

Impact of Spent Lithium-Ion Batteries Recycling on Economy and Environment

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Abstract

Lithium-Ion batteries (LIBs) are used in several products such as portable electronic devices like toys, wireless headphones, handheld power tools, mobiles, small and large appliances/gadgets, etc., and electrical energy storage systems. Also, more importantly it is projected to be used as an energy source for the next generation of electric vehicles with the aim of environmental friendliness. Therefore, it is predicted that the consumption of active materials will continuously increase, which results in increasing resources demand such as lithium, cobalt, nickel, manganese, copper, aluminum, etc. Furthermore, improper disposal of spent LIBs is affecting the environment and living things because it contains flammable electrolytes, heavy and toxic metals. Because of this, recycling of LIBs will be necessary to avoid shortage of rare resources and minimize the pollution due to hazardous components. If it is not properly managed at the end of their useful life, they can cause harm to human health or the environment. To overcome these issues, development of LIB recycling system is one of the attractive technologies in the field of research and development. Recycling assists to protect our environment, reducing the manufacturing cost and creates new job opportunities in the field of LIB waste recycling industry. Spent LIBs often contain among other useful metals high-grade copper, aluminium in addition to cobalt and nickel as well as rare earths. Recycling processes for lithium batteries are required to prevent a future shortage of cobalt, nickel, and lithium and to enable a sustainable life cycle of these technologies. From these processes, lithium, cobalt, manganese, nickel, copper and aluminium from spent battery cells are recovered. Depending on the separation technology, recycling processes recover approximately 25% to 96% of the materials of a lithium-ion battery cell. These technologies consist several steps, which are combined into complex process chains, especially considering the task to



recover high rates of valuable materials about involved safety issues. The purpose of the present chapter is to summaries structure and components of LIBs, their current state and some of the most promising technologies for recycling process of spent LIBs.

Keywords: LIBs, Heavy & Toxic Metals, Disposal, Recycling Processes, Challenges

Introduction

A battery is an electrochemical device that generates electrical energy by converting chemical energy contained in its active material via redox reaction. Its basic electrochemical unit is known as cell and battery assembled by using one or more cells. A cell consists of an electrode, electrolyte, separator and container. After the invention of battery technology, several primary and secondary batteries are discovered. Based on its performance and reliability, only few of them are used for commercial purpose such as Ni-MH, Pb-acid, Ni-Cd, lithium-ion battery (LIB), etc. Since 1990, lithium-ion batteries (LIBs) are widely used for various applications and dominating the power source market for various portable electronic instruments. Compared to other types, LIBs have longer life cycle, environment friendly, compact design, low self discharge rate, broad operating temperature range, higher cell voltage, less memory effect, etc., therefore LIBs become popular (Du *et al.*, 2017). Recently, in electronic vehicles (EVs) LIBs are used as an energy source due to their high energy density, lightweight and long-life cycle. These technical advantages make LIBs more attractive for various urban or industrial applications and next generation of electronic vehicles. Therefore, LIBs have one of the promising ways for the reduce of CO₂ emission. The aforementioned technical advantages and environmental importance reflects increase in production rate of LIB.

The combination of active material used as electrode and electrolyte for LIB governs the charge storage performance of LIB (Rajoba *et al.*, 2019). The materials like lithium, cobalt, nickel, aluminum, copper, palladium, titanium and manganese are used in electrode. Moreover LiPF₆, LiOSO₂CF₃ and LiClO₄ are used as an electrolyte with ethylene carbonate (EC) and dimethyl carbonate (DMC) (Bazito *et al.*, 2006; Liu *et al.*, 2016; Park *et al.*, 2010; Hofmann *et al.*, 2013). Some of these materials are toxic and having rare occurrence. However, the ever-growing energy demand for consumer electronics and EVs increases the utilisation of batteries and consequently increases consumption of various metals. The huge gap between consumption of these metals and their availability leads to continuous increase in price of LIB. On the other hand, today increasing utilisation of LIB generates multimillion metric ton heap of used LIB that could end in the trash (Zeng *et al.*, 2014). In addition, improper disposal of spent LIB leads to environmental issue and affects human health because they contain high percentage of heavy and toxic metals (Liu *et al.*, 2019). Thus, recycling of rare and toxic metals from spent LIB shows valuable way to prevent environmental pollution and raw material consumption. Unfortunately, LIBs are new like other E-waste spent, so all recycling technologies



are limited to laboratory level. Therefore, due to technical, economical and some other challenges, like collection, sorting and processing, only 5% LIBs are recycled today (Martínez *et al.*, 2019). Recycling lowers the manufacturing cost of new product, protects our environment and creates new jobs in the field of recycling industries. Another reason to recycle LIB is to keep heavy and toxic materials away from landfills because they can contaminate the soil and water (Wang *et al.*, 2015).

