Journal of Engineering Science and Technology Review 15 (1) (2022) 74 - 84

JOURNAL OF
Engineering Science and Technology Review

www.jestr.org

Review Article

A Mini Review on The Recent Progress on The Method of Recycling Lithium-Ion Battery: Pros And Cons In Environmental and Economical Aspect

Gusti Putu Surya Govinda Atmaja[1], Novat Pugo Sambodo[2] and Muhammad Akhsin Muflikhun[1,3]*

[1]Mechanical and Industrial Engineering Department, Faculty of Engineering, Universitas Gadjah Mada (UGM), Indonesia
[2]Department of Economics, Faculty of Economics and Business, Universitas Gadjah Mada (UGM), Indonesia
[3]Center for Advanced Manufacturing and Structural Engineering (CAMSE), Faculty of Engineering, Universitas Gadjah Mada (UGM), Indonesia

Received 1 February 2022; Accepted 6 May 2022

Abstract

Since the last decades, the demand for lithium-ion batteries (LIBs) is going rapidly. Many applications used lithium-ion batteries as a power source in electric vehicles, solar panel, wind turbine, power backups (UPS), laptops, cellphones, electric toys, and general consumer electronics goods. The huge demand of LIBs in large number can create a supply risks for natural lithium (Li) source reserves. In general, after the service life of LIBs, to keep the source of components that contains in LIBs, a proper of recycle process is mandatory. Recycle of LIBs purposed to alleviate limited resource in nature. Moreover, recycling is an excellent solution to reduce and prevent potential environmental pollution caused by improper disposal of LIBs. Hence this motivated many researchers to recycle LIBs in recent years. Considering the actual methods to recycle LIBs, the present study summaries and listed the methods that used by researchers and industries that are proven can be used to recycle LIBs with the pros and cons followed in each category. The study also comparing different methods that can give suggestions for improvement related to effectiveness and efficiency in the future, for a better environment and cost efficiency.

Keywords: Recyclng processes; Environment; Lithium-Ion battery; Resources; Valuable metals

1. Introduction

The Nobel laurate 2019 announced that the winner in the chemistry categories goes to John B. Goodenough, Akira Yoshino, and M. Stanley Whittingham for the development of lithium-ion batteries (LIBs). The Nobel committee argues that the development of LIBs gives a better ecosystem and condition for a wireless and fossil fuel-free society, and researchers that these impacts significantly benefit humankind [1–5]. LIBs have been widely used since the 1990s in portable electronic devices to electric vehicles due to long storage life, compact, lightweight, high energy density, wide range used in different temperature conditions, and low self-discharge efficiency [6]. In the past two decades, the production of LIBs increases rapidly. In 2000, up to 500 million cells produced when the annual production increased 800% in the next 10 year. Six years after 2010, the production of LIBs reached 7.8 billion that increased from 20115 with 5.6 billion. It is estimated that in 2020 the production can exceed 25 billion units with the total weight up to 500,000 tons [7,8]. In specific demand, the main element composed for LIBs, lithium used in modern type of battery due to high electrochemical potential and have low density with around 0.534g/cm3[9]. The demand of lithium has increased more than 85% from 265 thousand tons in 2015 to 498 thousand tons in 2025. This condition can make a supply shortage in the future [10–15]. The future of LIBs as an energy storage will remains increase since the wider application in automotive and structures that used electric as main source power. Moreover, the electric usage can be a solution for electricity lack problem in remote areas [16–21]. Hence, the stock as reverse is limited. This condition pushes many researchers and corporations around the world to recovery of lithium by recycle and reuse of it from all possible resources.

The limited amount of lithium in the world greatly impacts the research related to the recycling of LIBs. The focus of researchers is not only on how to reuse and recycle lithium as main material used in LIBs, but also how to recycle all its constituent elements. This motivation is triggered by the fact that in average, from 4000 tons of spent LIBs, it contains 27.5% of heavy metals and more than 10% of toxic electrolytes. Generally, spent LIBs contains of 5-20% Co, 5-10% Ni, 5-7% Li, 5-10% several metals such as Cu, Al, Fe, etc., around 7% Polymers, and the rest is organic compound [22]. The composition may differ from different manufacturers. With unproper recycling and reuse of spent LIBs, its waste can harm the environment and may infiltrate the soil, create serious environmental pollution, and risk human life. Moreover, to destroy spent LIBs by burning process is also not a solution since it will release poisonous gases such as Co and HF [23,24]. Regulator has made several policies to prevent unproper recycle and reuse process of spent LIBs. For example, EU in 2015 introduce a law to the member countries to recycle LIBs between 15-25% with the efficiency should reach 45-50%. Beside good for environment, several element in LIBs such as Li, Ni, and Co have high price in the market and can boost the economic benefits [22,25].

Based on the point of view background mentioned in the above, recycling and reusing of LIBs. For the researcher itself, optimizing the recycling process of LIBs can create less pollution and prevent harm gaseous that may contaminate human life. Nevertheless, the challenges and difficulties for

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*E-mail address: akhsin.muflikhun@ugm.ac.id
ISSN: 1791-2377
doi:10.25103/jestr.151.10

the recycling process remain and need to be resolved. This action is not easy since the diversity of the elements used in LIBs make the recycling process more complicated. In LIBs, the basic components generally consist of Cathode (LiMO2, LiFePO4/Al), Anode (Graphite/Cu), Electrolyte (LiPF6, EC, DMC, EMC, DEC, etc.), Separator (PE/PP), and Case (Al/SS/Polymers) [22,26]. Although the challenge and difficulties, the opportunities also appeared such as economical reason and benefits from the urban mining is coexist.

