmium Batteries

Another type of rechargeable battery is the nickel–cadmium battery. The battery uses nickel oxide hydroxide as an anode, metallic cadmium as the cathode, and potassium hydroxide as the electrolyte. Nickel–cadmium batteries were invented in 1899 by Waldemar Jungner, a Swedish engineer. Due to their terminal voltage of 1.2 V and steady discharge voltage, they quickly replaced primary zinc–carbon and alkaline batteries, which have a terminal voltage of 1.5 V, in many applications. These batteries are mainly used in portable electronic devices such as wristwatches and flashlights [10,24,25]. They have replaced primary batteries in devices such as cordless phones and toys. Despite being more robust and having a much higher energy density than lead–acid batteries, they are less energy dense than lithium-ion batteries. They also suffer from the memory effect. When this battery is charged and discharged to the same state of charge many times, it tends to remember the point to which it was discharged before recharging. If the battery is discharged beyond this point, it experiences a sharp voltage drop as if it is fully discharged [24,26].

2.2.3. Nickel–Metal Hydride Batteries

In recent years, nickel–metal hydride batteries have emerged as one of the better alternatives to nickel–cadmium batteries due to their low-level of toxicity. Similarly, to other batteries, they have a positive and negative terminal, but the negative terminal uses hydrogen-absorbing alloy as an electrode. The battery also uses aqueous potassium hydroxide as the electrolyte, which separates both positive and negative terminals [27]. These batteries are also used in place of alkaline AA batteries, since they have a similar voltage of 1.2 V and a much higher energy density. They were also being used in hybrid electric vehicles, but recently they have been replaced almost entirely by lithium-ion batteries, mainly because they have a much higher energy density than NiMH batteries [25,28].

2.2.4. Lithium-Ion Batteries

Lithium-ion batteries are a type of rechargeable battery in which positively charged lithium ions travel from the anode to the cathode during discharge and back to the anode while charging. The positively charged electrode is made of an intercalated lithium compound, while the negatively charged electrode is made of graphite [29,30].

Research on lithium-ion batteries had already started in the early 1970s, but the prototype lithium-ion battery was not developed until 1985. Finally, the first commercially developed lithium-ion battery was produced by a team of Sony and Asahi Kasei scientists led by Yoshio Nishi in 1991 [1,31]. Ever since, the influence of the lithium-ion battery on the technological and economic development of the world has been noteworthy. Its applications expanded to automobiles, electronics, and electrical appliances, and to glass and ceramics industries [8].

From the above discussion (refer to Tables 1 and 2) about different types of secondary batteries, we understand that lead–acid batteries are harmful to the environment due to toxic lead compounds. Nickel–cadmium batteries cannot be used for the long term because of the memory effect. Nickel–metal hydride batteries do not have an energy density as high as lithium-ion batteries; hence, they cannot be used for heavy applications. Therefore, we see that lithium-ion batteries (LIB), not having a memory effect and being the most energy dense, have major scope for reusing and recycling. Thus, this paper will focus on different methods of recycling lithium-ion batteries and their second-life applications.

Table 1. Advantages and disadvantages of lithium-ion batteries.

Advantages	Disadvantages
• The high energy density of approximately 100–265 Wh/kg or 250–670 Wh/L • No memory effects • The low self-discharge rate of approximately 1.5–2% per month • Low maintenance cost • The high cell voltage of 3.6 V during operation • They always remain easy to charge and are mainly unaffected by a reduced total amount of charge over time	• Possibility of overheating and damage at high voltages (flammable in extreme circumstances) • High cost (approximately 40% more than Ni–Cd) (Lander et al., 2021 [32]) • Costs are kept high due to the scarcity of cobalt and nickel • Ageing leads to a decreased capacity over time and charge–discharge cycles (Li et al., 2021 [33]) • They perform poorly at very low temperatures (below 0 °C)

Table 2. Comparison of batteries.

