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Recycling of valuable elements contained in waste lithium ion batteries

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Abstract

Today, fossil fuels that are not renewable are frequently used. Since their use has continued from past years, their reserves have decreased and since they cannot be renewed, renewable and recyclable materials are needed. These renewable and recyclable energy sources are frequently mentioned because they can be used over and over again or are ready for reuse after being used. In this article, Lithium-ion (Li-ion) batteries, which can be recycled, and the recycling stages of these batteries are mentioned since the authors of the article want to work in this field. Lithium-ion batteries (Li-ion) are a type of battery that can be charged and used as an energy storage device. Li-ion batteries are actively used today. The anode material of Li-ion batteries is usually graphite made of carbon. The cathode material is usually metal oxides. The electrolyte material is usually lithium salt in an organic solvent. Lithium cobalt oxide (LCO), lithium iron phosphate (LFP), lithium manganite (LMO), lithium nickel manganite cobalt oxide (NMC) are Li-ion battery types. Recycling of these types is carried out using pyrometallurgy, hydrometallurgy and mechanical separation methods. In this period when recycling is increasing day by day, this review article on Li-ion batteries is written to help readers in their studies and research.

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1.Introductıon

Energy is indispensable for human life. When we look at today, energy is used in many fields such as health, education, industry. A disruption in energy that will occur also causes disruptions in our lives [1]. In recent years, the focus on sustainable energy sources has been increasing [2]. Sustainable energy sources become inefficient when they are not suitable for use. It is important to store sustainable energy resources in order to use them continuously and efficiently. The storage of energy helps people at this point [3]. Efficient storage of the obtained energy is possible with batteries. The closest battery to today's batteries was launched in 1969 as a lithium sulfur dioxide (Li-S) battery [4]. Then, lithium-polycarbonate monofluoride batteries were available on the market in 1973 and lithium-manganese oxide batteries by 1975 [5]. The modern Li-ion battery was sold by Sony in 1991 [6]. As a result of various developments, Li-ion batteries have become what they are today. Batteries store energy in short energy in the following way: They have two main components consisting of an anode and a cathode. The oxidation reaction occurs at the anode part and the reduction reaction occurs at the cathode part. There are electrode between the anode and the cathode that allow the transport of electrically charged particles. Electrons transported in this way form an electric current [7]. They are widely used due to their characteristics such as long cycle life, specific capacity, energy efficiency, and the ability to be produced according to the area to be applied [8]. Batteries can be divided into groups such as nickel-metal hydride batteries, Li-ion, decoupled redox flow batteries and lead acid batteries. As a result of the acceleration of technology in recent years, Li-ion batteries have become more prominent [9]. Li-ion batteries are available in 4 different types Decoupled among themselves. CCCCLi NMC batteries have good capacity and stability[5]. Lithium cobalt oxide (LCO) batteries are highly suitable for electronic devices and electric vehicles [10]. Lithium Manganese Oxide (LMO) batteries have high thermal stability of the battery [11]. Lithium Iron Phosphate (LFP) batteries come to the fore with their low raw material costs and long-lasting structures [12], [13], [22]–[24], [14]–[21].

The general characteristics of Li-ion batteries include long cycle life, high cell potential and high energy density, usability between -20°C and 60°C, and fast charging[25]. Thanks to all these advantages offered, Li-ion batteries have a wide range of uses, such as electronic devices that we use in everyday life, electric vehicles and in the healthcare sector [26]. The wide area of use is increasing the demand for Li-ion batteries day by day [27]. But as a result of this demand, the number of batteries used is increasing and millions of batteries are thrown away every year [28]. The environmental and economic damage of discarded batteries is increasing every passing year. To produce a battery with 1Wh storage, an average of 110 gCO_2-equivalent greenhouse gases are released into the environment Tue [29]. By ensuring the recycling of batteries, it is estimated that it will bring sales of 23.72 billion dollars by 2030 [30]. In this context, it is important to ensure the recycling of Li-ion batteries. Current research on the recycling of various Li-ion batteries is included in this article [31], [32], [41]–[50], [33], [51]–[53], [34]–[40].

2. The status of lithium-ion batteries

Commercially used lithium ion batteries have charging and discharging characteristics. Lithium provides the entrance to the graphite developed through the anode component (FIG1). Then, from the cathode component in another direction of the battery, lithium ions are released towards the electrolyte part. This is the simplest description of the charging and discharging process of Li-ion batteries [54]. Lithium ion batteries are actively used in electrical household appliances, battery chargers, technological devices, electric vehicles and industry. As a result of the increasing demand for electric vehicles in recent years, interest in lithium-ion batteries has been increasing. Lithium ion batteries provide benefits in terms of recycling and recovery after use, as well as beneficial use for the environment [55].

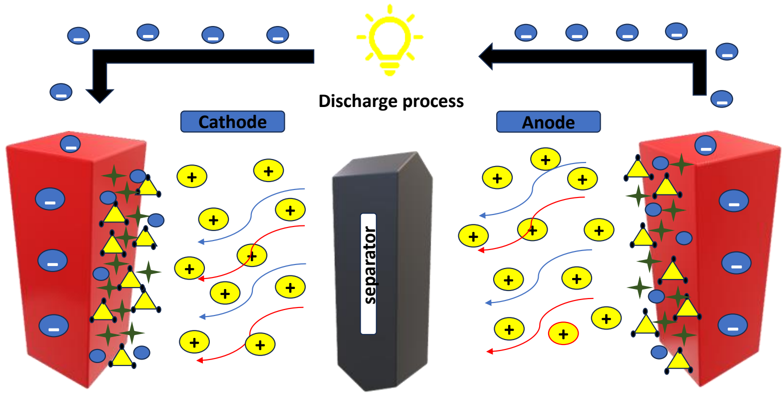

Fig 1. Energy flow diagram of Li-ion batteries from anode to cathode.

Li-ion provinces are preferred more due to the fact that electric vehicles are starting to be used instead of gasoline-powered vehicles. Lithium ion batteries have a positive effect in many ways. Lithium ion batteries greatly reduce greenhouse gas emissions. Lithium ion batteries make the energy collected from renewable energy sources such as wind, solar, geothermal of high quality [56] .