The recent article provides a progress report of the recent advances of the recycling and reusing process of spent LIBs. By reviewing previous research, this study can give the most up to date knowledge about recycle process with the latest technology, comparison between recycle methods, and overview of the method in lab-scale and industrial experiences. The present article also presents the development of alternative recycle methods that have environmental-friendly and sustainable processes.

2. Lithium-ion battery (LIB) structures and manufacturing process

The structure of LIBs can be arranged into 4 main important parts: anode (negative electrodes), cathode (positive electrodes), organic electrolyte and separator. Usually, the material used in anode is graphite with a maximum discharge capacity of 372 mAh g-1. Graphite was chosen as an anode material due to its layered incorporation of ions (Li +) into a solid form. In a recent study, to gain better properties, an anode assembled from a copper foil that coated with several materials such as graphite, a conductor material, a poly-vinylidene fluoride (PVDF) and so forth. A cathode arranged from LiMO2 that is coated with aluminium foil together with conductor material, a PVDF, and other materials [27]. An electrolyte solution consists of LiPF6, LiBF4, or LiClO4 that dissolved in an organic solvent (DMSO, PC, and DEC). A recent study conducted by Marcinek et al. [28] studied electrolytes that usually consist of LIBs. There are several electrolytes in LIBs that are reported have been used: organic carbonates and esters liquid electrolytes (EC, PC, BC, NMO, DMC, DEC, EMC, EA, MB, EB, γBL, and γVL), organic ethers liquid electrolytes (DMM, DME, DEE, THF, 2-Me-THF, 1,3-DL, 4-Me-1,3-DL, 2-Me-1,3-DL), polymer electrolytes (LiTFSI, LiBETI, LiFAP, LiTFAB, LiBOB, LiBBB), ionic liquid electrolytes, and other approaches.

Since the LIBs manufacturing policy became stricter, the hazardous material that potentially can create pollution for soil and air should be treated carefully. To fulfill this procedure, at now a day, the quality of LIBs became more greener and cleaner than other types of energy storage devices due to an improvement of new materials that have better environmentally friendly.

Several studies have successfully used the integration model of the supply chain model in the manufacturing process of lithium-ion components [29–32]. Hou et al. [31] used waste tires to be used as electrode materials. The method was used waste tires that have been carbonized and coated with sucrose. This combination then further processes to be nano sulfur carbon (NSC) and carbon-coated nano sulfur carbon (NSC/C). The results show that by using waste tires materials, the battery's performance was excellent, and the process can fulfill the requirement of the battery component and have advantages in the economical and environmentally friendly process.

In the market, generally there are 3 common types of lithium-ion cells: cylindrical, prismatic, and pouch. Before the final types spread to consumers, the electrode fabrication begins with manufacturing process of electrodes. The process to manufacture of electrodes is shown in the Fig. 1.

Fig. 1 Electrode fabrication process.

The fabrication process of LIBs has an important component namely separator. Its function is to isolate cathode and anode to prevent short-circuiting that happen when the electron passes through freely from the positive and negative electrodes. It is well common knowledge that the ionic conductivity has directly influenced the battery's performance. The basic structure of an electrochemical in cathode and anode can be seen in Fig. 2.

2.1. Cylindrical cell fabrication

The common structure of cylindrical cell of LIBs can be seen in Fig. 3. The fabrication process of cylindrical LIBs starts by combining two electrodes using a winding machine. To avoid short-circuits, between cathode and anode layers inserted by separator. After winding process, the cathode and anode layer then taped and rolled to keep it tightly. The rolled layers then measured its impedance to ensure no short-circuits occurred. The process then continued by inserted the materials into the cylinder with hollow mandrel. Circular insulator then placed on the top while the anode tab is placed at the bottom and cathode is placed on the top. Later, an annular groove is made by using grooving machine and the cell is sealed by crimping. The CID and PTC then paced into the hollow and then ultrasonic welding is applied. The cell then filled with electrolyte using a vacuum injection process to ensure the electrolyte distribution before the cell is sealed. The dry condition is set to prevent moisture change. The cell then gains final check and clean using electrolyte leakage test and alcohol to remove any debris [6,33,34].

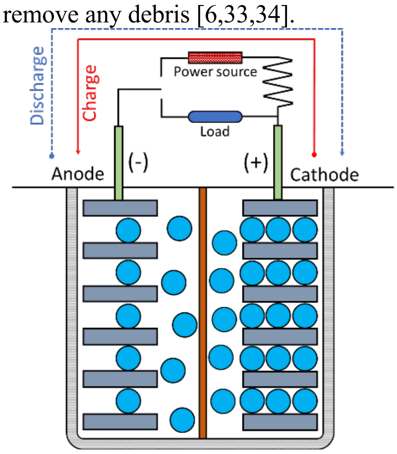


Fig. 2. General structure of LIBs with electrochemical power source.

2.2. Prismatic cell fabrication

The first step to manufacture LIBs with prismatic type is generally similar to cylindrical cell structures where the binding and applied separator between cathode and anode were applied. The difference between cylindrical and prismatic is that there is absence of electrode tab in prismatic. Instead, the electrode terminal is directly connected. To prevent short-circuiting, the asymmetric layups is applied using ultrasonic welding. After that, the core of the cell is taped. There are 2 different case, where one is metal that have good conductivity such as aluminium. In this case, the surface is lined with nonconductive material. Before the cell is placed in the case, the electrolyte is filled inside using vacuum injection technique. The through case then sealed before final inspection and cleaning procedure [36].