The gap between recycling and production currently represents untouched source of rare material (Martínez *et al.*, 2019). Therefore, this chapter presents discussion on the current technology for recycling of LIB. To deeply introduce current status, challenges and future outlooks for the recycling of the spent LIB's, we first introduce the structure in this chapter then configuration of LIB and analyse available technology for the recycling of LIBs. Further, we focused on the main challenges in the current technology.

Structure of Lithium-Ion Battery

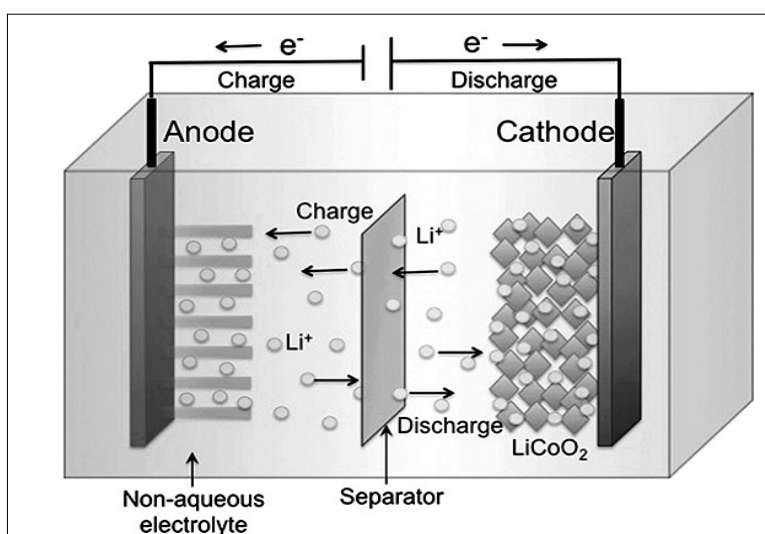


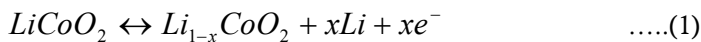
Figure 1 Schematic Representation of Charging and Discharging of Lithium-Ion Battery

As we can see in Figure 1 LIB consists of cathode, anode, electrolyte and separator. Generally, active material such as lithium-based materials, carbon-based materials, tin based materials, silicon-based materials, and transition metal oxides are used as an anode and it is coated on copper foil (Rajoba *et al.*, 2021a; Park *et al.*, 2010; Liu *et al.*, 2016; Li *et al.*, 2018; Rajoba *et al.*, 2021b). Additionally, active materials such as layered compounds (LiMO_2), spinel compounds (LiM_2O_4), olivine compounds (LiMPO_4), silicate compounds (Li_2MSiO_4), tavorite compounds (LiMPO_4M) and borate compounds (LiMBO_3) are used as a cathode material and is coated on aluminum foil (Xu *et al.*, 2012; Chen *et al.*, 2013; Tao *et al.*, 2011; Ni *et al.*, 2015; Xiao



et al., 2013; Barpanda *et al.*, 2015). Here M is used for metal ion. During the preparation of electrode material conducting carbon and polyvinylidene fluoride (PVDF) is used with active material. Both electrodes are separated by porous polymer membrane. Lithium hexafluoride (LiPF_6) dissolved in organic solvent ethylene carbonate (EC) and di-methyl carbonate (DMC) is used as an electrolyte. During charging, lithium ions are de-inserted from the positive electrode and inserted into the negative electrode through electrolyte. At the same time, electrons are freed from positive electrode and move towards negative electrode through external circuit. In case of discharging, the process is reversed (Linden *et al.*, 2002). The detailed chemical reactions on the electrodes are

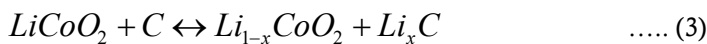
On positive electrode:



On negative electrode:



Over all reaction:



Multiple Ways for Final Disposal of Batteries

There are different alternatives to the final disposal of batteries (Bernardes *et al.*, 2004):

Landfill

Now a day's most of the household batteries especially the primary batteries are sent to a sanitary landfill.