Battery → / **Parameters ↓**	**Lead–Acid**	**Nickel–Cadmium**	**Nickel–Metal Hydride**	**Lithium-Ion**
Energy density	Low	Medium	High	Very high
Overcharge	High	Medium	Low	Low
Voltage	2 V	1.2 V	1.2 V	3.6 V
Toxicity	High	High	Low	Low
Cost	Low	Medium	High	High

2.3. Circular Economy in the Battery Market

A BCG study showed that, as of 2020, there were more than 32 million electric vehicles in use, of which 8 million were fully electric and plug-in hybrid electric vehicles, and the remaining 24 million were hybrid electric and medium hybrid electric vehicles. It also predicted that over 300 million electric vehicles would be on the roads by 2030 [7,32]. In addition, the governments of various countries around the world have been promoting the use of electric vehicles to tackle climate change. This trend suggests that the required number of LIBs will only increase in the following years. Even though using electric vehicles is less harmful to the environment than fossil-fuel-based vehicles, lithium mining is not mainly green [3,34]. The extraction of 1 ton of lithium takes roughly 1900 tons of water, and it is also predicted that the current supply of lithium will not be able to fulfil the demand in 2023–2025 unless the batteries are recycled at a 90% efficiency [35,36].

This increase in the requirement for LIBs would create an immense load on the environment owing to the mining of new lithium. Hence, it is of the utmost importance that a circular economy model is introduced in the battery industry, promoting the reuse and recycling of the batteries already in use and reducing the load on the environment [33,37]. The only way forward is the effective end-of-life treatment of LIBs, which mainly includes recycling or a second-life application.

1. Recycling LIBs involves recovering valuable metals from the battery, such as lithium, cobalt, and manganese. This task is executed by companies that specialize in recycling such batteries. The metals thus recovered are then sold to other companies which manufacture batteries.
2. Second-life applications involve repurposing the battery and usage, usually in stationary applications such as static energy storage systems. A specialized company carries out this repurposing.

These two alternatives are better than the disposal of batteries after use in landfills, since there is no recovery of valuable metals. Introducing the circular economy model in the battery industry will ensure that the valuable metals are used up to their capacity before disposal, which can help reduce toxic waste and load on the environment [8,9].

3. Methodology and Workflow

This review has used as a guideline the works of [12] for the overview of a circular economy, and [18–20] for methods of recycling LIBs. It focuses on the papers published between 2010 and 2022 to explore the different ways and stages in the recycling process of LIBs and the techniques followed by a number of well-established companies. The references have been collected from databases such as ScienceDirect, ResearchGate, and Google Scholar since they provide comprehensive and peer-reviewed publications. The search consisted of 3 primary keywords or phrases: 'Lithium-Ion battery reuse,' 'Lithium-Ion Circular Economy,' and 'Critical Mineral Extraction from batteries.' It included research papers, review papers, textbooks, websites, and presentations. The initial search yielded 85 research articles, after which an elimination process, as shown in Figure 2, was adopted to streamline the database and content for the review. Finally, the scope of the paper was narrowed down to employing a circular economy in the batteries industry by recycling lithium-ion batteries, and it contained 16 articles. Table 3 provides the name of the journals in which those 16 articles were published.

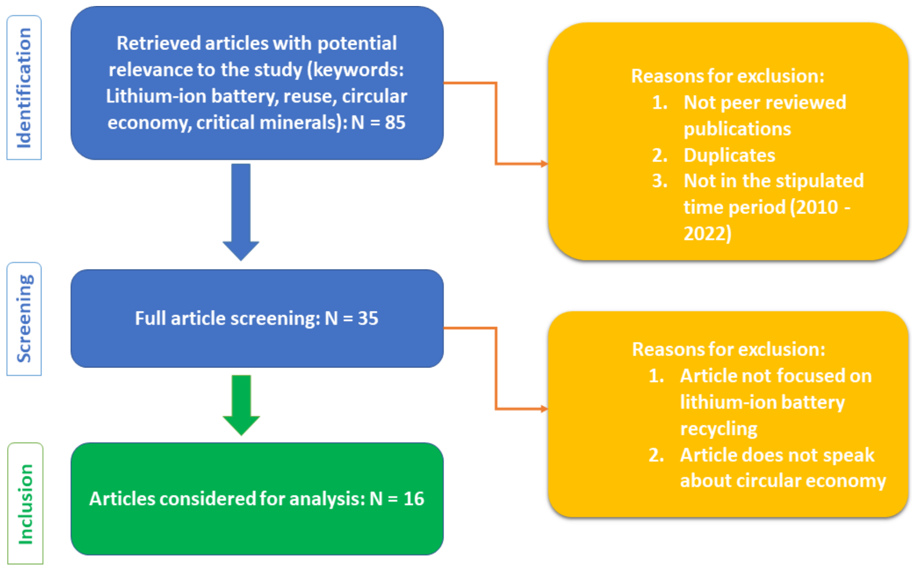

Figure 2. Flowchart for selecting articles.

