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An overview on Lithium-ion batteries recycling processes

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Abstract. In the last few decades, well-developed lithium-ion batteries (LIB) with high capacity showed great performance in electric vehicles. However, efforts must be made in the field of LIB recycling to reduce the negative effects caused by spent LIB on the environment. The processes for lithium-ion batteries recycling are presented through three different portions includeing pre-treatment, secondary treatment and further treatment. In pre-treatment, the LIB is firstly discharged and disassembled manually. Secondly, the unit cells are shattered and separated into black mass mainly by physical properties through secondary treatment. Lastly, the black mass is decomposed into raw materials for recovery or resynthesizing in further treatment. Interdisciplinary approaches and secondary use feasibility are also discussed for future application prospect.

1. Introduction

The wide use of high energy, lightweight, rechargeable lithium-ion batteries has mitigated the increasing power demand of electronic devices and automotive vehicles for the last few decades. On the other hand, the recycling process of LIB also needs close attention due to the high cost of raw materials, safety concerns toward stockpiled batteries, and environmental concerns regarding LIB waste which is hazardous to the nature[1]. The recycling of lithium-ion batteries (LIB) aims to recover components such as Li, Ni, Co, Mn, Al and Cu. It is estimated that the global LIB recycling market will grow from $12.2 billion in 2025 to $18.1 billion by 2030[2]. However, since Sony commercialized the first LIB in the 1990s, there has been challenges to develop a sustainable LIB circular economy. The various cell structure, complicated system design and chemicals required for the whole process have caused the in-fancy of LIB recycling industry. It is reported that in 2016, only 5% of LIB were recycled in the UK, and the other 95% ended up in landfill[3]. The United States has not established a standard procedure for LIB recycling as a result of the vast varieties of batteries need to be recycled by different processes to avoid potential danger[4]. Chinese government has been popularizing renewable energy vehicles since 2013, and it is estimated that there will be a massive retirement of automotive LIB in 2021 consequen-tly. The annual spent batteries will reach 500,000 tons and goes up to 1.16 million tons in 2023[5]. On the bright side, EU set 25% minimum LIB collection rate in 2012 and 45% in 2016. Twelve European Economic Area countries have achieved 45% recycling rate by 2015. Switzerland set 71% LIB collec-tion rate in 2015[1].

As a result, the recycling process of LIB needs to be taken into serious consideration. A standardized recycling process are crucial to improve the sustainability of LIB, in which case the process applies to the most or at least a part of spent LIB, gather the most valuable parts of the battery and achieve separation to the greatest extent under an acceptable component purity. Currently, the recycling process of LIB could be divided into three steps: pre-treatment, secondary treatment and further treatment[5]. The pre-treatment mainly focuses on the fully discharge of the battery and manually disassemble; the

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secondary treatment includes comminution and the first-time separation driven by mostly different physical properties of the cell; the further treatment applies a wide range of physical and chemical methods to achieve the premium separation and recycling rate. Herein, we present a detailed discussion on this widely accepted and industrialized three-step process, as well as a series of progressive and innovative methods on the topic of LIB recycling.

2. Pre-treatment

In the pre-treatment process, battery cells are firstly disassembled from spent batteries manually, then discharged through different approaches. For safety sake, certain approaches may require discharging before manually disassembling. It is also arguable that to what extent the battery cell should be disassembled. A wide range of laboratory studies claim that electrodes should be separated and collected prior to secondary treatment[6-9]. However, in industrial practice few examples were seen since this pro-cess could be extremely time and labor consuming. In most cases, the outcoming product of pre-treatment is a combination of the battery shell, electrodes, electrolyte, adhesives and separator. As for discharging, several common techniques as well as their pros and cons are presented below.

2.1. Electrical discharging

In electrical discharging, spent batteries are connected to a resistor in a close circuit to release the remaining electricity. Electrical discharging has relatively low operating difficulty and security risk. Besides, there have been arguments about whether the electricity recycled through this process could be collected and make up some of the capital cost. According to Sonoc et al., the energy can be collec-ted through electrical discharging are less than 1% of the total capacity[10]. Due to the uncertainty of the state and residual energy of the spent LIB, the idea of obtaining residual electricity is still question-able.