Stabilisation

In this process, technology is used to avoid contact of metals with the environment in a landfill. However, due to high cost this alternative is rarely used.

Incineration

Generally, this alternative is used when spent batteries are sent to a municipal waste combustion facility. During this process hazardous materials such as mercury, cadmium, lead and dioxins release and merge in to environment.

Recycling

By using special technique, spent LIBs can be used to recycle metals present in it. From last decade researchers are concentrated to improve technology of LIB recycling. Now things are starting to change.



Phases of Recycling Process

Battery recycling involves processing of spent batteries and its schematic representation is shown in Figure 2. The prime aim of processing is to reuse spent batteries rather than disposing. Recycling is very important because it avoids improper disposal. It is also important to know recycling process of spent LIBs (Huang *et al.*, 2018).

Collection

In this step, recyclers collect the spent LIBs from collection points or other locations.

Sorting

In this step, recyclers split spent batteries into two parts of which, one is plastic materials and another is metal components. Both materials are useful during the manufacturing of new products.

Smelting

In this step, metals are recycled from the separated metal component. This process is also known as metal reclamation. Various metals such as Nickel, chromium, manganese, iron *etc* are recycled which is useful for making new products. There are some safety risks associated to collection, transport and storing of spent LIB.

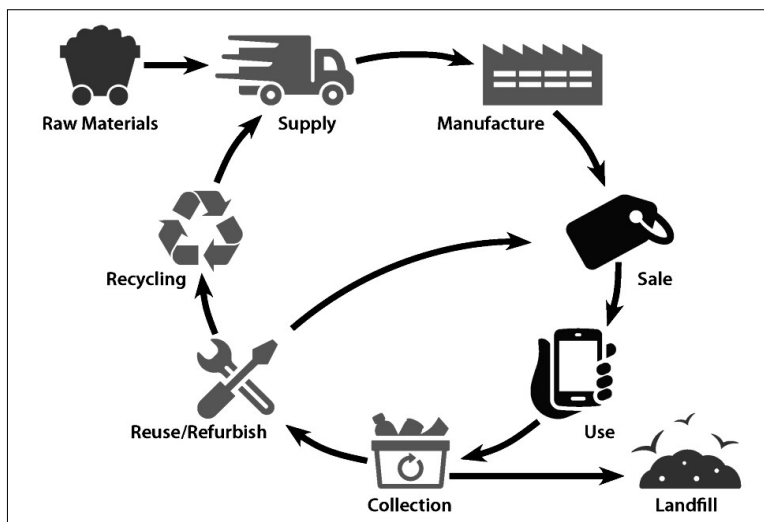


Figure 2 Schematic Representation of Recycling System of Lithium-Ion Batteries

(source: <https://www.epa.gov/recycle/used-lithium-ion-batteries>)

Economical and Environmental Motivation for Recycling Lithium-Ion Batteries

The following points are in support of the recycling LIBs



1. Spent LIB contains various rare, toxic and flammable active materials and electrolyte. The presence of these materials in spent LIB has considerable risk because they can catch fire easily. Recycling alleviates toxicity and flammability risks.
2. Mostly LIB ends in a trash; consequently, the presence of these materials creates solid waste. When this solid waste comes in contact with liquid or water, some compounds are dissolving in liquid or water. Later dissolution of these compounds turns into the composition of the leachate. There are numbers of factors which governs rate at which compounds are getting dissolved in the material such as pH, nature of casing, charge left in battery, oxygen content in landfill. Besides, these materials can contaminate the soil and groundwater, which is hazardous to our ecosystem.
3. Cost of required raw materials will be reduced by recycling of LIB.
4. Most of the countries in the world are depending on specific countries for LIB raw materials. Recycling offers the opportunity for bypassing the imports of raw and refined LIB materials.
5. To reduce cost of transportation, there are strong incentives for localising at least part of the recycling infrastructure. The developed recycling industry will generate billion rupees in tax income and jobs.

Safety and environmental issues, economic development and independency on raw material motivate recycling of LIB.