Table 3. Number of papers in academic journals.

Journal Title	Number of Papers
The International Journal of Life Cycle Assessment	1
Resources, Conservation and Recycling	1
Batteries	1
Journal of Environmental Management	1
Journal of Industrial Ecology	1
Advanced Energy Materials	1
Journal of Energy Storage	1
Mineral Processing and Extractive Metallurgy Review	1
Journal of Hazardous Materials	1
Separation and Purification Technology	1
Critical Reviews in Environmental Science and Technology	1
International Journal of Production Research	1
Energy Policy	1
Journal of Cleaner Production	1
Sustainable Materials and Technologies	1
Waste Management	1
Total	16

Primarily, this paper discusses the incentive and motivation behind recycling LIBs. Although there is a wide variety of methods of recycling LIBs employed by different companies, we have chosen first to explain the broad stages of recycling for this paper. These stages are (1) sorting, discharge, and disassembly, (2) pyrometallurgy, (3) mechanical processing, and (4) hydrometallurgy. After giving an overview of these methods, we move on to a number of well-established recycling processes used in the industry.

4. Discussion

4.1. The Circular Economy Perspective

As mentioned before, the main goal of the circular economy model is to keep all products at their highest level of utility for the maximum amount of time. This aim is fulfilled by the seven Rs of the circular economy. These seven Rs of the circular economy and their current applications in the LIB industry are explained below [7,15,20,38]:

I. Rethink: Involves rethinking and reformulating the various stages of the processes and making them lean, keeping in mind the impact LIBs have on the environment (the generation of waste and pollution). To extract minerals such as lithium and cobalt for LIBs, a large volume of water and energy is required [15,39], which eventually becomes one of the many downsides of producing LIBs. Rethinking these steps and optimizing the use of natural resources to extract these minerals is essential for making the production process greener [18].

II. Reduce: Involves reducing both the dependency on a particular product and the usage of natural resources to manufacture the product. However, as mentioned earlier, the use of LIBs will only increase in the future and reducing their usage would not be viable. However, reducing the depletion of natural resources used to manufacture these batteries would make their extensive usage sustainable [8,23].

III. Repair: This involves repairing partially damaged products instead of throwing them away and replacing them with a new product. It promotes the user to use the product to its maximum potential and reduces waste outflux. However, in most cases, if a LIB is damaged in a product, the battery is replaced, e.g., in smartphones. Here, a slight change in the design can help immensely to reduce the wastage of products still in working condition [30,35].

IV. Refurbish: Involves recovery and renewal of used and discarded goods to be sold again instead of making new products and wasting the previously used material. Refurbishing in the LIB industry is mainly functional in the smartphone sector, which means that the phone had a previous owner and is being resold after minor fixes, such as rectifying physical damage. These refurbished phones are offered at cheaper rates and hence are preferred by many buyers. They are also environment-friendly, since they reduce the number of new units introduced into the cycle, promoting the total usage of the products' potential [40,41].

V. Recover: Includes salvaging energy from non-recyclable waste by processes such as combustion. However, applying this to the LIB industry would end up doing more harm than good to the environment, as most of their components are toxic. Recycling and reusing, on the other hand, are more viable options [37].

VI. Reuse: As the name suggests, it involves reusing the product until it has been used to its full potential. It has been adopted in the LIB market as a second-life application. It also includes LIBs that were a part of products that have reached their end-of-life. LIBs find applications in devices such as electrical storage systems for residential and commercial power [10,39].

VII. Recycle: Includes disassembly of the product, separating the valuable components, and concentrating and preparing them for use in a new product. It is one of the most widespread activities undertaken in the LIB industry, and the process followed is explained in detail in this paper [17]. After searching the literature and other research regarding the application of CE principles to the LIB industry, it was found that in the last decade, most of the studies focused only on the re-usage and recycling of LIBs.