2.2. Brine or salt solution

LIB cells could also be immersed into brine or salt solution to achieve self-discharge. In this case, the batteries need to be further disassembled into single units to improve efficiency. Despite the labor cost that could be saved and economic benefits in this process, studies have shown the potential risk for further disassembling the battery into small units[6, 7]. A great many of damaged structures and unevenly distributed energy zoon is included in spent LIB, such as regions being open circuit and short circuit. Although these areas could be detected by current or voltage meter, areas where energy is unevenly distributed could still cause danger to the operator owing to overcharging effect. Additionally, highly concentrated charge on both electrodes could lead to surface corrosion and make the cell surface rusty. The waste effluent will also need cleaning after discharging.

2.3. Cryogenic processing

Cryogenic processing is believed to be a safe but relatively energy consuming process. Liquid Nitrogen is used to solidify the electrolyte and lower the reactivity of the active components in LIB. It also reduces the chance of combusting and explosion during the disassembling process. Nevertheless, the scalability of cryogenic processing is limited since it takes up a considerable amount of energy to maintain low temperature[11, 12].

2.4. Thermal processing

D. Lisbona, C.K. Lee and Granata heated the LIB cell to 100-150 ℃ for 1 hour to discharge prior to shredding[13-16]. Through this process, the cell will be loosed and easy for unpacking. However, if the cell is not fully discharged, thermal runaway might occur. It is also reported that the electrolyte is irreversibly damaged, and hazardous gas is released. This process was once widely used for recycling LIB with high Ni and Co concentration since it is easy to operate and the purity of metal that could be recycled is elevated, even though it might cause secondary pollution. According to G. Zhang et al., to fully remove the electrolyte and separator, a temperature above 400 ℃ is required[17-20], which consu-mes more energy. It is also notable that thermal processing will totally burn down the anode material for LIB

using graphite as anode. European Commission claimed that Cobalt and graphite were at great supply risk thus need to be properly recycled for future sustainability[1, 21]. Thermal processing grows narrowly applied recently due to the fluctuation of noble market price and environmental concerns[2].

Table 1. Different stabilization techniques in comparison[3].

Stabilization technique	Advantages	Disadvantages
Electrical discharging	Potential for energy recovery	Difficult to scale energy recovery
Brine or salt solution	Low cost	No energy recovery, Waste effluent may require cleaning
Cryogenic processing	Safe, even in cases of metallic Li or Li plating	Energy intensive, high cost, poor scalability
Thermal processing	Degrades binders and removes volatile components simultaneously	Electrolyte cannot be recovered, produces HF

3. Secondary treatment

3.1. Comminution

The comminution process typically uses a granulator, a shredder or a hammer mill. The granulator rotates fast, and the gear blades are fine. The static friction between materials and blades provides centripetal force to keep the material inside the granulator. As the material being granulated into smaller particles, the static friction continues to decrease until the maximum static friction cannot provide enough centripetal force to hold the material inside. As a result, the tiny particles will fall off. In comparison, shredder and hammer mill rotate at a low speed, and the outcome particles are larger. Therefore, fewer laboratory studies designate shredding and hammer mill as treating methods.

The comminution environment is also critical since the cathode materials are likely to be oxidized once exposed in the air. Wet comminution and inert gas comminution are commonly used for protecting the cathode materials. Wet comminution is a low-cost method which could also separate the electrolyte simultaneously. It also provides convenience for the following separation process, which will be discussed in later sections. The disadvantage, on the other hand, is like the use of salt solution which requires effluent cleaning. Inert gas comminution consumes more resources which makes it less popular than wet comminution. Furthermore, fire will not stop once started during crushing and smashing, so the whole process needs to be supervised.