In addition to being successful, lithium-ion batteries also have negative effects in some cases. The production of lithium hydroxide, which leads most lithium electrodes, takes a long time. The start and end period of production of lithium hydroxide material can cover a time envelope of 2 years. The long duration of the production phase causes it to be affected by economic variability and increase the cost[57].

Lithium-ion batteries mainly consist of electrodes, electrolytes and separator components. Anode materials in commercially used batteries are mostly composed of nano-sized graphite. On the other hand, cathode materials may vary depending on the product to be produced [58]. The current state of the most preferred cathodes;

It is known as LCO, LFP, LMO and NMC. Lithium ion batteries manufactured from these different cathodes have different positive and negative properties. If some of these features need to be mentioned;

- Batteries produced from the LO cathode are more expensive and it is observed that O_2 gas comes out in case of overcharging.
- Batteries produced from NMC cathode show slower speed compared to others.
- On the other hand, a negative feature of LFP batteries is that the electrical conductivity condition is low.
- The batteries produced with $LiCoO_2$ have a high-performance electric conduction feature.
- As observed in the current situation, LCO is preferred more when the properties are compared. The most preferred cathode product in the portable electronics sector is $LiCoO_2$ [56].

Various chemical and physical changes such as volume changes, phase transition, side reaction can be observed while lithium ion batteries are in operation. There are complex layouts that Li-ion batteries have realized. These complex layouts show the technique of connecting directly to the electrochemical reaction of a Li-ion battery.

The characterization represents the principle of operation of the batteries and the status of the analyzed studies. In order for the research to be put into practice, the batteries must be fully operational under the conditions required by the defined order. This process often brings with it specially developed devices and measuring devices. When controlled by taking these studies into account, it is seen that the methods of characterizing lithium ion batteries have been developing more rapidly in recent years. In this regard, especially in the studies carried out after the increasing interest in recent years, the projects developed also have an important place in the research [59].

3. Ensuring recycling of used lithium-ion batteries

The recent increase in the use of lithium-ion batteries has resulted in thousands of tons of battery waste [60]. Recious metals are used in the production of these batteries. In this context, it is obvious that the recycling of precious metals in used batteries is of great importance. Recently, as can be seen in Graph 1, when we look at the studies with the words "lithium-ion batteries" and "recycling"; it is seen that there were 3 articles in 2012 and 149 articles in 2022. This situation also emphasized the importance given to recycling.

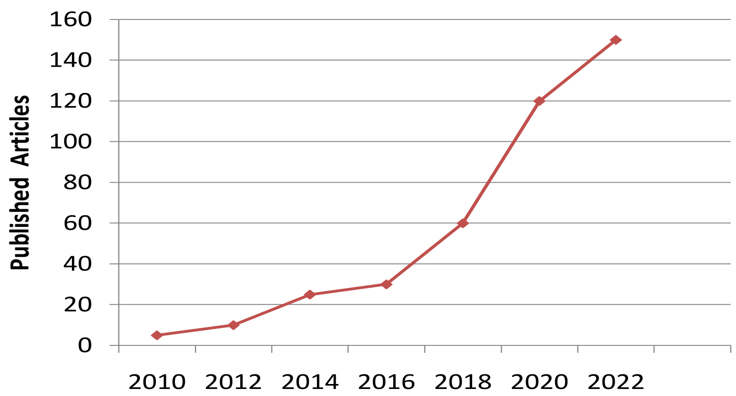

Fig 2. Distribution of articles written about Li-ion batteries by year.

In addition, with this increase in the number of articles, it is seen that it makes a high contribution to the recovery of valuable and scarce resources in the world in order to ensure a sustainable future [61], [62]. In this context, it is known that many different methods are used in recycling studies. Mechanical, pyrometallurgical and hydrometallurgical methods can be given as examples[63].

3.1. Recuperation of cathode substances from discarded LCO batteries

Lithium cobalt oxide (LCO) is the first cathode material to be released to the market. For more than 30 years, the market has been widely used. LCO batteries have become a popular energy source with the increase of electronic devices and electric vehicles [64]. Research shows that by 2025, the need for LCO batteries and the raw materials to make them will increase [65]. Considering the developing and growing Sunday, recycling of LCO batteries that have become unusable is of great importance when considering their macroeconomic and environmental impacts [66]. Recycling of LCO batteries can be achieved by various techniques: chemical categorization can be achieved by direct regeneration, pyro metallurgical, hydrometallurgical techniques.

3.1.1. Recycling processes by hydromealgic technique

Hydrometallurgical recycling technique can be characterized as the place where various leaching(leaching=precipitation and calcination processes are performed [67]) it includes transactions. Precious metals are obtained with chemicals that do not show solvent properties in the leaching process. Organic acids are preferred in leaching processes today. Organic acids are used in the internal processes of LCO. Qu et al., for instance, leached the LCO cathode material following high-energy ball milling using 1.25 M citric acid [68]. When the experimental results were examined, it was shown that the optimum leaching process of 94.91% for cobalt (Co) and 97.22% for lithium (Li) was realized with the optimum grinding speed of 500 rpm and the optimum experimental condition of 80°C in 30 minutes. In another example, it was performed by Chaudhary and others [69].1.25 M citric acid and 1% H_2O_2 were combined by Chaudhary et al., for the leaching procedure. He finished the leaching process at 90 °C in 5 hours. At the end of the process, the yield was found to be 99.93% for cobalt and 99.7% for lithium.

In another study, Verma Et al., [70] LCO leaching was performed using 0.58 Moxalic acid ($H_2C_2O_4$) and 1.16 M hydrogen peroxide (H_2O_2). It has been shown that the mixing speed is realized with an optimum experimental condition of 600 rpm and a temperature at 80°C. After that, KOH was added to raise the pH, causing it to rise over 13. Lithium was precipitated with potassium carbonate (K_2C0_3) and Li_2CO_3 was released. Then, for the recovery of cobalt, $CoC_20_4•2H_20$ was gradually added. KOH has been added because it is necessary to keep the pH high. Weak Co yield has been observed.