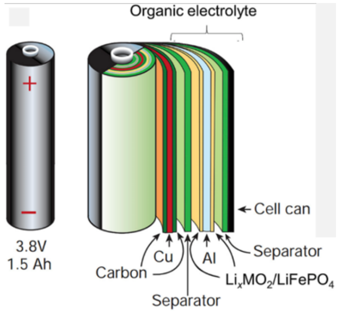


Fig. 3. Typical cylindrical cell structures of LIBs. Ref. [35].

2.3. Pouch cell fabrication

Typical structure of pouch cell of LIBs can be seen in Fig. 4. The system consists of multi layers of cathode and anode that separated by separator layer. Since the number of layers does not limit the system, the number of layers differs from each case due to the manufacturer and application sectors' purposed. Unlike the manufacturing process of cylindrical and prismatic cells, the pouch cell manufacturing process consists of cathode and anode connected by endless separator until the number of layers is achieved. After the separator was cut, the laminate was taped and then arranged together based on cathode and anode tabs before welded together. The laminate was then inserted in the aluminum layer coated with insulator to prevent short-circuits. The humidity and other debris then removed in the vacuum condition. The remaining unsealed part is used for filling electrolyte using injection technique. After that, vacuum-sealed is applied for better penetration of electrolyte. The second sealed process is applied by discharge the gas that may be filled inside the case to prevent exhausted gases produced during the initial charge. Final process was then applied with a leakage test and cleaning [33,37].

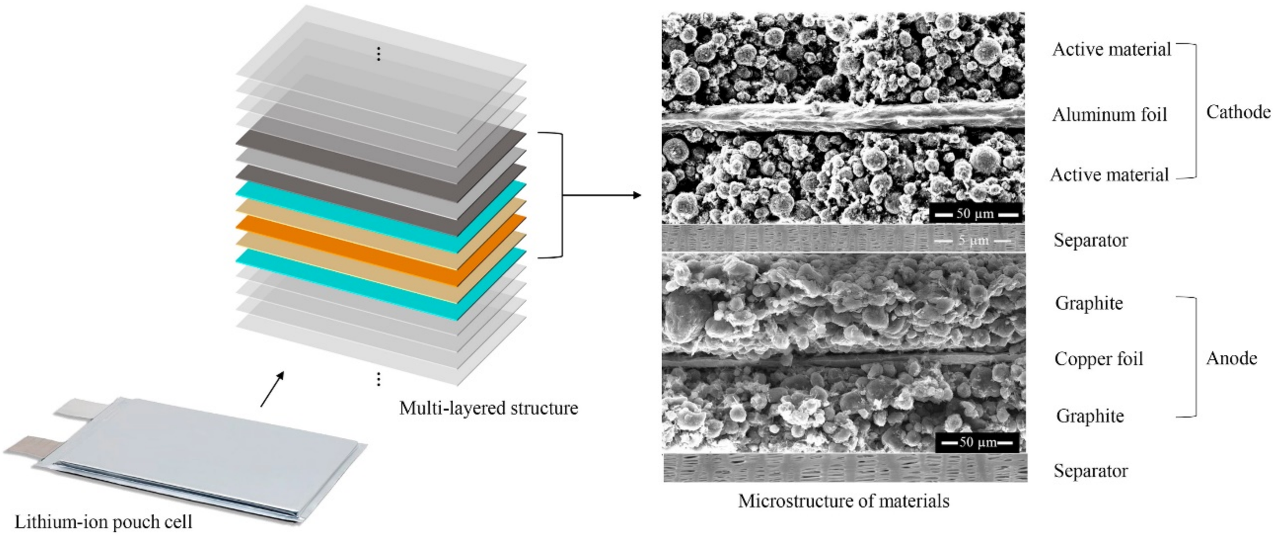


Fig. 4. Structure of pouch cell LIBs with microstructure layers. Ref. [38].

3. Pretreatment process

The pretreatment process to recycle LIBs are aimed to discrete elements and components based on the different properties. However, before separation is induced, it should be discharged first to prevent short-circuit that may occur in spent LIBs. Several methods reported have been used to discharge spent LIBs. In general, there are 2 techniques to discharge spent LIBs: chemical and physical. In chemical discharging technique, it is well known to use a salt solution ($NaCl$, $FeSO_4$, $MnSO_4$). In physical technique, graphite and copper can be used [39–41]. The different discharge methods with each element's composition are shown in Table 1. According to previous study conducted by Yao et al. [40], the best discharge solution are occurred for $NaCl$ solution and $FeSO_4$ solution where if it related to environmental issue, $FeSO_4$ solution have advantages compared to $NaCl$ solution.

After discharging process is obtained, pretreatment process is proposed to separate materials, and metallic scraps of spent LIBs. There are two types of pretreatment after the discharge process of spent LIBs: manual and automatic/semi-automatic. In manual technique, spent LIBs case dissembled using cutting devices like a screwdriver, knife, and plier. In many cases, such as cylindrical type of LIBs, using cutting devices can easily split the component of LIBs into several parts such as metallic or polymer case, anode, and cathode foil. In small-scale recycle LIBs, manual technique can give high purity metal and polymer material. After dismantling, the metal case can be easily collected and then recycled or reused for other purposes. Nevertheless, in large scale process and efficiency purposes, many recycle center and researchers used automatic or semi-automatic technique by directly crushing spent LIBs into smaller size and then used magnetic to separate metallic parts. The smaller parts then can be further processed to separate electrode material from Al, Cu, plastics. In this step, there are segmented component that called plastic fraction, attaching parts, and leaves electrode (battery cell materials) scraps or powder-like [34,42]. Separation process during pretreatment can absorb more than 20% of LIBs total weigh. Fig. 5 shows LIBs parts that can be separated into 5 different parts based on the study of Georgi-Maschler et al. [43].