Challenges in Recycling of Lithium-Ion Batteries

1. Ideally, recycling means restore spent battery material to their original condition, but in reality, it is not possible. Recycled materials are less pure compared to the virgin material. Therefore, if it is used in LIB, it may reduce cycle life of LIBs.
2. Transportation and collection of spent LIBs must be considered in recycling process to make environment friendly recycling system. Role of transportation is minimal as compared to overall recycling costs however proper transportation system is required to minimize environmental impact in this process.
3. Generally, LIB is formed by using multiple electrochemical cells, which are connected in series or parallel and enclosed in metallic or plastic case. One of the foremost challenges in recycling of spent LIBs is metal extraction from active materials which is used for making electrode. Because, it is mixture of conducting carbon and binder; in addition, it is deposited on copper and aluminum foil. To simplify this process, researchers choose manually dismantling to individual gather the electrode material. On the other hand, some other techniques are proposed to simplify this process such



as heat treatment (In this process active materials and binders are pyrolysed at high temperature to separate these materials), solvent soaking (in this method different solvents such as NaOH, Trifluoroacetic acid, N-methylpyrrolidone are used to dissolve organic binders and separate out active material from substrate) and mechanical treatment (in this process mechanical force like crushing and magnetic separation, is applied to separate out active material (Fan *et al.*, 2020; Xiao *et al.*, 2020).

4. Separating metal elements from active materials is also challenging during recycling of spent LIBs. Generally metal elements were separated by using acid/alkali leaching, bioleaching and chlorination process. The main aim of recycling is to reduce environmental issues, which is created by spent LIBs. However, recycling process consume significant amount of chemical, electrical and thermal energy. Furthermore, toxic gases, solid waste and contaminate water are released as a byproduct.
5. Separating everything from spent LIB is not always the best way for LIB recycling but utilising as much of their existing structure as possible is efficient way for LIB recycling. However, the technology for properly disassembling LIB is not yet developed. It is challenging to develop technology (Gaines *et al.*, 2018).
6. Market price of recycled material is combined cost of collection, transportation, storing and processing. Recycled material gains market when its price is comparatively lower than the cost of raw material. However, the cost of material recycling is higher as compared to the cost of extracting and refining new resources. To reduce the cost of recycling is also challenging task (Beaudet *et al.*, 2020).

Recycling Processes for Lithium-Ion Batteries

Various processes are developed for recycling today's LIB. Generally, these processes are classified into two different categories, one is physical process and another is chemical process. For both the processes, first stage is for casing and connection materials which includes skinning, removing of crust, crushing, sieving and separation of materials in order to separate the cathode materials from other materials is removed in advance. The stepwise pretreatment process is (Xu *et al.*, 2008),

1. By using small knife or screwdriver, plastic cases of the batteries are removed.
2. Before removing cover of the battery *i.e.*, metallic shells, it was immersed into liquid nitrogen (for safety precautions) and fixed in a lathe machine.



3. The ends of the metallic shell are removed firstly and then internal material of the battery is smoothly removed by longitudinal cut. Then anode and cathode are separated manually.

For safety precaution, above mentioned all steps are carried out using glasses, gloves and gas masks (Mossali *et al.*, 2020; Shin *et al.*, 2005). The prime aim of pretreatment process is the improvement of metallic portion, reducing scrap volume and energy consumption, improvement of recovery rate, manage safety issues and take advantage of different chemical and physical properties of LIBs components.

Various recycling processes for LIBs are described one by one below.

Mechanical Separation Process

Shin *et al.*, reported mechanical separation process for the recovering lithium and cobalt metal from spent LIBs for possible application to a commercial scale plant (Shin *et al.*, 2005). Separated electrode material goes through series of mechanical processes which include crushing, sieving, magnetic separation; fine crushing and classification were carried out to yield enriched particles of lithium cobalt oxide in sequence. In order to remove small parts of substrate from active material and for satisfactory separating metal particles from waste, crushing and sieving was taken two times. It improves the recovery efficiency of target metals. Purification process of the leachate is applied, because PVDF binder and carbon does not dissolve in acid. But the lithium dissolves in the acid solution during leaching. In the crushing process, lithium hexafluorophosphate (LiPF_6) decomposes into lithium fluoride and phosphor pentafluoride. Propylene carbonate (PC) and diethyl carbonate (DEC) were evaporated in this process. Several metals which are penetrating into each other are difficult to separate by using this process.