This is due to the sudden rise in the usage of LIBs in burgeoning electric vehicles and smartphones. Hence, this paper concentrates on the various steps involved in the recycling process through examples from different companies [4,25,42].

4.2. Motivation behind Recycling of Lithium-Ion Batteries

The volume of LIBs produced is set to surge in the coming years, and the global cumulative capacity of LIBs is predicted to increase over five times in the current decade [43]. It brings a surge in the waste flux generated, which has prompted an increase in research in the area, as shown in Figure 3. Mentioned below are the various incentives and benefits of recycling LIBs.

I. Economic Benefits: One of the most important benefits is that the material acquired from the recycling of used batteries can be utilized to manufacture more batteries. This is particularly beneficial because raw materials account for over half the cost of a LIB [44]. Many LIBs have a greater concentration of elements such as lithium and manganese than their natural ore, so their recycling will be cheaper and more beneficial due to the higher yield of critical components. It has also been observed that recycled lithium is more porous, which aids in faster charging [45].

II. Environmental Benefits: The main advantage of recycling LIBs is that it reduces the amount of waste produced, reducing the amount of electronic waste in landfills [18]. It not only ensures that all the critical minerals already in use are utilized to their full potential but also reduces the risk of these elements leaking into the environment, contaminating the soil and groundwater and harming the ecosystem [43,44].

III. Health Benefits: Lithium is toxic, and its infiltration into groundwater can adversely affect the ecosystem that thrives in it and the human population that consumes it. Recycling will reduce the number of batteries that end up in landfills and thus reduce the chances of groundwater contamination [4].

IV. Humanitarian Benefits: There are many humanitarian issues linked with mining. Over 50% of the cobalt used in these batteries comes from the Congo, where the fundamental human rights of the mine workers are frequently violated [32,39]. Recycling batteries and making the most out of the cobalt and lithium already in use will reduce the dependency on new minerals from mines, hence improving the humanitarian state of the laborers [1,3].

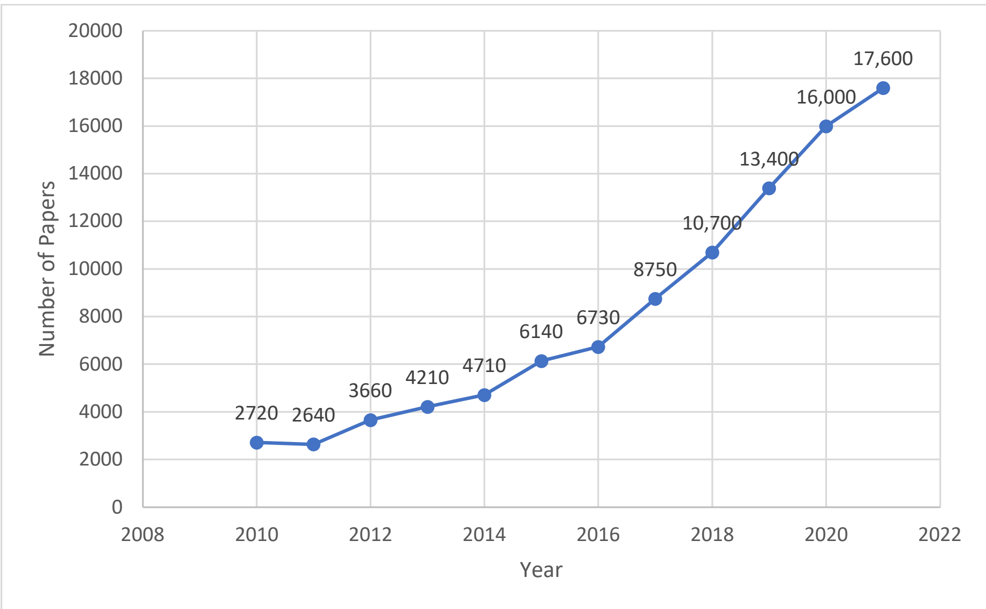

Figure 3. Papers about lithium-ion battery recycling—volume published per year.