Table 1. Discharge effect to LIBs with different materials [40]

Type	Element	Time of discharge (min.)	Sediment (Element)	Supernatant (Element)	Output Gas (Element)	Stability
Chemical	NaCl	30	Fe, Al	P, Li, Co	N_2, CO_2, H_2O, C_xH_y, $CH_3OCOOCH_3$	Stable
	MnSO4	140	-	Ni, Al, Fe, Mn	N_2, CO_2, H_2O	Stable
	FeSO4	5	Fe	Cu, Fe	N_2, CO_2, H_2O	Stable
Physical	Graphite	14	-	-	-	Unstable
	Copper	153	-	-	-	Unstable

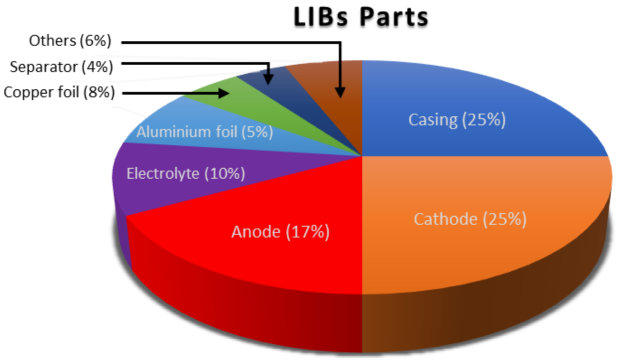


Fig. 5. Average LIBs material content.

4. Recyling methods

The recycling classification methods to recycle lithium-ion batteries listed in the current study was summarized based on the previous research. The process of recycle is focused on the recycling cathode, anode, and electrodes materials that contains many valuable elements

4.1. Wet and Dry crushing methods

The comparison between dry and wet crushing methods was evaluated by several researchers [44–47]. The recycle process give output materials with clustered into 2 different particles. Aluminium and copper foil, plastics, and diaphragm fiber resulted in coarse particle. Zhang et al. [47] used electrode materials (graphite and LiC_OO_2) composed as fine particles. The result shows that using wet crushing methods gives less efficiency due to water flow that carried fine particles and lost it. The wet crushing method also has problems from the water containing hazardous materials (LIBs electrode materials). This contaminated water needs further action to recycle process second.

4.2. Thermal treatment + hydrometallurgical process

The hydrometallurgical process often induced with the thermal treatment to increase metal mining percentage from LIBs recycling process, [48–50]. The process was proven and capable of increasing efficiency. However, the environmental effect of this process with toxic and corrosive behavior may give a severe pollution to the environment. Lombardo et al. [23] investigate this effect in their study. The corrosion can reduce the amount of Cu and Al during the recycling process and hazardous gases for the atmosphere (CO) reported in this study and need further treatment.

4.3. M_NSO_4 solvent method

The traditional method to release residual electricity from LIBs that used NaCl gain challenge by environmental expert [51–53]. To prevent and to change the existing method, the experimental was done using different solutions to gain the most efficient with greener impact [41,54–57]. A cleaner method to discharge spent LIBs was reported by Xiao et al. [41]. The study revealed that using water electrolysis, the discharge is less efficient compared to solution that structurally can destruct batteries. Among the lists of solutions, the results show that using $MnSO_4$-based solution, the efficient result was achieved with cleaner results for the environment compared to previous method. The said solution can successfully use in recycling process of LIBs since it can avoid galvanic and organic leakage by using pH adjustment. the ion can be converted from the isolation layer and reduced contact with anode. The discharging process give no effect to the LIBs as shown in the following process.

$$Mn^{2+} + 2OH^- \rightarrow Mn(OH)_2 \quad (1)$$

$$4Mn(OH)_2 + O_2 \rightarrow 4MnOOH + 2H_2O \quad (2)$$

4.4. Solvent method mixed with Drying, Grinding, Screening

The performance of spent lithium iron phosphate (SLFP) and spent lithium manganate (SLMO) cathodes that used as adsorbents of heavy metal in water were evaluated [39,58–61]. Several effects were examined such as co-existing ions on adsorption, adsorption time, and initial adsorbate concentrations. Heavy metal such as Cu^{2+}, Pb^{2+}, Zn^{2+}, and Cd^{2+} can be adsorbed with SLFP (30-40 mg g^{-1}) and SLMO (25-30 mg g^{-1}). This indicates the method have highly efficient to recover the heavy metal consists in LIBs. Moreover, the cathode material can be recycled as adsorbents with the concept of waste to treat waste.

4.5. A three-dimensional microbial fuel cell

The recycle process of lithium-ion battery focused on the how to construct and recover Co from lithium ion battery were gained interest from researchers [62–65]. A three-dimensional microbial fuel cell (3D-MFC).with two-chamber system combined with granular activated carbon (GAC) microelectrodes was applied in the research that conducted by Huang et al. [62]. The study used stripping cobalt sulfate as a solution. The results show that more than 98% recovery of Co can be obtained with the power density was about 11.34 W/m^3. The study also concluded that adding ammonium carbonate into the system can increase the Co's precipitation. The system presented in this paper successfully achieved by using electromigration. The study further concluded that the chemical precipitation has more effectiveness on the adsorbing Co ions compared to pure electrostatic method.