Liao Et al., tried a different approach. Biolic process it has managed to separate 99% of Co, and 100% of Li from batteries that have become unusable with [71]. A mixed culture containing *Acidithiobacilluscaldus* and *Sulfabacillus* thermos sulfidooxydans was formed during the biolic process. Lossless removal of metals from "Stemphyliumleafblight" (SLB) has been facilitated by Fe $^{2+}$ -MCB process. For this reason, the harmful SLB has become harmless and has lost its toxic effect. Deep eutectic solvents (DEC) are often preferred in analytical chemistry applications today. DECs are an alternative to ionic plasters. They exist in liquid form, such as ionic liquids, at room temperature. When combined with different solvents, it provides low vapor pressure and is economical. According to Peters et al., using DEC. Co and Li have managed to decompose from the locos that have become unusable [72]. It is also produced with choline chloride and ethylene glycol. Next, a 10 mL glass bottle was filled with 100 mg of LCO and 5 ml of liquid ant to begin the leaching process. It was mixed for 24 hours at 900 rpm between 180°C-140°C degree and leaching process was performed. The optimum leaching yield was found to be 91.6% at 180 °C at 900 rpm under 24-hour conditions.

Peters et al., showed different approach to DEC and proposed an economical and environmentally friendly solvent with choline chloride-citric acid solvent [73]. The study provided evidence that, in the presence of copper and metallic aluminum in LCOS, a 2:1 M solution of citric acid and choline chloride diluted with distilled water is an effective solvent. With this method, LICoO2 was filtered at 40 °C and 60 minutes and >98% of cobalt (II) was separated.

Liu et al, tried a low-cost DEF approach and achieved Li and Coextraction efficiency close to 100% [74]. They used microwave to separate Co and Li from LCO. In the scope of the study; DEF was used for leaching in microwave, low viscosity and low temperature. The mixture was mixed at 80°C for 30 minutes until it became homogeneous. Then he glided away. Saturated sodium carbonate solution was added and Co recovery was realized. The filter process has been applied and dried.

3.2. Recovering cathode material from LFP batteries that have become empty

LFP batteries are a type of battery that is mostly used in electric vehicles and smart systems [75]. Especially the interest and increase in electric vehicles has also increased the interest in LFP batteries[76].The increase in the use of LFP batteries has led to an increase in the number of waste batteries [77].Waste LFP batteries must be disposed of carefully. The most valuable component of expired LFP batteries is lithium [78]. Due to the distinctive feature of LFP crystals, the Li component can be selectively removed using engineering techniques. Li extraction has aided in the advancement of several useful technologies, including electrochemistry, pyrometallurgy, hydrometallurgy, and mechanochemistry [79].

3.2.1. Hydrometallurgical technical recycling processes of LFP battery

The recycling stages of LFP batteries by applying hydrometallurgical processes can be found in the relevant sources [38],[81]. To give an example of these recycling steps; Fan et al. during the work he did, the battery was charged and the supply of iron (III) phosphate (FePO4) and lithium graphite, which will be used to extract lithium, was provided Jul[82]. He first used water as a leaching material [67]. The removal of lithium from the anode is given in the following equation (reaction 1). In the second and third equations, the equation created with the NaOH cathode for the recovery of iron and phosphorus is stated [82] (reactions 2-3).

$$2LiCx + 2H_2O \rightarrow 2LiOH(aq) + 2xC + H_2 \quad (1)$$
$$FePO_4 + 3NaOH(aq) \rightarrow Na_3PO_4(aq) + Fe(OH)_3 \quad (2)$$

$$Na_3PO_4(aq) + 2LiOH(aq) \rightarrow 3NaOH(aq) + Li_3PO_4 \quad (3)$$

The Na_3PO_4 solution was mixed with LiOH solution. As a result of this mixture, lithium phosphate was formed. For this reason, NaOH was formed again and a loop was created to filter it again. Three parameters were used in this study. These parameters are the volume of the substance of the filter, the concentration of the substance of the filter and the filtration time. In the study, suitable leaching conditions were determined as 0.5 hours, sodium hydroxide (NaOH)/Fe molar fraction of 4.5 and 1 mol/L NaOH, and the appropriate leaching yield was determined as 99.7%. It has been observed that the method used with the help of these efficiency values is better and more suitable than the traditional pyrometallurgical and hydrometallurgical methods. Removing the components used as an anode or cathode from the LFP battery determines the flow contact. Removing the components used as an anode or cathode from the LFP battery determines the flow contact (Figure 2) [56].

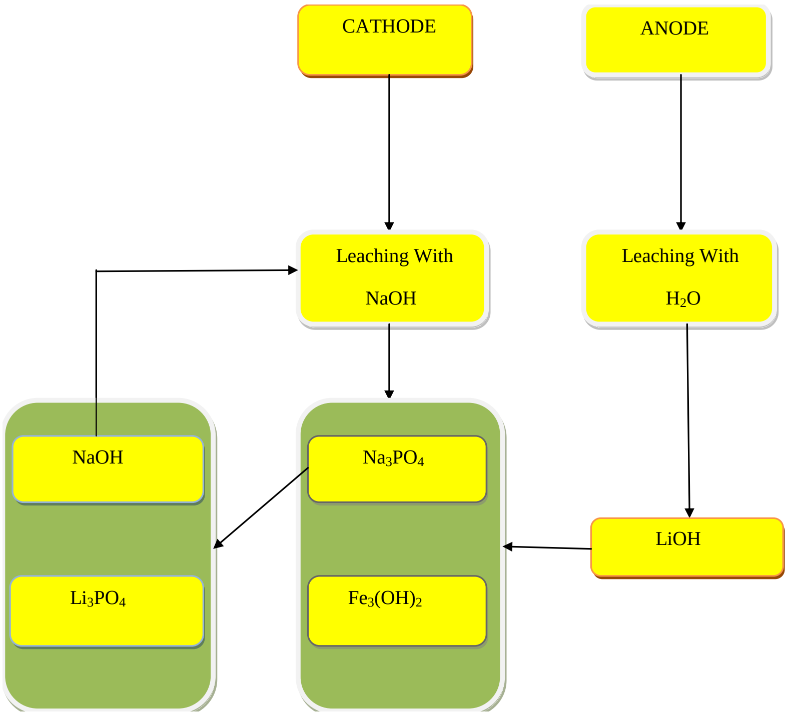

Fig 3. Flow diagram of removing components used as anode or cathode from an LFP battery.