3.2. First-time Separation

First-time separation mechanics is mainly based on the physical properties of the material, such as particle size, magnetism, density and hydrophilicity/hydrophobicity (Fig. 1). In size separation, the materials are fed to a series of vibrate sheets with porous structures. Each sheet has a certain size for filtering. After multiple degrees of separation, coarse particles will remain on top while fine particles at the bottom. Magnetic separation uses a conveyor belt and magnetic wheel which intermittently gains and loses magnetism to separate the material. As the material is delivered to the “cliff” on the conveyor belt, the magnetic wheel below gains magnetism to make sure only substances without magnetism are filtered. As the materials go underneath, the wheel loses magnetism to collect materials with magneti-sm. Density separation uses a liquid phase or gas phase with a designed density to separate materials. In the carrying fluid phase, materials with higher density will sink, and lighter ones will come to the surface. Froth flotation takes advantages of hydrophobicity/hydrophilicity of materials. A bubbled sol-ution is created through constant stirring, and materials are added to the solution. After a while, hydro-philic materials will float up together with bubbles as hydrophobic materials sink to the bottom. Depe-nding on different separating methods, the produced components are also different, thus could be divi-ded and utilized into various further treatments.

4. Further treatment

Further treatment requires the cathode materials collected by previous steps. The treating methods are not limited compared with pre-treatment and secondary treatment. Currently, three main types of LIBs are on the market, including $LiCoO_2$, $LiNi_xCo_yMn_{1-x-y}O_2$ and $LiFePO_4$. Among which common treating methods such as precipitation, extraction, electrochemical deposition and ion exchange are applied[5]. Additionally, several newly discovered techniques also include recovery and secondary use of the spent cathode materials which will also be mentioned in later section. Compared with $LiNi_xCo_yMn_{1-x-y}O_2$ and $LiFePO_4$, the recycling process of $LiCoO_2$ is well

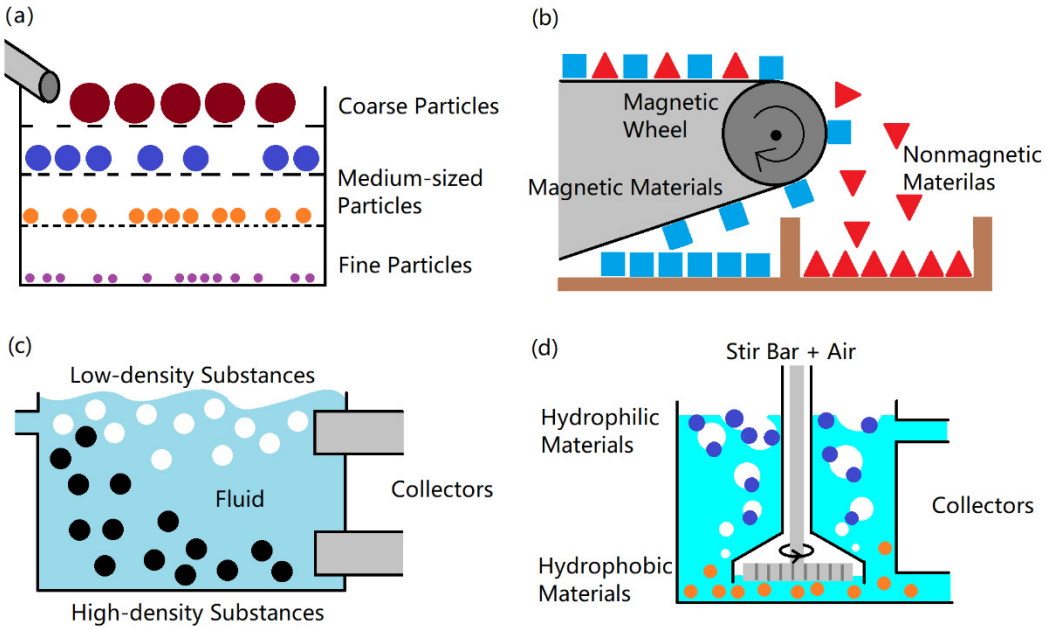


Fig. 1. (a) Size separation; (b) Magnetic Separation; (c) Density Separation; (d) Froth Separation.

developed, as a result of early discovery and long-term usage. However, with the improvement of battery capacity and energy density, $LiNi_xCo_y$-$Mn_{1-x-y}O_2$ and $LiFePO_4$ are replacing $LiCoO_2$ as the market domain. As a matter of fact, cathodes with multiple metal components are much harder to handle, and it is because of the various types of cathode components that the recycling process must be discussed separately.