4.6. Metallurgical-mechanical method

In term of capacity, Yun et al. estimated that in 2020, more than 250000 tons of batteries must be disposed and recycled [66]. Until now, the technology that can recycle all these dumped batteries in one year does not exist due to the complex recycling process, government regulation, and worldwide recycle standard. Although the system does not fulfill the world requirement, the technology developed by many researchers is still on progress with a good prospects [67–70]. Among many recycling techniques, the technique called the Metallurgical-mechanical recycling method is a hot topic. The technique is proven can be embed with more advanced technologies such as automatic and intelligent recovery system such as genetic programming [71], and artificial neural networks [72], advanced recovery process for slag, electrolyte, and anode-cathode [66].

4.7. Hydrometallurgical method

The components of battery such as copper, manganese, cobalt, nickel, and lithium can be recycled and separated by using the hydrometallurgical method [73–76]. The study obtained by Cheng et al. [76] show that In the final product, Nickel can be recovered as Ni $(OH)_2$, and Lithium as Li_3PO_4, after filtration and drying process. The study summarized that the following element can be recovered with superior percentages: 100% for copper; 99,2% for manganese; 97,8% from cobalt; 99,1% for nickel; and 95,8% for lithium. These metals were recovered using multiple chemical reactions, which consisted of specific metal recovering processes. The hydrometallurgical process is predicted in this paper to be a candidate for the effective separation and comprehensive recovery of all metals from the leaching liquor. This leaching liquor of spent LIBs consists of some heavy metals that are known to be detrimental to the environment and could be one of potential redundant sources of metal for the multitude of applications, including cathode material of new batteries. This could lead to more sustainable recovery of heavy metal and valuable metal. The study concluded that the hydrometallurgical processes could become one of the most promising candidates for recovering metal from complicated waste material stream. This is due to the capabilities of the process to recover high-value added metal as well as low-value added metal.

4.8. Chemically dissolve method

Recycling lithium ion batteries using chemicals combined with the dissolve method gained interest from many researchers [77–80]. The study conducted by Zhang et al. [77] developed a closed-loop process for recycling $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ from the cathode scraps for lithium-ion batteries. This study conducted using trichloroacetic acid (TCA) with hydrogen peroxide (H_2O_2) that had been actively refined to dissolve $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ from the cathode scraps. The study is conducted to optimize the recycling process of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ by obtaining lower Al dissolution rate without further sacrificing other metal. The study concluded that the optimization can be achieved through increasing the concentration, leaching temperature and time, and decreasing the S/L ratio. This is proved the feasibility of a closed-loop recycling of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ from the cathode scraps.

5. Technical Aspect Related to Recycling Battery

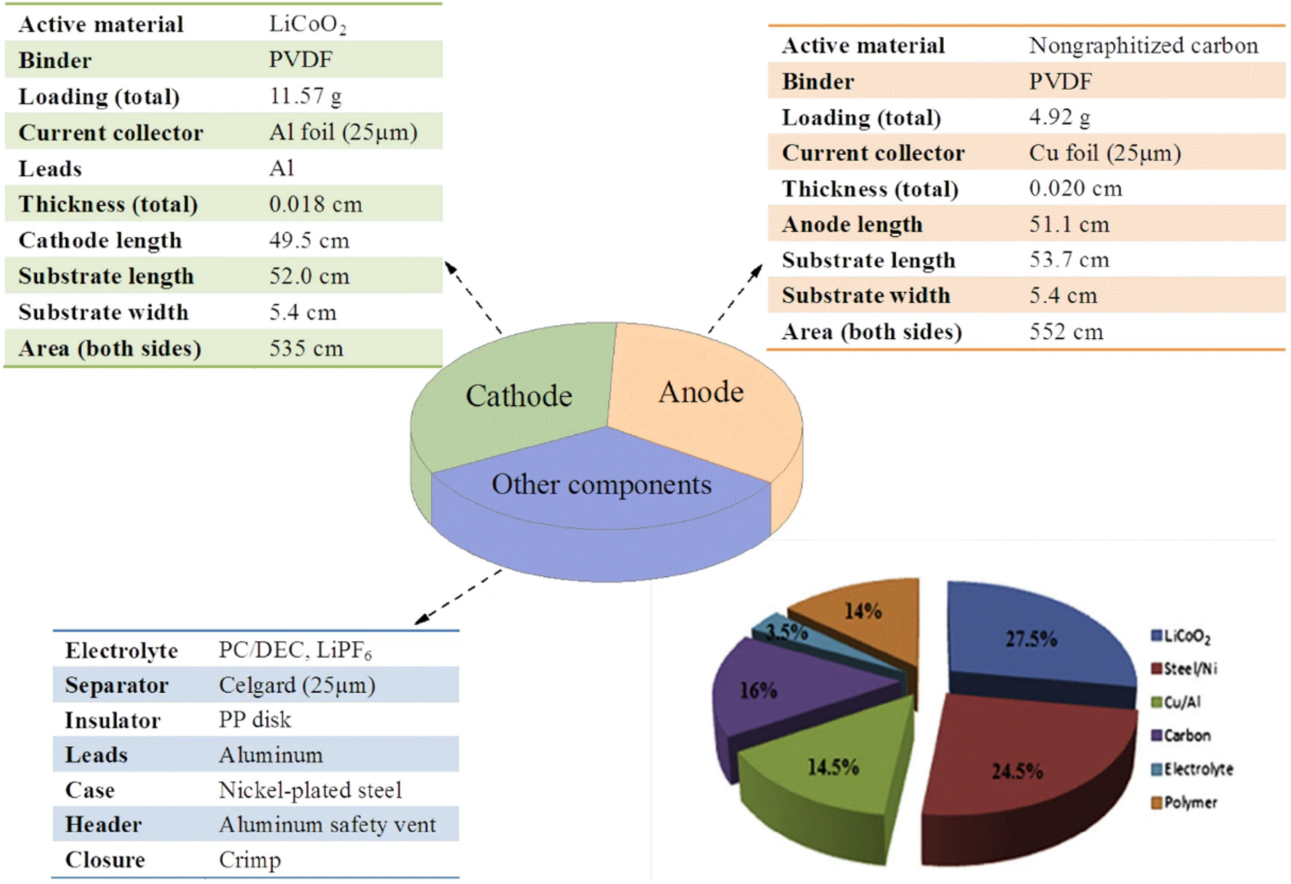
The recycling process of the LIBs gained huge interest from many researchers due to its elements related to the economic aspect [81,82]. The mining technology, known as urban mining, is closer to adequate LIBs. Urban mining has challenges that used secondary LIBs to process into different elements that can be sold. Since the LIBs have many different economically prospect elements, this recycling process gains attention from different people. From collecting the second LIBs, processor, to the buyer of the final elements that already pured by the recycling technology. Generally, before recycling process can be processes, we need to determine the element inside the LIBs. By determine the elements that composed the LIBs, the typical recycle technique can be applied to get the lowest cost and highest purity results. Fig. 6 showed the typical ement used in the LIBs that can be devided into 3 main portions: Cathode, Anode, and other components [83]. These composnents can be used to prioritize the receyling process, which is the element that can be gathered first. The important elements listed, such as $LiCoO_2$ and $LiPF_6$ can be the first priority, followed by the other elements such as Graphite (which could be a small part of it contains graphene), metal parts, other electrolyte elements, and then isolator element for separator [84,85].