Thermal Treatment

In most recent decade the majority of commercial LIBs are designed by using lithium and cobalt containing electrode material (Lee *et al.*, 2002). Thermal treatment is one of the attractive methods to recycle these metals from spent LIB. In this method, first, spent LIBs are thermally treated at 100-150 °C. Then the spent LIBs are disassembled with a high-speed shredder. In next step, two-step thermal treatment is performed to separate out active material from substrate. Then, obtained active material is heat treated at a temperature range 500-900 °C. In next step, obtained active material is reacted with nitric acid solution and then gel is formed. The obtained gel is calcined in the temperature range 500-1000 °C and desired powder is formed.

This method has simple and convenient operations, however by using this technique recovering of organic compounds is not possible due to combustion of carbon and organic compounds.



Mechanochemical Process

Saeki *et al.* reported mechanochemical method for recovering Co and Li from spent LIB (Saeki *et al.*, 2004). In this process active material is grinded with polyvinyl chloride (PVC) to form Li and Co-chlorides. The obtained chlorides are soluble in water, therefore the resultant material mixed with water to extract Co and Li.

Dissolution Process

Contestabile *et al.* reported dissolution process. In this process, spent LIBs are recycled without separating cathode and anode electrodes (Contestabile *et al.*, 1999). The collected battery rolls are reacted with N-methylpyrrolidone (NMP) at 100 °C. During this treatment, active material is effectively separated from substrate. It also recovers substrate such as aluminum and copper. This is simple process due to metals are separated without separating electrode from spent LIB. However, for dissolving active material N-methylpyrrolidone (NMP) is used which is too expensive. Therefore, cost of recycling process increases.

Chemical Processes

In chemical industries normally spent LIBs are recovered by chemical process, this process is connected with the two processes, one is leaching steps in acid or base medium and another is purification processes in order to dissolve the metallic fraction and to recover metal solutions. In order to recover metals, in purification process various steps are involved which are chemical precipitation, filtration, extraction, etc. In this process precipitate is formed by optimising pH of solution and by adding some reaction agent. In addition, metals are also separated by solvent extraction. During this process organic solvents are used. Organic solvent binds metallic ion and separates metals from solution. Then separated metals can be recovered by electrolysis process. In this section chemical process is describe in detail

Acid Leaching

In this process, generally inorganic acids such as H_2SO_4 , HCl , HNO_3 and $\text{NH}_4\text{OH.HCl}$ etc are used as leaching agents (Zhang *et al.*, 1998). The experimental result reveals that the leaching efficiency of Co is highest in hydrochloric acid among above mentioned three leaching agents. In addition, leaching efficiency of Co enhances with increasing temperature. However, during this reaction, chlorine (Cl_2) gas is released which leads to serious environmental problems. In order to solve this issue, special equipments are required to treat Cl_2 but this technology is not available. To overcome this problem H_2SO_4 and HNO_3 are used with hydrogen peroxide (H_2O_2), Here H_2O_2 is used as a reducing agent. This reaction is reported by Mantuano *et al.*, and Lee *et al.*, and its results reveals that due to H_2O_2 leaching efficiency is increased by 45 % and 10 % for Co and Li respectively (Mantuano *et al.*,



2006; Lee *et al.*, 2002). In addition, its efficiency is also governed by temperature and concentration of HNO_3 and H_2O_2 .

Bioleaching

Bioleaching is also attractive method for recycling spent LIB due to its cost effectiveness and higher efficiency. In this technique, chemolithotrophic and acidophilic bacteria, with acidithiobacillus ferrooxidans are used. Its energy source is elemental sulfur and ferrous ion and it produce metabolites such as sulfuric acids and ferric ion in the leaching medium. Till today this technology is in research stage and it is not used for commercial purpose.

Solvent Extraction

In this technique Co, Cu and Li are recycled from spent LIB, during reaction here di-phosphoric acid, trioctylamine, diethylhexyl phosphoric acid, 2-ethylhexyl phosphonic acid, mono-2-ethylhexyl ester are generally used as an extractants. Very high purity of recovered material is achieved by this technique. In addition, this technique is having easy operational conditions, low energy consumption and good separation effect. However, expensive solvents are used during reaction which results in increasing recycling cost. By choosing proper solvent we can reduce recycling cost (Xu *et al.*, 2008; Lin *et al.*, 2003; Tanii *et al.*, 2003).