4.3. The Recycling Process

Figure 4 illustrates the lithium-ion battery recycling process.

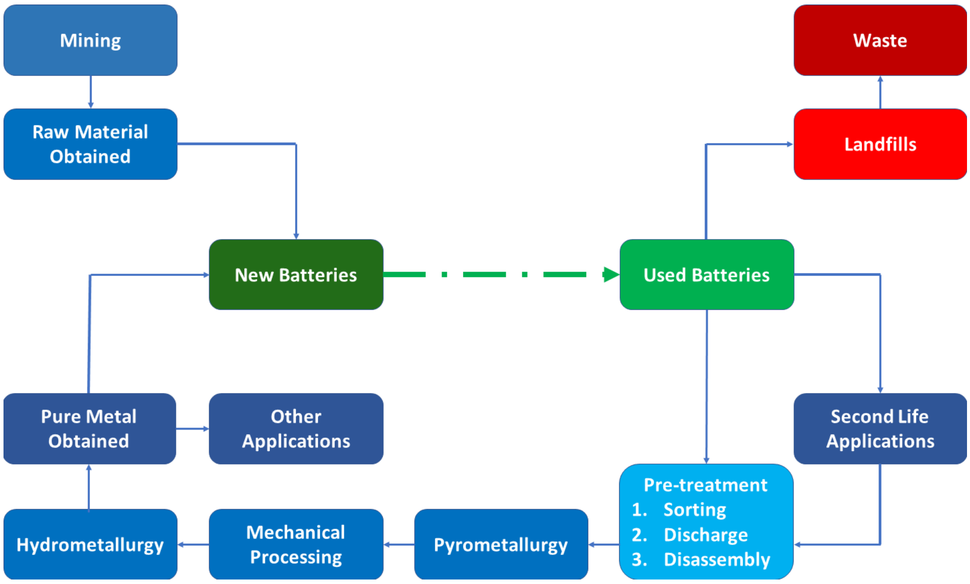

Figure 4. Lithium-ion battery recycling process.

4.3.1. Sorting, Discharge, and Disassembly

Initially, preprocessing is performed, where the chemical composition of the battery is not altered. This phase involves disassembly, stabilization, and sorting according to the chemical composition, battery type, density, shape, and size. After the preprocessing phase, the batteries are refined and concentrated using one of the three methods discussed in the next sections [1,46].

4.3.2. Pyrometallurgy

The main aim of the pyrometallurgical process is to deactivate the batteries by discharging them before mechanical treatment, as it is crucial for the safety of the workers. The pyrometallurgical process involves a high-temperature furnace which is first emptied and then filled with pure nitrogen gas. The metals and oxides in the battery are melted and converted into copper, cobalt, iron, and nickel alloy via redox reactions at approximately 500–600 °C. Additional separation processes follow the pyrometallurgical treatment, and hydrometallurgical processes recover these metals. The aluminum and lithium oxides generally become a part of the slag and are not recovered. The process must be carried out under vacuum conditions if the aluminum is to be retrieved in pure metallic form. As such, the output of this process is alloys, gases, and slag [8,18,47].

4.3.3. Mechanical Processing

Mechanical processing and pretreatment are essential before the hydrometallurgical treatment of the batteries. In this, the task is to separate the metals such as iron, cobalt, copper, aluminum, lithium, and nickel (called black matter). This step will increase the surface area by crushing the component metals so there is greater efficiency in the metal dissolution during acid leaching [33]. Equipment such as rotary shears and hammer mills pulverizes the battery components. After this, a number of separation techniques are employed to separate the various metal shreds from each other. Magnetic separation is first used to remove the external steel casing [37]. Density separation, froth flotation, sieving, and vibrating drum screens are other methods to separate the component metals. To subject the batteries to crushing and other mechanical processes, they must be deactivated and discharged first, as was done in the disassembly and the pyrometallurgy phase [40]. If this is not carried out correctly, there is a risk of battery explosion or fires. If the thermal deactivation phase is to be omitted, the pulverization can be carried out under cryogenic conditions or in an inert atmosphere. It prevents the metals from reacting with oxygen and catching fire. However, this is significantly more expensive [8,36,37].