Recycling processes of LIBs generally can be listed in the Fig. 7. The process can be categorized into three different activities: pretreatment, leaching, and resynthesis [8]. The pretreatment process start with the discharging activities. This step have purposes of normalizing the currect of the battery. By using solution process, or counter-discharging method, the target to normalize battery can be achieved. After that, dismantling is a common way to split the metal case with the anode and cathode. This process need relatively simple process using blade to break the metal case. The next process was to use NaOH solution to split Al solution with the following drying and calcination process. The next step was leaching process. This process have purposes to separate the Lithium, Nickel, Cobalt, and Mangan. The final step namely resynthesis was performed to gain the rest of the element also to produced NCM powder [83,86].

Other researcher provided different technical recycling process of LIBs that can be determined in the following steps

1. Crushing, ultrasonic washing, acid leaching, and chemical precipitation are some of the methods used [87].
2. Mechanical shredding, electrolyte extraction, electrode dissolving, and cobalt electrochemical reduction are all steps in the cobalt electrochemical reduction process [68].
3. Dismantling, chemical deposition, and solvent extraction are all steps in the process [50,88,89].

4. A mixture of mechanical, thermal, hydrometallurgical, and sol-gel procedures include crushing, acid leaching, heat treatment, and chemical precipitation [87,90].
5. Dismantling, acid leaching, solvent extraction, and chemical precipitation are all steps in the process [22,66,81].
6. Mechanical disassembly and separation, electrochemical and thermal treatment; a combination of mechanical dismantling and separation, acid leaching, chemical precipitation, and solvent extraction [22,34,66]
7. Leaching, solvent extraction, and electrowinning [91,92].
8. Dissolution, heat treatment, acid leaching, and chemical pre-capitalization are all examples of chemical pre-capitalization.



Active material	$LiCoO_2$
Binder	PVDF
Loading (total)	11.57 g
Current collector	Al foil (25μm)
Leads	Al
Thickness (total)	0.018 cm
Cathode length	49.5 cm
Substrate length	52.0 cm
Substrate width	5.4 cm
Area (both sides)	535 cm

Active material	Nongraphitized carbon
Binder	PVDF
Loading (total)	4.92 g
Current collector	Cu foil (25μm)
Thickness (total)	0.020 cm
Anode length	51.1 cm
Substrate length	53.7 cm
Substrate width	5.4 cm
Area (both sides)	552 cm

Electrolyte	PC/DEC, $LiPF_6$
Separator	Celgard (25μm)
Insulator	PP disk
Leads	Aluminum
Case	Nickel-plated steel
Header	Aluminum safety vent
Closure	Crimp

Fig. 6. Typical elements that used in the Lithium Ion Battery [83].