Liu et al.'s study claims that certain mechanisms involving sodium hypochlorite (NaCIO), hydrochloric acid (HCl), an acidic environment, and oxidants caused reactions to occur in reconstituted LFP batteries [83]. In this context; HCl/Limolar ratio, HCl concentration and CIO/Li ratios were investigated. The results of this study indicated that the most suitable operating parameters for the internal efficiency of Li were; T= 15 °C; HCl - LLI molar ratio= 1.3; NACIO - NUNLI ratio= 0.6; t=5 min. In addition, Li-ions contained in the leaching solution for the extraction of Li_2CO_3 were precipitated with the help of Na_2CO_3 (sodium carbonate). In addition, the leaching residues produced from $FePO_4$ can be used for the regeneration of LFP powders. This process allows the separation of metal ions, which are difficult to separate, and as a result of the leaching process, Li_2CO_3 and $FePO_4$ were obtained [83].

Yue et al. in his study, it is possible to recycle LFP batteries with visible light photo decay ($NaFeS_2$) produced in addition to the recovery of Li and P [50],[52]. First, LFP powder was added to sodium peroxidisulfate ($Na_2S_2O_8$) solution, then it was mixed for 30 minutes. As a result of the processes performed to obtain LFP powder, the Li ion was recovered. After these steps, it reacted with LFP and Na_2SO_8 at room temperature. This reaction produced a photocatalyst ($NaFeS_2$) and phosphorus was formed [87].

The appropriate parameters are listed as LFP/Na_2S= 1; LFP/Na_2SO_8= 1:0.9; reaction temperature= 24°C; mixing time= 30 minutes. The ability of $NaFeS_2$ on absorption and photocatalytic synergistic has been tested. The results were then compared with methylene blue (MB)and TiO_2. As a result of the comparison, it was determined that MB was 67 times faster than TiO_2 when the degradation rate of methylene blue and TiO_2 by $NaFeS_2$ was compared. Thus, it has been revealed that the synthesized $NAFeS_2$ is suitable for removing organic paint pollution [88].

3.2.2. Recycling processes of LFP battery by pyrometallurgical technique

One kind of extractive metallurgy that is particularly useful for removing and recovering metals from materials is pyrometallurgy [82].This method can show processes such as concentrating, roasting, simplified recycling process and high temperature melting technique. Although this method provides important advantages such as high processing capacity and high applicability, it shows high energy consumption [89],[81].

In pyrometallurgy, research is done on recycling LFP batteries using techniques such sodium salt aided roasting. Zhang and associates carried out a study with two distinct sodium salts. Na_2CO_3 and naoh are these salt kinds [90].The carbothermal reduction approach is another one of these investigations. Using this method, a spent Li_2CO_3 battery was converted into the same product using an LFP battery. The following is the response for obtaining Li_2CO_3 again [56] (reaction 4 and 5).

$$2LiFePO_4 + (NH_4)_2S_2O_8 \rightarrow 2FePO_4 + Li_2SO_4 + (NH_4)_2SO_4) \tag{4}$$
$$Li_2SO_4 + Na_2CO_3 \rightarrow Li_2CO_3 + Na_2SO_4 \tag{5}$$

3.2.3. Recycling processes of lfp battery by mechanochemical technique

The mechanochemical method is a method used to extract Li from the LFP battery [91], [92] In this method, sodium citrate is a common grinding agent that does not contain pollution and provides recovery. After mixing the LFP powder with sodium citrate, it was mixed with hydrogen peroxide. Then, the zirconia was placed in a grinding jar with a bal. 98.9% of the Li obtained from LFP is selectively usable, and the remaining Li is converted to Li_3CIT and dissolved in water. Under favorable conditions; Sodium citrate/LFP mass ratio 10: 1; addition of 1mL H_2O_2; 500 rpm rotation speed and milling time in 5 hours; Reactions of this process (reactions 6 and 7) [92];

$$2Na_3Cit + 6LiFePO_4 + 3H_2O_2 \rightarrow 2Li_3Cit + 6FePO_4 + 6NaOH \ (pH < 8) \tag{6}$$
$$FePO_4 + 3NaOH \rightarrow Fe(OH)_3 + Na_3PO_4 \ (pH > 8) \tag{7}$$

3.2.4. Direct regeneration processes

LFP recycling can be performed with the direct regeneration technique. The used LFPs become reusable with a functionalized pre-separation separator and electrochemical treatment of the electrode.

The regeneration process was carried out to reconstruct the cathode particle structure, which had undergone decay, without damaging the bulk phase [93], [94].

Pre-lithification of the pre-separation separator started with the electrochemical deposition of a thin layer of Li-rich material on a commercial separator [95]. In this context, the reason why lithium oxalate (LiC_2O_4) is chosen for the transportation of Li is its air stability and low cost.

3.2.5. Recycling processes by electrometallurgical technique

Lithium is extracted from salt water by using lithium iron phosphate ($LiFePO_4$) cathode materials. Salt lake water and Li minerals are the primary sources for lithium products [96]. The high concentration of Li in the brine obtained from the Salt Lake provides cheap extraction costs. For this reason, Li extraction from salt water is one of the most preferred options. Characteristically, salt water variably has the elements Li, K, Ca, Na and Mg. Electrochemical extraction method methodology (ELEM) has been developed to provide Li extraction from salt water [97]. This method is environmentally friendly, easy to use and does not need additional chemical reagents. Thanks to these features, it is more advantageous than traditional extraction methods [98].

3.3. Recovering cathode substances from LMO batteries that have become empty

Due to the scarcity of minerals to be used for battery production and the high cost of processing, it is very important to recover these minerals from used batteries. Li-ion batteries used to store energy today have short lifetimes. Sustainable recycling methods of these batteries have been one of the most focused topics in recent years [99].Recycling plays an important role in the sustainability of Li-ion battery production, as it reduces the demand for virgin material and the environmental pollution caused by irresponsibly disposed batteries [100].