4.3.4. Hydrometallurgy

Hydrometallurgy is the next step in extracting valuable metals such as cobalt, lithium, copper, and nickel. It involves the leaching of the black matter followed by crystallization, solvent extraction, membrane separation, electrochemical processing, and precipitation. Hydrometallurgical processes are beneficial because less energy is required, since there is no need for elevated temperatures, and lithium is recovered in its carbonate form [2,48]. During the leaching process, the black matter is dissolved in a strong inorganic acid such as hydrochloric acid (HCl), sulphuric acid (H_2SO_4), nitric acid (HNO_3), or phosphoric acid (H_3PO_4). Research is also being carried out on adding reducing agents to aid leaching. It was found that adding H_2O_2 helped convert insoluble $CO_3{}^+$ to soluble $CO_2{}^+$, improving cobalt and lithium yield from 40% and 75% to 85% each. Organic acids such as citric acid, malic acid, and aspartic acid have also been used for leaching and have shown satisfactory results [4,20,29,36,37,47].

4.4. Recycling Processes Followed in the Industry

In this section, we discuss a number of significant companies currently recycling LIBs for various metals. Although the methods followed by the companies are based on the same general processes, they are unique and mainly dependent on the metal they are trying to obtain. Every process in the industry is a hybrid, and this is no exception. This process starts with the mechanical treatment of the batteries and progresses to pyrometallurgical methods to make them inactive. This process finally ends with hydrometallurgy, where the desired metal is precipitated.

4.4.1. Sony–Sumitomo Process

The Sony–Sumitomo process has a capacity of approximately 150 tons per year and was developed by a team that comprised members from both Sony Electronics and the Sumitomo Metal Mining Company. The process is a combination of hydrometallurgy and pyrometallurgy. It starts with the calcination of the battery at approximately 1000 °C, where organic materials and electrolytes are burned off. After this step, the remaining mixture is mechanically separated so that minerals such as aluminum, copper, and iron are isolated and can be reused [1]. The resulting powder is now subjected to hydrometallurgical treatment, in which lithium and cobalt are obtained and reused in the manufacturing of batteries [38].

4.4.2. Retriev Technologies Process

Previously called Toxco, the Retriev Technologies process can recycle all kinds of LIB compositions and has a capacity of 4500 tons per year. This process combines cryogenic, mechanical, and hydrometallurgical treatments to recycle LIBs and recover all cathode metals. The process starts with disassembling the larger battery packs and shredding them in a brine solution or a cryogenic liquid nitrogen environment to prevent violent reactions. Generally, this nitrogen is used when the number of batteries is high. It is also helpful to make the outer plastic casing hard and brittle to break off quickly in the shredding process. The smaller batteries, such as the ones in smartphones, are not disassembled, and they enter the process directly [49].

After the shredding process, the slurry is further processed with a hammer mill with lithium brine, which enables the separation of the bigger particles by screening. The crushed material enters a water-sprayed shaker table and is separated into three phases. The first phase is the copper cobalt product. The second phase is the lithium-ion fluff, which is treated with sodium carbonate to precipitate lithium carbonate. It is then washed, cleaned, and sold. The third phase is the slurry, which is filtered and results in a sludge containing cobalt, copper, nickel, manganese, and iron. It is then processed to make a cobalt manganese product sold to smelters [1,38].

4.4.3. Accurec Battery Recycling GmbH Process

Accurec Recycling GmbH is a German company, and the process developed by them was first meant for nickel–cadmium batteries. However, this process has been extended to many other battery types, including LIBs. It is a hybrid process, which includes steps of mechanical, pyrometallurgical, and hydrometallurgical processes. The batteries are first sorted, cleaned, and dismantled, followed by mechanically processing them to remove as much plastic material as possible [1,38]. This processed feed is then transferred to the vacuum thermal treatment chamber for pyrometallurgical treatment at approximately 250 °C. It ensures that the electrolytes, solvent, and volatile hydrocarbons are removed, and the LIB becomes inert. After this step, the remaining mixture is crushed via various milling and grinding processes and then passed through mechanical separators such as magnetic separators, sieves, air separators, vibrating screens, and zig-zag separators [50].