Chemical Precipitation

In this technique, various precipitate agents are used to form precipitate of metal. Generally, ammonia, NaOH *etc* is used as a precipitate agent and it is inexpensive. By using pH meter and controlling pH of solution, one can easily control the precipitate. This technique is useful for industrial application because as compared to other technique this technique has simple reaction and by choosing proper precipitate agent, highly pure products are recovered using this technique.

Some of above-mentioned chemical processes are attractive for metal ions recovery from spent LIB (Huang *et al.*, 2018). Though, large amount of acid sludge is produced as a byproduct during chemical processes. The obtained acid can be easily recycled or neutralised, but the remaining heavy metal in a chemical waste is harmful for environment. Therefore, commercialisation of this process is possible by enhancing the extraction of metal with minimum steps.

Current Status of Recycling LIBs in India

Most of the raw materials like lithium, cobalt, nickel, etc., are found in few countries Chile, Congo, China, etc. It is well known that India doesn't have much of minerals to manufacture LIBs. Owing to these resources, India has to import LIBs in huge quantity and does not manufacture LIBs. Because of shortage of raw materials, the production of LIBs in India is not cost effective. Developed countries such as Korea



and Japan import spent LIB from India for recycling purposes. In the Indian market recycling LIBs industries will grow after awareness of reusability and it is possible once government brings a well-defined policy framework. In October 2019 Indian government announced one policy that offer tax concession to recycling industries. Like western countries, the Indian government offers several schemes such as quick approval to the manufacturing of hybrid and electric vehicles. Nowadays in India, electric vehicles (EV) are becoming as a part of the new transportation sector, initially, EVs are powered by lead acid battery but now it is replaced by LIBs. Based on the utilisation of LIB's, environmental awareness in people and government policies, it is predicted that in the current decade recycling market in India will certainly grow. Several global companies including Umicore in Europe, Glencore in Canada started the LIB recycling plant but India is lagging in the race. India has greater recycling capacity if the government pushed to a recycling business. Currently, Various Indian companies also have started to set up recycling industries for example, Gravita India Ltd., India's largest battery manufacturing company is entering the electric vehicle battery recycling business. Before five years Attero Recycling successfully commercialised a lithium-ion battery recycling plant in India. In August 2019, Tata Chemicals Ltd started a recycling plant in Maharashtra. They recover almost half the valuable metals, including aluminum, cobalt, copper, lithium, manganese and nickel. In the first stage, the project is started at the pilot scale and now they plan to develop a plant that recycles 500 tons of spent lithium-ion batteries. Mahindra Electric also announced to develop EV battery recycling plant. Raasi Solar has announced a new plant on lithium battery recycling along with battery assembling and cell manufacturing. Apart from these famous industries, some other industries are also working on spent LIBs in India few of them are summarised here.

Table 1 Some LIBs recycling industries in India

S.N.	Company name	Location	Technology
1	TES-AMM	Chennai	Mechanical
2	Exigo	Haryana	Mechanical
3	Sungeel Hi-metal	Andhra Pradesh	Hydro Metallurgy
4	ECOTantra	Pune	Mechanical
5	EXIMO Recycling	Gujarat	Mechanical

Conclusion

An energy storage device having greater energy and power density like a lithium-ion battery (LIB) becomes one of the essential devices in the current technological world. It is used in every portable electronic device as well as in electronic vehicles. LIBs are manufactured by combining various heavy, rare, flammable and toxic elements such as lithium, cobalt, nickel, aluminum, copper, palladium, titanium, manganese, etc. When its useful life is over spent LIB generates multimillion metric tons of E-waste. Its improper disposal leads to various environmental issues which affect human



health. There are different alternatives to the final disposal of batteries among the various alternatives recycling provides a better approach because the prime aim of recycling is to reuse spent batteries rather than disposing of them. In addition, this leads to a decrease in the manufacturing cost of the new product, protects our environment and creates new jobs. Government motivates to recycle the LIB due to its safety and environmental issues, economic development and independence on the raw material motivate the recycling of LIB. However, separating metal elements from the combined LIB is also challenging task during the recycling of spent LIBs because the technology for properly disassembling LIBs is not yet developed. Recently, the government announces well defined policies which offer tax concession, quick approval to the manufacturing of Hybrid and Electric Vehicles. These policies trigger various startup companies to set recycling industries.

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