Lithium manganate ($LiMn_2O_4$) batteries are more advantageous than lithium cobalt (LiCo) batteries. LMOs have a preferred cathode material due to its high operating voltage, low cost, low toxicity, high reserve, reliable by nature and environmentally friendly [101].

Some used Li-ion do not contain a single cathode material. Due to this, differences may be observed in recycling methods. Various methods are used for the recovery of cathode materials of intact lithium manganese oxide (LMO) batteries [56],[102].

3.3.1. Recycling processes of LMO battery by hydrometallurgical technique

The hydrometallurgical process is the first method for the Li-ion battery to be recovered [102].It involves the leaching and separation of metals in an aqueous environment. The process steps consist of acid/alkali leaching or bio-leaching, chemical deposition, solvent extraction and electrochemical deposition [103]. Leaching systems are made with inorganic acids such as HCl (hydrochloric acid), H_2SO_4(sulfuric acid) and HNO_3(nitric acid). In addition, although it has been tried to be used in H_3PO_4 (phosphoric acid) and organic acids, H_2SO_4 has been preferred due to its prices and reactivity [104].

Wu Et al., renewed the spent $LiMn_2O_4$ battery by chemical reduction method, the Li source of which was lithiatedpyrene (Pyrene- Li), using the hydrometallurgical process. The Pyrene-Li compound is a reagent that can give electrons (e-) and Li^+ and has a high amount of reducing agents. In this experiment, it was preferred to eliminate the Li deficiency in the waste LMO cathode. It has been concluded that Li-deficient LMO and degraded LMO prepared by acid deletion method are originally recoverable due to long-term stability and superior speed capacity, which can be compared with intact LMO cathode [105]. In another example, Wang et al., chose to use acid leaching to recover LMO. In this experiment, Wang et al. combined acid leaching with the resynthesis of cathode material. Here, Li and Mn leaching yields of over 94% were achieved using 60 g/L solid-liquid ratio, 1.0 M citric acid, and 1 h leaching. Spinel LMO was resynthesized by sol-gel method using the leachate generated after leaching[106].The studies carried out were not limited to these two experiments.

In another example, Zhou et al. succeeded in recycling $LiMnO_{24}$ cathodes without using acids, bases or chemical reductants, only a pH gradient provided by neutral water electrolysis [107]. Mohanty et al. used Na-D_2EHPA as an extractant and sulfuric acid as a decomposer. They investigated the recovery of MN from used oil using hydrometallurgical process [108].Yao et al. tried to recover LMO both inexpensively and in the most environmentally sound way. For this purpose, they used citric acid as leaching reagent and glucose as reducing agent [109].

3.3.2. Recycling process of LMO battery by pyrometallurgical technique

In the pyrometallurgical process, non-metallic components are decomposed into gas. Metallic components, on the other hand, reduce the spent Li-ion in a high-temperature furnace, where they are reduced to alloys or metals. But there are negative aspects such as high energy consumption, the product formed as a result of the process is not pure enough, the release of secondary polluting gases such as dioxin, furan [110]. Looking at the studies conducted; Tao and colleagues have pioneered studies on the recovery of $LiMn_2O_4$ batteries with one-step pyrolysis that does not contain acid leaching as a reductant. After, LMO powder was placed in the quartz tube and electrolyte evaporation was demonstrated between 30-180°C in the first stage. In the second stage, high temperatures (180-800℃) were reached and CO_2 emission was observed while the cathode material was reduced. It has been observed that manganese (LMO), which has a high valence, is reduced to 500 ℃MNO. The leaching efficiency of Li at 1 hour pyrolysis time with a nitrogen flow rate of 50mL/min at 500℃ has been calculated as 99% and the leaching efficiency of manganese is 99.4% [111].

Liu et al stated that water and sulfate purification leaching methods can be used to recover Mn and Li from LMO. $NaHSO_4$.LMO powder combined with H_2O is roasted between 308-773℃. The Li element in the sample, which was subjected to roasting at a temperature of 600℃ and for 30 minutes, was taken back as $LiNaSO_4$. A change in the valence of the Mn element has been observed. This change has been observed from +4 to +2. After the roasting process, water leaching conditions were applied and the electrification rate of Li was found to be 96.6% and the extraction rate of Mn was found to be 9.7% [112]. Liu LMO has not been limited to investigating the recovery of batteries by the pyrometallurgical process only with this experiment and has investigated the low-cost and environmentally friendly recovery of LMOs using NMP (N-methyl-2-pyrrolidone) in another experiment [101].

Han et al. colleagues investigated the effects of roasting temperature and duration on surface modifications by using the roasting-assisted flotation method from LMO batteries. It has been observed that $LiMn_2O_4$, which was subjected to flotation after the roasting process, was easily separated from graphite. Other analyses have shown that the main reason for the difficulty in separating $LiMn_2O_4$ from graphite is due to the organic binder and electrolyte coating on its surfaces. As a result of this experiment, it was observed that the effects of organic films decreased by changing the contact angles through roasting [113].

Studies of Li and Mn recovery from LMO cathode spent by He et al. method of salt roasting at low temperature [114], Pindar et al. recovered manganese and Li from coin cells by heat treatment using organic acid leaching [115] these are the other studies that we come across in the literature for pyrometallurgy. Hydrometallurgy and pyrometallurgy can be used separately or combined [116]. It is a method that has made its name in the combined pyro-hydrometallurgical process in recent years. This method, which has emerged in accordance with the principles of green chemistry, starts with a thermal pretreatment step involving the collection of cathode materials. Then it continues with the unified recovery process of cathode materials. As the last step, the subsequent separation and extraction process is also carried out [117].