This treatment leads to the formation of metallic fractions, producing an iron–nickel fraction, an aluminum fraction, and some aluminum–copper fraction from which the metals can be directly extracted. A binder is added to the remaining fraction and then agglomerated and compressed into briquettes. These briquettes now undergo a two-phase pyrometallurgical reduction process. In the first phase, the briquettes are treated in a rotary furnace at 800 °C and in the second phase, they are treated in an electric arc furnace. After this, pure cobalt is obtained, while manganese and lithium are lost in the slag. However, lithium can be recovered from the slag through its leaching by means of hydrometallurgy [1,38,51].

4.4.4. Redux Battery Recycling GmbH Process

REDUX Recycling GmbH is a German company that recycles all lithium-ion batteries. Battery recycling generally involves four stages. In the first stage, i.e., unloading, the battery systems are identified, assessed, and discharged. The batteries with high energy values require full discharge. The energy obtained is fed back into its grid. In the second stage, i.e., disassembly, the energy storage systems are dismantled by hand. Given the extensive variation in sizes and types, it is the most genuine way to accomplish the highest output of secondary raw materials. Depending on the type of battery, this process takes 20–60 min. This process yields plastics, cables, aluminum, or electronic components. Subsequently, the cells of the batteries are deactivated using a unique thermal treatment process, after which the coating of the electrode conductor foils, and the separator electrolyte are removed.

Contrary to conventional methods, this stage even extracts aluminum conductor foil with the help of process control. This third stage is called thermal pretreatment, in which the maximum amount of material is extracted from the thermally deactivated batteries through mechanical processing [52]. It is followed by mechanical fragmentation and separation in a special-purpose system. Recycling products produced in this process are aluminum, iron, aluminum/copper mixture, and active mass. The latter is not contaminated, allowing a hydrometallurgical treatment process to recover nickel, cobalt, copper, and lithium [53].

4.4.5. Primobius Battery Recycling GmbH Process

According to [54], Primobius GmbH offers recycling solutions for LIBs which are scalable, sustainable, and efficient. The output generated is in the form of materials of better purity and reduced carbon footprint to be used in the supply chain of batteries.

The recycling process includes several steps such as shredding, aeration, and beneficiation. In the first phase, the battery is disassembled, and plastics and metal components are separated from the black mass. This is followed by aeration and beneficiation to further separate the metals from the plastics. In this process, the prior discharge of cells is not required, as wet shredding and an inert gas-covered environment are provided to safeguard against fire hazards. One of the important components recovered at this stage is black mass, which contains the active materials required for battery operation.

The hydrometallurgical process employed in the next phase helps in extracting critical minerals such as lithium, nickel, and cobalt so that they can be reused in making the electrodes of the battery [55,56]. As these materials are separated, the process also provides a concentrated leach solution. When this solution is processed, it helps in extracting sulfides of critical minerals such as sulphates of lithium, nickel, and cobalt as well as manganese and copper sulfates. These can then be directly sold back to the lithium-ion battery (LIB) supply chain. Thus, using a two-phase recycle process, Primobius ensures that critical minerals are extracted and pushed back into the LIB supply chain for further industrial use.

4.4.6. Fortum Battery Recycling Process

While Primobius focused more on the hydrometallurgical process, there is another company, Fortum, which combined both mechanical and hydrometallurgical technologies. The mechanical process applied here helps in recovering plastics as well as metals such as aluminum, copper, and black mass. Similarly, to the previous case, the company's hydrometallurgical process is used for recycling black mass, whereas other materials recovered from the process are recycled separately [57].

The hydrometallurgical process here applies a precipitation technique rather than leaching. The critical minerals recovered at this stage are then directly provided to the industries in the supply chain. According to [58], a combination of mechanical and hydrometallurgical processes helps in processing 80% of the battery materials. The company claims that the hydrometallurgical technology used helps them in recovering 95% of the valuable critical minerals from recycling.

These companies continuously look into ways and processes to extract a higher yield and purity from the battery materials (refer Table 4). However, no matter how well-established these processes are, certain drawbacks and difficulties are faced while recycling these batteries. Some of these challenges are mentioned below.