4.1. LiCoO2

Acid leaching is believed to be a simple and high-efficiency method for $LiCoO_2$ separation. Li Li et al. reported an electrochemical approach to recover the $LiCoO_2$ film using nitric acid as leaching agent followed by an electrochemical deposition process in 2011[8]. Despite the good cycling performance, this method could hardly be applied at large scale or for industrial process. The same research group used 1.25 mol/L ascorbic acid under 70 °C resulting in a recover rate of 94.8% for Co and 98.5% for Li in 2012[7]. Due to the reducibility, ascorbic acid played the roles of both dissolving agent and reduc-ing agent. M. Wang et al. used a mechanochemical method which went through a co-grinding process of the cathode material together with EDTA to form water-soluble chelates Li-EDTA and Co-EDTA in 2016[6]. Followed by the addition of NaOH and Na_2CO_3, a recover rate of 98% for Co and 99% for Li was achieved. An outstanding solid-state thermal treatment was also discovered in 2016 by Shi Chen et al[9]. In this process, after filtering, washing and drying, the cathode materials were simply mixed with Li_2CO_3 followed by a thermal treatment at 850 °C. With the addition of only 5 wt.% of Li_2CO_3, a total recover ratio of 95.78% was achieved. One disadvantage for recovery processes, however, is that the atom ratio of several metal components needs to be adjusted which requires extra efforts in recyc-ling practice.

4.2. LiNixCoyMn1-x-yO2

Rujuan Zheng et al. designed a close-loop process for $LiNi_xCo_yMn_{1-x-y}O_2$ recycling in 2017[22]. With acid leaching, pH adjustment, the addition of Ni_2SO_4, $CoSO_4$ and $MnSO_4$ followed by a coprecipitation process, the products could be directly resynthesized into $LiNi_xCo_yMn_{1-x-y}O_2$ cathode. This process is possible to separate the mixture of $LiNi_xCo_yMn_{1-x-y}O_2$ and $LiFePO_4$, which is of great significance to the future of unclassified battery recycling. S. Anwani et al. used a collective method to compare the product purity, overall process efficiency, environmental and economical indices of 10 different acids including sulfuric, hydrochloric, nitric, phosphoric, formic, citric, oxalic, DL-malic, ascorbic and tarta-ric acids[23]. The separation process is shown in Fig. 2, and 10 different acid leaching towards the same cathode material was studied. The result showed that oxalic acid-based recycling process is the premi-um selection.

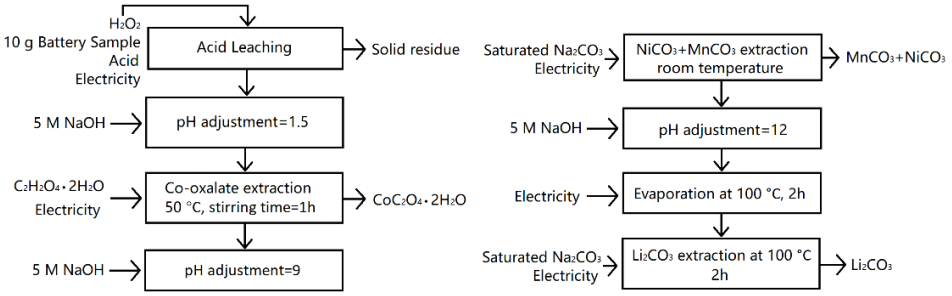


Fig. 2. Flow chart for component separation[23].