3.3.3. Recycling processes of LMO battery by electrochemical technique

Liang et al. the parameters of recycling manganese element from LMO battery were determined by electrochemical method. These parameters: graphite rod for the counter electrode, a working electrode coated with LMO powder, and finally Ag/AgCl electrode (reference). As a two-electrode system, a constant voltage deoxidation process was determined by using graphite clasp as the cathode and graphite plate as the anode. After these procedures, the temperature of electro-oxidation at a voltage of 0.5–3.0 V was set at 750 °C, and the reduction process of LMO was observed for 12 h. In this experiment, Li could be recovered to the desired extent, while Mn could not be recovered at the desired level. As a result of these experiments, it can be said that the hydrometallurgical process, which is both low cost and environmentally friendly, is more usable for the recovery of Mn and Li [118][56].

3.4. Cathode material recovery from used NMC batteries

Lithium nickel manganese cobalt oxide ($LiNi_xMn_yCo_zO_2$,NMC), one of the important materials in batteries, has high energy density[119]. Thanks to its layer-shaped structure, it changes its volume at a rate of 2% [120]. Promising Li-ion batteries for the future make up the cathode material group [121]. During the addition and removal process, the Li content can reach up to 20%, providing electrochemical stability. NMC provides affordable cost thanks to its high capacity and Mn [122]. The three NMC batteries have chemically similar properties. For this reason, selective and efficient recycling of used NMC batteries has become a difficult goal. When new NMC batteries are produced from used NMC batteries, selectivity is a less important feature in recycling. The separation of cathode materials from used NMC batteries is usually carried out in recycling plants.

The recovery process allows the precious metals contained in the batteries to be separated and made reusable. This process reduces environmental impacts and also reveals an economical method because it produces less electricity [123]. Three main processes are needed to recycle NMC batteries: Hydrometallurgical, pyrometallurgical and mechanical [124].

3.4.1. Recycling processes of NMC battery by hydromealgic technique

The hydrometallurgical process involves reactions that take place in the liquid phase. It mostly involves obtaining the desired precious metals as a result of the decomposition of a metal in the form of a special. In cases where it is difficult to separate some metal ores by other methods, the hydrometallurgical method is used. These processes are carried out by purification methods such as precipitation, liquid-liquid extraction, adsorption, ion exchange after the leaching process [125]. Hydrometallurgical processes provide the recovery of a larger amount of metal compared to pyrometallurgical processes. Ilyas and others [126] in a study conducted by, it is a recovery method that occurs with the precipitation of Li_2CO_3, which occurs as a result of filtering Li at 90 ℃ for the used NMC cathode and adding Na_2CO_3 at the ideal dose of Li+: CO_{2-}=1:1.5 Li_2CO_3. After that, the cathode material was leached at 90 ℃ for about 3 hours using a 3 M HCl solution. As a result of this process step, the dissolution of Co, Mn and Ni was achieved with an efficiency of over 99%. Then, at Mn 80℃ in the leachate, $KMnO_4$ was added at a stoichiometric ratio of 1:1.25 with a pH of 2.0, resulting in selective precipitation. Furthermore, Co and Ni pH were extracted from leachate using an ionic liquid (organic: aqueous) with a ratio of 5.4 and 2:3. Two milliliters of 2 M H_2SO_4 solution were used to carry out the ionic liquid's breakdown. Co_SO_4 with high purity has been degraded via evaporation. It created the crystals of xH_2O. At 50 °C, Ni ions precipitated as [$NiCO_3$·2Ni $(OH)_2$] with a pH of 10.8, and a stoichiometric ratio of Ni_2+: CO_2-= 1:2.5.

Wang et al., in a study he conducted, calcination process was applied to aluminum foil in a CO_2 atmosphere at 600 ℃ for 2 hours [127]. As a result, the aluminum foil is separated from the cathode strips of used Li-ion batteries. The slag obtained after calcination is stripped from the aluminum foil. The goal was to get a solution that contained Mn^{2+}, Ni^{2+}, Co^{2+}ions. For this purpose, it was diluted with 1.5 M HCI for 2 hours at a rate of 20 g/L (solid: liquid). The aluminum foil, which was recovered after separation from the cathode strips, was used to obtain Ni-Co-Mn nano-sized powders from the leachate. Aluminum foil served as a reducing agent here, replacing Ni^{2+}, Co^{2+} and Mn^{2+} ions. In this way, a low-cost agent was used with aluminum foil. In addition, the reduction of waste has been ensured. The recovery yields of Ni, Li, Mn and Co in this method can be listed as 97.5%, 94.05, 4% and 99.3%.

According to Li et al.'s research, NMC cathode powder was leached for 60 minutes, using a 2.5 M H_2SO_4 solution, at 50 °C that contained 5% H_2O_2 at a rate of 25 mL/g (liquid: solid) [128]. The internal yields of Ni, Co, Li and Mn were found to be 97.20%, 99.12%,99.54%, and 99.23%, respectively, following the leaching procedure. Following the addition of dimethylglyoxime (DMG) to the solution, nickel ions precipitated as Ni-$(C_4H_8N_2O_2)_2$ with a 99.35% yield. Subsequently, Co and Mn were extracted with yields of 99.95% and 99.35%, respectively. For the extraction of Co, a solvent consisting of 10% C272 by volume was used at a ratio of 1:1 (O/A) at pH 6. For the extraction of Mn, a P_2O_4 solvent of 10% by volume was used for 10 minutes at a pH of 3.5 in a 3:1 (O/A) ratio. The last step is to remove the metal ions loaded into the organic phase with a dilute H_2SO_4 solution. MnO_2, Li_2CO_3 and CoC_2O_4 have been recovered in the form of. As a result of all the process steps, the extraction yields for Ni, Mn, Li, Co were defined as 96.84%, 92.65%, 91.39% and 81.46% respectively.

In a study by Xuan et al., presented a method to recover the utility of the NMC111 cathode using antisolvent precipitation [128]. [129]. The filtration step was performed with 1.5 M citric acid at 20 g/L (solid) at 50℃. Non-solvent ethanol, acetone, etc. with the addition of dissolved metal ions turn into citrate form and precipitation process is performed at 25 ° C. Precipitation of Li has not been achieved. For this reason, the separation of Li has taken place. Then, acetone was added to the leaching solution in a ratio of 2:1. The recovery yields can be listed as 97.0% Co, 86.9% Ni and 99.7% Mn. The final stage involved calcining the collected sediments at 900 °C and 50 mL/min of oxygen. The outcome of this process was a mixed metal oxide product.