I. High cost: The cost of recycling LIBs is very high, mainly due to two reasons: the existence of minerals that are not very valuable, which increases the cost of segregation and separation from the valuable minerals, and the loss of valuable minerals to the slag during hydrometallurgical processing.
II. Lack of batteries to recycle: In many countries, the bulk of LIBs used in devices such as smartphones and laptops are directly discarded in dustbins, resulting in landfills. Even rechargeable AA batteries meet a similar fate. This pollutes the environment with toxic chemicals and reduces the number of LIBs available for recycling.
III. Dangers of transportation: Transporting LIBs is very dangerous and expensive since they can catch fire and explode under mechanical stress. Therefore, different countries place regulations on them when being transported.
IV. Dangers of handling: Even after the batteries are transported successfully, there are other risks involved in treating them. The batteries can catch fire if the dismantling process is not performed carefully. These batteries also contain toxic chemicals that pose a health hazard to the people working on them.

Table 4. Comparison of the processes of the different companies.

Company → / Parameters ↓	Sony–Sumitomo	Retriev Technologies	Accurec Recycling GmbH	Redux Recycling GmbH	Primobius Recycling GmbH	Fortum
Type of process	Pyrometallurgical + hydrometallurgical	Cryogenic + mechanical + hydrometallurgical	Mechanical + pyrometallurgical + hydrometallurgical	Mechanical fragmentation and separation + hydrometallurgical	Mechanical + pyrometallurgical + hydrometallurgical	Mechanical + hydrometallurgical
Capacity	150 tonnes/year	4500 tonnes/year	4300 tonnes/year	10,000 tonnes/year	3650 tonnes/year	3000 tonnes/year
Final products	Copper, iron, lithium, and cobalt oxide	Lithium carbonate, copper cobalt, and manganese cobalt	Lithium chloride, lithium carbonate, and cobalt	Pure nickel, cobalt, lithium, and copper	Lithium cobalt oxide (LCO) cathodes and lithium nickel manganese cobalt (NMC) cathodes	Lithium, nickel, cobalt, and manganese

5. Conclusions

In the rechargeable battery space, lithium-ion batteries (LIBs) have created their own niche. This is due to their superior quality in terms of high density, their better retainment of charge over time, and the low cost of buying them. Having a low self-discharge rate makes them yet more valuable, as it means that they can retain charge when not in use. One of the risks associated with LIBS is that of them catching fire if proper care is not taken in their storage or if they are not charged as per the given instructions. Their design ensures that electrodes cannot touch each other, but it is better to ensure that the battery is not damaged, as damage can lead to fire hazards. Irrespective of the perennial issues of aging and retaining less charge over time, LIBs have still retained a superior position as a potent energy source for most devices.

The introduction of the circular economy in the LIB industry begins with rethinking and reformulating the industrial manufacturing processes as well as the design of the product so that it becomes more conducive to reusing and repairing processes. Recycling should only be carried out when there is no scope for further reuse and refurbishing processes. Even the recycling processes should be optimized so that the amount of wasted valuable minerals is reduced and the cost is proportional to the yield. Lithium, as mentioned before, has vast applications in various industries, and its demand is set to increase drastically, considering the growth of EVs in the past few years. In a situation like this, tackling the number of wasted LIBs takes a high priority, not only to protect the environment from pollution but also to reduce mining activities that destroy the flora and fauna of the area.

There is no correct way to recycle batteries, since they all have advantages and drawbacks, and they still have scope for improvement. This paper elaborates on the emerging and established recycling processes that agree with the circular economy model, hoping that the information compiled in it may help further research in improving the processes.

The findings of this research may attract the attention of a number of battery recyclers, as they suggest through a secondary case that many companies are actively working in this area and also focusing on CE adoption. The result of the analysis may assist sustainability professionals and operations managers to convince their business leaders and senior managers of the operational benefits of CE adoption and its effect on LIB recycling.

The present study is based on a review of the literature and secondary case studies. It will be interesting to see how CE adoption is actually working in lithium-ion battery recycling. Primary case studies will be helpful in improving the understanding of the practical relevance of CE adoption in the battery recycling industry. Future studies can also focus on a survey of battery recycling industries to understand the enablers, challenges, practices, actions, etc. related to CE adoption.

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