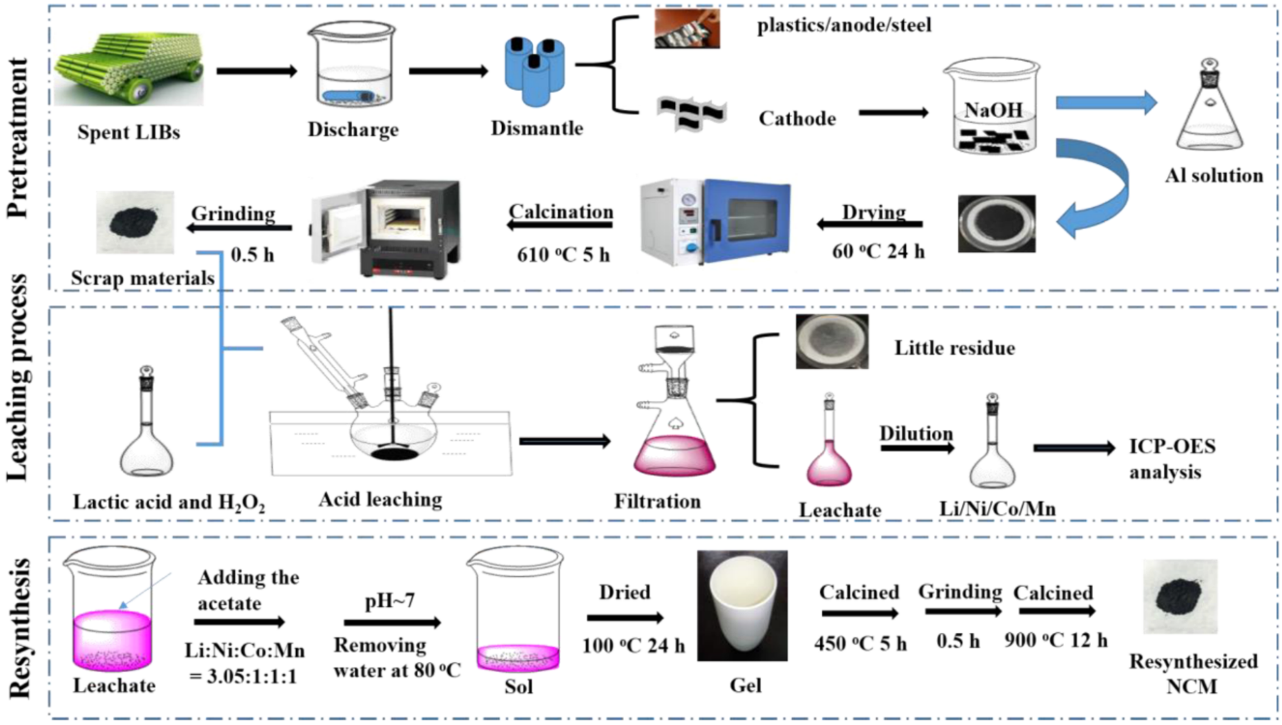


Fig. 7. Three steps of recycling battery in general [8].

6. Output products and influence on the environment and economic aspects

The recycling process of LIBs produces a multitude of materials. Each recycling method focused on recycling certain materials or metals from spent LIBs. Therefore, each method would eventually imply different environmental burdens [35,42]. The wet and dry crushing method produced recycled material composed of two groups, fine particles like electrode materials and the coarse particle-like copper foil, plastics, and diaphragm fiber [47,93,94]. The thermal treatment with the addition of hydrometallurgical process produced higher concentration of LIBs constituents. However, the thermal with hydrometallurgical process has environmental solid impact [23]. The CO level in the pollutants causes this strong effect. In addition, surging in the CO level could substantially reduce the output of recycled LIBs [23].

The M_NSO_4 solvent methods used the chemical agent of the M_NSO_4 to efficiently destruct the used LIBs [41,95–97]. This method promised better and cleaner impart to the environment with respect to the previous methods. This process can be further controlled by adjusting the pH settings of the cell [41]. The next method, the Solvent method mixed with Drying, Grinding, Screening, can absorb heavy metal elements in the LIBs [39]. This method actually the better methods for the environment.

Next, A three-dimensional microbial fuel cell is usually used to recover cobalt element in the LIBs [62,98–100]. This decomposition system uses a three-dimensional microbe with a two-chamber system combined with granular activated carbon (GAC) microelectrodes. This method proves to be the most suitable apple and the Google Phone. This method can ultimately recover by as much 98% of the Cobalt content in the LIBs [62]. This is considered as good news since the cobalt usually considered heavy metal. The metallurgical and Hydrometallurgical method is still the hottest topic on recycling the used LIBs [75,76,101,102]. The Metallugrical Mechanical dan hydrometallurgical is usually used to recover a lot of cheap metal. This method also provide little impact on the environment [76]. The last method presented here is the chemically dissolve method used on the LiBs. This method is done using the thrichloaceticacid (TCA) and the hydrogen peroxide. The chemically dissolve method was environmental. The environmental impact of this method is miniscule. The used of free corrosive acid and strong oxidant process during recyclye of LIBs have been proposed by Wang et al. [103]. The method began with co-grinding with different additives in a hermetic ball milling system, after which A water leaching approach easily recovered co and Li.The method began with co-grinding with different additives in a hermetic ball milling system, after which A water leaching approach easily recovered co and Li. It was discovered that EDTA was the best co-grinding reagent.It was discovered that EDTA was the best co-grinding reagent.It was discovered that EDTA was the best co-grinding reagent. In general, the method consists of three steps: (1) co-grinding LiCoO2 powder with EDTA, (2) leaching the ball milled combination with water, and (3) chemical precipitation to extract Co and Li in the form of Co_3O_4 and Li_2CO_3. The sulfur dioxide and lead fume emissions from classic pyrometallurgical melting furnaces have the most direct onsite environmental impact; this is especially obvious for the reverberatory process, which emits significantly more sulfur dioxide. The government's environmental protection division has conducted a series of environmental inspections for direct pollution emissions. Because of the high recycling rates from leaching at low temperatures, the onsite pollutant emissions of the new processes are obviously reduced [104].

Moreover, in the environmental aspect, the recycling process of LIBs can have positive and negative impacts, which are closely related to the economic aspect [81,82,100,105]. The characteristic of recycling LIBs from lab-scale is shown in Fig. 8 based on It is shown that the highest cost comes from leaching cost. Omrani and Jannesari [105] conducted the other study related to the use of LIBs. The study used different scenarios to determine the effect of using secondary LIBs (reused) to be implemented in different applications. The original battery used in the electric vehicle is then reused in energy storage stations, residential, industrial, and photovoltaic power plants. A deeper analysis of the social-economic-environmental impact of the scenario using different mechanisms has been studied previously [82]. The reward and penalty that the government implements then being evaluated. The scenario was clustered into 3 models: no policy intervention, subsidy, and reward-penalty. After the study, there are 3 conclusions gained.