In the part to be explained, deep eutectic solvents (DES) were used. Deep eutectic solvents have high dissolving capacity. Melting points are low. It is also known that it can dissolve different metal oxides. Deep eutectic solvents are also known and used as green solvent in the separation and recovery of metals [129]. Liquid form is accessible at temperatures ranging from ambient temperature to 70ºC. Because of this, it functions as a leaching agent in the hydrometallurgical process' leaching step [130]. It works well for the sustainable and ecologically friendly recycling of used Li-ion batteries.

Luo et al. in his research, deep eutectic solvents based on choline chloride (CHCl) and oxalic acid were used for the recovery of used NMC523 [131]. The precious metals in the NMC523 cathode have completed the filtration process in 20 minutes and at 110℃. Researchers have suggested that filtration systems can be used that allow Li-ions to be selected from water.

Ma et al. in a study he conducted, a deep eutectic solvent based on tartaric acid and choline chloride (TA/ CHCl) was employed to extract the spent NMC cathode [87]. An antisolvent crystallization approach was applied to the leachate by adding ethyl alcohol. The deep eutectic solvent used was leached at a temperature of 70 ℃ and within a short period of time. Thanks to antisolvent crystallization, the precipitation efficiency was realized at a rate of 98.5% for Ni, Co, Mn. The precipitate released because of antisolvent crystallization provided the recovery of DES and antisolvent and was proposed as the starting point for a new cathode.

3.4.2. Recycling processes of NMC battery with pyrometallurgy technique

Pyrometallurgical processes take place because of high temperature reactions. After heat treatment is applied to metal ores, it aims at the recovery of precious metals [132]. As a result of heat treatments, the material undergoes chemical and physical changes. Applications such as drying, calcination, roasting, refining are included in the pyrometallurgical process [133]. The high cost of pyrometallurgical processes and the fact that they are not environmentally friendly due to the gases released during combustion events are considered among the disadvantages of this process.

To recover Li from a used NMC battery, Zhang et al. there is a research that he has done [134]. In the research conducted, it has been proposed to perform a carbothermal reduction process in addition to the multi-step leaching process. Based on the acquired data, NMC was reduced to CoO, Co, Ni, Li_2CO_3, NiO, and $LiAlO_2$ following the reduction process. Li's recuperation was accomplished in three stages. Water leaching was used to first recover the Li salts that were soluble in water. This process's efficiency was found to be 73.80%. Li was subsequently extracted from Li-Al compounds using the alkaline leaching technique. This technique has been shown to have an efficiency of 13.86%. The process of adjusting pH is the final step. Li, which collapses together with aluminum during pH adjustment, has been recovered by roasting process together with water leaching. The efficiency of this process has been determined as 96.3%. The recovery of Li from the NMC cathodes used with a multi-step approach has been achieved with a total extraction efficiency of 87.15%.

A pyrometallurgical approach has been adopted by Yan et al. for the recovery of Li from used NMC batteries [135]. Mn persisted as an oxide at temperatures below 800 ℃. Metal has been reduced to Co and Ni. Mn persisted as an oxide at temperatures below 800 ℃. Metal has been reduced to Co and Ni. It has been transformed into the forms of Li, Li_2CO_3 and Li_2O at temperatures higher than 900 ℃. Li was extracted using a water leaching procedure for 30 minutes at a rate of 5 mL/g (liquid: solid), yielding lithium hydroxide and carbonate. It has been established that this method has a 93% efficiency. Furthermore, the impact of contaminants on Li's recovery rate was examined in this investigation. By using the lithium aluminate synthesis process, it has been found that the Li recovery rate drops in the presence of Al and that Cu has no discernible influence.

Hydrogen gas can be used as a reducing agent. This method is a non-harmful alternative for the environment used in the recovery of precious metals. It does not require the use of a dangerous chemical.

Bhandari and Dhawan's in their study, the recovery of metals from waste NMC powder was achieved thanks to the reduction of hydrogen. Li salts (LiOH, Li_2CO_3) were recovered in 60 minutes and at 500 ℃. Selective recovery was obtained with 91% and 63% efficiency using pure H_2 and 10% H_2+90% Ar. The two methods used to achieve decontamination were pure H_2 and 10% H_2 + 90% Ar. Bhandari and Dhawan observed that the amount of Li recovered is decreased by the FIBER salt that is emitted during Li recovery. They declared that the PVDF ought to be eliminated. Following the removal of PVDF, the H_2 gas's reduction efficiency rose, and in an hour at 500°C, the recovery of Li increased from 63% to 91% [136].

3.4.3. Recycling processes of NMC battery with mechanochemistry technique

Liang et al., have tried a new mechanochemically supported way to recycle the cathode materials used. The cathode material used in this experiment was ground with ammonium persulfate ($(NH_4)_2S_2O_4$) and sucrose in an iron grinding bowl. Then the cathode material obtained was leached. The leaching yields were found to be 97.1%, 94.0%, 87.6% and 93.8% respectively for Li, Ni, Co and Mn. Although this experiment is innovative, recovery processes are required in metals subjected to leaching [137].

Cuhadar et al. they used magnetic separation and buoyancy techniques to remove plastics and precious metals from waste MNC cathodes, and reverse buoyancy technique to separate plastics. Then, the remaining black substance was filtered for two hours at 60 ° C using 2M H_2SO_4 solution at a dosage of 40 g / LH_2O_2.The leach yields for Co, Ni, Mn and Li are respectively %96, %94, %95, %98 it was found as [138].

Qiu et al., he used pretreatment and flotation techniques for the separation of graphite and NMC found in waste LIB. The effect of pretreatment processes on the flotation process has been investigated. The preliminary processes start with 0.5 M Fenton reagent. Roasting is performed at 450 °C for 30 minutes. Under optimal conditions, the efficiency of NMC cathode active materials and graphite was found to be 90% and 75%, respectively [139].

Liu et al. he preferred the process of grinding the Li-ion found in the waste state at low temperature. The low temperature properties of the binder have been investigated. Examining the experimental data, it was found that the electrode plates exhibited great selectivity when manufactured at a low temperature for five minutes as opposed to grinding at room temperature. [140].