Besides traditional methods, an innovative biomass pyrolysis treatment using macadamia shells as biochar is presented by Yunze Zhao et al[24]. According to the author, the cathode & macadamia powder was roasted at 500 °C for 30 min then roasted at 750 ℃ for 25 min. The $LiNi_xCo_yMn_{1-x-y}O_2$ cathode material went through a series of biochar reduction reactions as is shown in reaction (1)-(6). Subsequently, a series of filtering and precipitation processes were applied and a purity of 99.98% for Li_3PO_4 could be obtained. The interdisciplinary study has certainly provided great potential due to reduced raw material and operating costs, moderate energy consumption and excellent separating efficiency.

$$Li\left(Ni_{\frac{1}{3}}Co_{\frac{1}{3}}Mn_{\frac{1}{3}}\right)O_2 \xrightarrow{biochar} \frac{1}{2}Li_2O + \frac{1}{3}NiO + \frac{1}{3}CoO + \frac{1}{3}MnO_2 + \frac{1}{12}O_2 \quad (1)$$

$$2MnO_2 + C = 2MnO + CO_2(g) \quad (2)$$

$$2NiO + C = 2Ni + CO_2(g) \quad (3)$$

$$2CoO + C = 2Co + CO_2(g) \quad (4)$$

$$2Li_2O + CO_2 = Li_2CO_3 \quad (5)$$

$$Li\left(Ni_{\frac{1}{3}}Co_{\frac{1}{3}}Mn_{\frac{1}{3}}\right)O_2 + \frac{7}{12}C = \frac{1}{2}Li_2CO_3 + \frac{1}{3}Ni + \frac{1}{3}Co + \frac{1}{3}MnO + \frac{1}{12}CO_2(g) \quad (6)$$

There are also studies showing that the spent cathode material could be utilized for Oxygen Elevation Reaction (OER) without further treatment. Luyang Wang et al. used a thermal treatment and dispersed the cathode material with alkaline ion exchange membrane into N-methyl-2-pyrrolidone (NMP) to form an ink solution, which was then dropped on a glassy carbon electrode, dried under infrared light and used as the catalytic active substance for CO_2 reduction[25]. The result showed that the recycled material had a pristine OER performance of 80% of the pure, raw materials. It was also believed that the atmosphere during thermal treatment and the mole ratio of Ni, Co, and Mn were crucial to the catalytic performance.

4.3. LiFePO4

Qifang Sun et al. has reported a similar thermal treatment using Li_2CO_3 to recover the $LiFePO_4$/C component as is mentioned previously in 3.1[9, 26]. The cathode material is firstly sintered at high temperature to form Fe_2O_3 and $Li_3Fe_2(PO_4)_3$, after which $LiFePO_4$/C is resynthesized with the addition of 1.4 wt.% Li_2CO_3. Furthermore, as a similar way to the EDTA method mentioned in 3.1, Li Li et al. also used a mechanochemical approach but with different reaction agents in which a co-grinding process, filtering process and precipitation process were involved[6, 27]. In contrast, citric acid was used as the co-grinding agent and H_2O_2 was used for the following extraction. The extraction efficiencies of Li and Fe are 97.82% and 95.62%, respectively. Apparently, the recycling difficulty for $LiFePO_4$ is relatively low, and the process resembles that of $LiCoO_2$ in many aspects.

At the same time, the secondary use of LIB is also highly recommended especially for countries like China where the consumer market is rapidly growing whereas the effective path of secondary use has not been established[5]. Bingxiang Sun et al. has developed a thorough study regarding the $LiFePO_4$ LIB secondary use in power load peak shaving[28]. Practical parameters such as battery purchasing, scre-ening, regrouping, operation, maintenance, and replacement costs are discussed in detail to analyze the economic feasibility. The author concludes that if only the batteries could go through a rough selection process to eliminate the severely damaged portion, for most LIBs that are down to 80% of its initial capacity, the recycled LIB could achieve Net Present Value (NPV)>0 for 20 years, and Dynamic investment payback period (DIPP) of 4 years.

5. Conclusions

A standardized LIB recycling process has not been established which requires close attention due to the rapidly growing world LIB market and its environmental effects. We present a study towards the traditional approaches for pre-treatment and secondary treatment as well as newly discovered methods for further treatment. Herein, initial battery designs that could provide convenience for future recycling are recommended such as using soluble adhesives and electrolytes with low toxicity. Interdisciplinary studies and secondary use for LIB showing great future potential are also strongly encouraged.

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