1. The reward-penalty method has a more significant impact on recycling rates and social welfare than the subsidy approach.
2. The consumer surplus and the profit of the EV producer are the two key driving variables of social welfare under the subsidies scheme. Reduced environmental impact is another major contributor to the reward-penalty approach.
3. A low minimum recycling rate benefits the environment, consumer excess, and EV manufacturers' profits. A relatively high minimum recycling rate, on the other hand, is advantageous in terms of lowering both policy implementation costs and the environmental burden produced by mistreated EV batteries.

The economic revenue from the elements composed of LIBs also became an intensive study by researchers [104,106]. The elements are clustered into two parts: input and output, based on the average price, as shown in Fig. 9 and Fig. 10. It is shown that the input and output from the recycling process of the spent lead-acid battery can be illustrated in detail. The combination of material input consist of lead paste and the chemical reagents (Sodium carbonate, Coal, Sodium carbonate, Iron scraps, Calcium oxide (CaO), Sodium citrate, Sodium hydroxide (NaOH), Hydrogen gas (H2), and Water). This input element is used in the recycling process to gain the output product, as shown in Fig. 10.

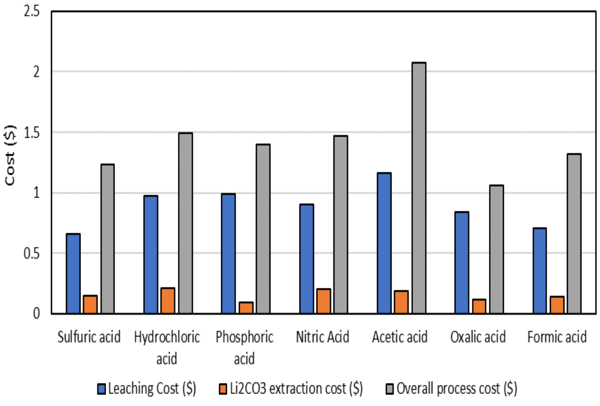


Fig. 8. Cost of LIBs recycling in lab scale. Overall cost included other processes such as evaporation and other components extraction costs.

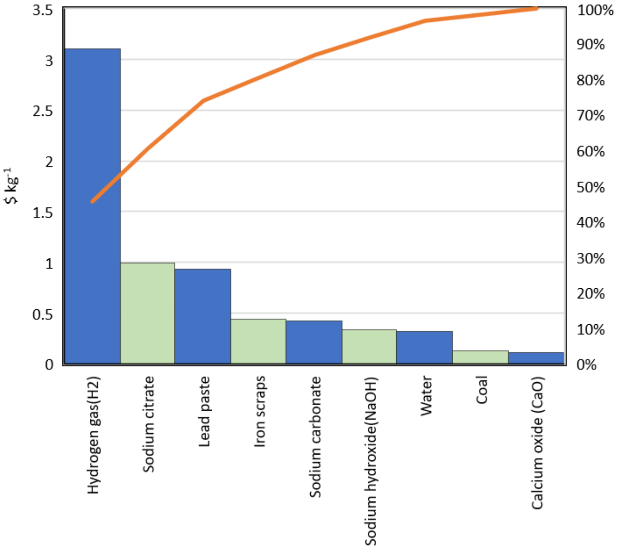


Fig. 9. *Cost of Unit price from the input product [104].*

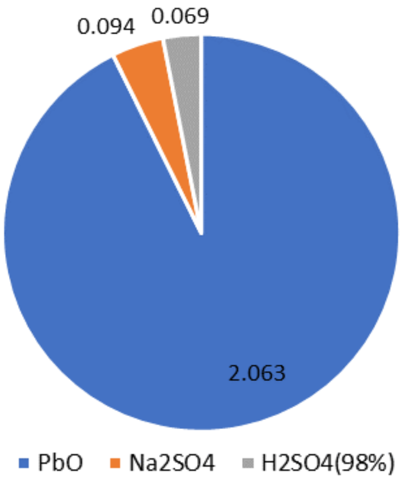


Fig. 10. Cost of Unit price from the output product [104]. The value unit in \$ kg^{-1}.

7. Conclusions

The sharp increase of the consumption and demand for LIBs will result on numerous spent LIBs in the future project [83]. Choosing the appropriate recycling method and optimization of the existing recycling process is as important as the recycling processes [107–110]. The LIBs contain a multitude of valuable metal that exists in very small amounts. This makes the efficiency and the optimization of the recycling process a must. Ideally, this optimization will eventually lead to low energy recovery and environment-friendly processes. To date, the LIBs recycling process has been developed in a rapid pace [111,112]. The process that decades ago deemed impossible is done on numerous industries now. However, the simplification of the process is still being developed. Although LIBs had been developed to the point where it is economically viable, the technology still accompanies lots of complicated and special processes that increase the cost of recycling. Therefore, much effort and research are still needed to develop more powerful recycling technology.

Moreover, there are challenges which hampers the development of the LIBs recycling processes. The first and foremost is the ever-evolving LIBs technology. Although this development is good for the costumer and the market, this development is leaving the recycling technology development behind. The second most important thing would be the absence of regulation, standard, and laws regarding to the LIBs' recycling process. This standard will be needed to ensure safe handling, disposing, and recycling of the LIBs. Eventually, in order to further speed up the development of LIBS recycling process, further effort and research may concentrate on, but not limited to the following (1) Identifying and sorting process of the LIBs. (2) Designing the LIBs with recycling in mind. (3) Accelerating the advancement of the LIBs recycling process by supporting and incentivizing the sector.

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