4. Prospects for battery recycling in the future

With the increase in population in the world, existing resources are being depleted day by day. It is becoming even more important to use existing resources more effectively and more efficiently [56]. Apart from increasing energy demands, the population growth in the world, together with the changing lifestyle and the fossil fuel resources that are about to be depleted, leads to dependence on renewable energy sources [141]. Considering environmental and socioeconomic concerns, extensive studies have been conducted in the literature to examine the use of alternative fuels, especially in the transportation sector, and lower carbon fuels [142].Since their invention in the 1970s, Li-ion batteries have gained significance in the market for portable electronic devices because of their special qualities, which include a high energy capacity and a long service life. In contrast to other battery types (such NiMH and Pb-acid), LIBS provide less of a harm to the environment. Its lifespan is extended. It is well-designed and better resistance against self-discharges is provided by it. It offers greater output voltage and resistance to high temperatures [143]. Li-ion batteries with these superior properties will be widely used in the production of electric vehicles today and in the future. At this point, the need and demand for minerals in the production of these vehicles and also in battery storage technologies is important. In this context, Li metal in particular is expected to be used 40 times more than cobalt, nickel and graphite [141]. There is a significant gap in the production and recycling of Li-ion battery. Globally, less than 5% of Li-ion are recycled, while in India alone, more than 50,000 tons of Li-ion waste are produced annually. Some non-recycled batteries are used in second-life applications such as energy storage systems [144]. Recent patents show that waste batteries offer high potential for profitable recycling. In the study of Wang et al., it was stated that pyrolysis achieved an electrolyte removal efficiency of almost 100% [56]. However, it will also release a lot of dust and harmful fumes that include fluorine and organic materials that need to be treated. Another method for handling used electrolyte is solvent extraction. Currently, there are two types of solvent extraction, which are used to process the used electrolyte. The first is organic solvent extraction. To be used in the production of new batteries; A technique has been developed to recover Ni, Mn, Co, and Al metals from Li-ion batteries. At this point, it became possible to reuse the cathode active material following thermochemical reactions. Tran et all,. ın a study conducted by; Regarding this issue, they suggested converting Li-ion batteries using green chemicals and technologies. With their study, they showed that high extraction efficiency can be achieved with an environmentally green method using deep eutectic solvents at low temperatures[56].

4.1.Recovery of Li-ion battery anode materials and electrolytes

4.1.1. Electrolyte recovery

In a Li-ion battery, the electrical insulation between the decoupled electrodes is shielded by an ion-permeable membrane. An effective ionic conductor can also be an electrolyte. Lithium hexafluorophosphate, which dissolves in carbonate solvents, has been stated to be the best electrolyte for Li-ion batteries[56]. There is a significant degree of fluctuation in the conversion rates reported in the literature; although there is a lot of variability from one region to another, there are still low rates. As an example, Call2Recycle, the organization responsible for collecting Li-ion used in Canada, reported that less than 200 tons of Li-ion battery were collected in 2017. Gies et al., while this amount corresponds to a collection rate of 25%, it is believed that their assessment is an overestimate when considering the sales volumes of electronic and electric vehicles (EV) [56]. Wang et al. based on statistics on the sale of electronic devices as a destination markup, the recycling rate in North America is estimated to be lower than 10% in 2012. To look at another comparison, the collection rate observed in Europe in 2016 by Eucobat was reported to be slightly higher than 15% [105]. Based on these data and the inability of existing pyrometallurgical processes to recover Li, it is estimated that there is a much lower rate of recycling Li from spent Li-ion battery than 1%, which clearly does not comply with the principles of sustainable development. For this reason, it has become mandatory to increase both the collection and recycling rates of Li-ion battery [143].

4.1.2.Recovery of anode materials

To get rid of spent lithium and use it more in new Li-ion batteries, it needs to be more economical and sustainable. Redeployment is not just an anode in libs (lithium-ion batteries), it is more than that. It has also been said that the recovery of graphite holds great promise for use in the fields of supercapacitors, catalysis and water treatment. In addition, the recycling of graphite from waste Li-ion batteries will also have a great impact on the protection of the environment. In light of all this, one of the most interesting transformations in recent years has been the recycling of waste [143].Water infiltration has been demonstrated to be a successful technique for extracting graphite from anode components in a spent Li-ion battery by Bhar and associates. In this instance, an organic solvent based on dimethyl carbonate was used to first wash the recovered graphite. After that, the graphitization time was extended and the intermediate phase of leftover carbon black and white electrolyte that had formed as a result of electrolyte on the graphite surface decay was removed by post-treatment at 750 °C[145].

5. Conclusion

The reasons for the selection of these materials are that the materials used in battery production are considered as strategic raw materials and the use of batteries has increased and therefore the rate of waste batteries has also increased. In addition, the cost of battery production has decreased considerably due to the increased use of batteries. While the cost of Li-ion battery production was 1183 dollars per kilowatt in 2010, this value decreased to 137 dollars in 2020.

Waste has become a big problem today. Therefore, recycling is of great importance. In this article, the recycling of Li-ion batteries, which have been frequently mentioned in recent years, is discussed. Procedures and techniques used in the recycling of Li-ion battery types including LFP, LCO, NMC and LMO batteries are discussed. These methods are categorized under 3 headings: hydrometallurgy, pyrometallurgy and mechanics. Efficiencies to be obtained from recycling in these methods are mentioned.

It is aimed that this article, which provides information about the recycling of Li-ion batteries, will contribute to the studies and research of the readers.

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The authors dedicated this publication to the 100th anniversary of the Republic of Türkiye. As scientists raised by Türkiye, they are proud to be citizens of this country.

Author Contribution

T. K., H. K., M. G., G. S., C. A., C.A., I.H., E. H., R.B., M.A., M.B.; Collected Data, Wrote the Paper. **T. K., M.B.;** Conceived and designed the analysis**. M.A., R.B.;** Performed the analysis. **H. K., M. G., G. S., C. A., C.A., I.H., E. H.;** Contributed Data.

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