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Metallurgical and mechanical methods for recycling of lithium-ion battery pack for electric vehicles

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ABSTRACT

Due to enormous growth of production of electric vehicles, it is estimated by the year 2020 about 250,000 tons of battery must be disposed or recycled. The technology to recycle this much amount of batteries in a single year does not exist., neither does the methods for recycling are standardized because of different configurations of battery packs. A challenge strictly poses on how to deal with lithium ion batteries, which are embedded in hundreds or more in a battery pack. Furthermore, the recovery of materials from the battery in the pack is essential to ensure the growth and sustainability of the electric vehicle market. It is desirable to establish a framework that is semi-automated/automated for ensuring faster disassembly of battery pack, identification and detection of residual energy of batteries in packs and recovery of materials from batteries. This review paper summarizes the two main basic aspects of recycling battery packs: mechanical procedure and chemical recycling (metallurgical). The work summarizes the existing recycling technology in these two aspects and identifies important research problems in the process of recycling of pack such as (i) automatic and intelligent recovery system, (ii) efficiency and safety disassemble of battery pack (iii) Adjustment of Chaos in recycling market (iv) Recovery processes for slag, electrolyte and anode, (v) Application in industrial scale, and (vi) development of recycling methods for new batteries having components with different properties. This paper also proposes a framework to push the recycling process from conception to practicality, both on government incentive polices and effective recycling technology.

1. Introduction

Within the last two decades, lithium-ion batteries (LIBs) technology has been extensively applied in wide-scale electric storage instruments, such as portable electronics, renewable power systems, and electric vehicles (EVs) because of their outstanding characteristics of small size, high voltage and energy density, long cycle life, and low self-discharge (Nitta et al., 2015). For instance, the global consumption of LIBs in 2000 rose to 500 million Li-ion cells, with the growth and expansion of 800% over course of 10 years starting from 2000. Furthermore, the increase of the EVs industry has been undergone a great change in the usage of LIBs. It is predicted that the large amount of spent LIBs in 2020 will surpass 500 thousand tons and 25 billion units (Zeng et al., 2014). With increasing concerns regarding environmental issues (Gaines, 2012) and the limited availability of lithium and cobalt, there have been stringent laws put in place world-wide in order to treat spent LIBs to properly recycle valuable metals as well as toxic chemicals (Meng et al., 2017). Therefore, the development of efficient recycling technologies for spent LIBs has become much attracted in the present time and future.

In recent years, many studies have focused on single recycling methods based on mechanical and metallurgy processes (Meng et al., 2017; Golmohammadzadeh et al., 2017). Mechanical processes comprise of disassemble of battery pack to modules, module to cells as well as the process of crushing single lithium-ion battery and sorting of materials. Metallurgical processes include pyro-, hydro-, bio-metallurgy and hybrid methods.

However, there has been a limited literature that combines both of them. Most of the literature review has been on materials recovery, life cycle assessment,policy development and cost-environmental benefit analysis of recycling of batteries (Wang and Wu, 2017; Awasthi and Li, 2017; Kumar et al., 2017; Liang et al., 2017). Thus, a systematic review

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summarizing the combination of recycling processes (mechanical and metallurgical processes) could pave the way for proposition of framework for efficient and effective recycling of battery pack. In this review paper, we combine the Metallurgical and Mechanical methods used in recycling of battery pack for electric vehicles (EVs). Fundamental recycling of spent Lithium-ion batteries was also summarized in context of metallurgical processes.

The remaining structure of the paper is as follows. Section 2 illustrates the components of lithium-ion batteries for EVs. Details in the recycling processes includes mechanical and metallurgical methods are introduced in Section 3. The critical gaps with new directions of research in the recycling process are discussed in Section 4. Based on research gaps, a framework for recycling of battery module is illustrated in Section 5. Finally, conclusions of the study are discussed in Section 6.

2. Components of lithium batteries for EVs

Electric vehicle (EV) packs typically comprises of battery modules and the battery management system. Battery modules comprises of batteries connected in series and parallel.

The LIB cell is comprised of four fundamental components: anode, cathode, electrolyte separate and shell casing (Xiao et al., 2017). The chemical compositions slightly vary as per manufacturers. The cathode materials in commercial LIBs are usually either lithiated metal oxide or lithiated metal phosphate such as: $LiCoO_2$ (LCO), $LiMn_2O_4$ (LMO), $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ (NMC), and $LiFePO_4$ (LFP). Table 1 shows the summary of upsides and downsides of the electrode materials used in commercial LIBs (Kushnir, 2015; Ordoñez et al., 2016). Carbon materials are used for anode component such as graphite and hard-carbons. These are usually the top choice used in current industrial manufacturers because these involves the combination of low cost, abundant availability, and cycle life. The anode and cathode material are linked with current collector sheets (Cu, Al) through adhesive agent like polyvinylidene fluoride (PVdF) binder. There are various electrolytes for LIBs including liquid electrolytes, polymer electrolytes. Liquid electrolytes are effectively utilized for a variety of battery applications. These consist of lithium salts ($LiPF_6$, $LiBF_4$, $LiClO_4$) dissolved in a single or combination of several organic solvents, e.g. ethylene carbonate (EC), propylene carbonate (PC), and dimethyl sulfoxide (DMSO). Polymer electrolytes have also been widely applied in portable electronics (laptops, smart phones, and tablets…) owing to their lightweight and shape flexibility of battery in any desirable configurations. Common polymer hosts used in LIBs comprise poly(acrylonitrile) (PAN), polyethylene oxide (PEO), polymethylmethacrylate (PMMA), polypropylene oxide (PPO), and many other (Zeng et al., 2014).

A typical LIB contains about 25–30% cathode, 15–30% anode (including current collector sheets), 10–15% electrolyte, 18% cell can, 3–4% separate and 10% other components (Nayaka et al., 2015; Winslow et al., 2017). The chemical composition of some typical LIBs are shown in Table 2 (Winslow et al., 2017). Fig. 1 shows material composition of a $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ battery.

3. Recycling processes

With the rapid development of lithium ion battery and electric vehicles in recent years, the recovery of lithium battery has also become a hot area of research (Liao et al., 2017). From 2008 to 2018, more than 3000 research papers are associated with this topic. In summary, the process of recycling combines two stages (see Fig. 2). At the first stage, mechanical recycle process (also known as physical process), includes disassemble, crushing, screening and separation. The purpose of this process is returning of lithium-ion batteries out of electric vehicles and separation of the cell into particles that can be directly reclaimed by chemical recovery. The main challenges in the physical process are as follows: a) Different design and connection of battery pack enclosure in EVs. b) The un-uniformity of size and shape of battery module and

Table 1
Upsides and downsides of the electrode materials (Kushnir, 2015; Ordoñez et al., 2016).

Material	Voltage vs Li/Li^+	Specific capacity ($mAh.g^{-1}$)	Volumetric capacity ($mAh.cm^{-3}$)	Advantages	Disadvantages
$LiCoO_2$	3.8	145	550	High Li^+ ion and electronic conductivity, performance	High-priced and harmful Co, low capacity.
$LiFePO_4$	3.4	170	589	Low-priced, environmentally friendly, cycle life, rate capability	Low voltage and energy density, low Li^+ ion and electronic conductivity, high processing-cost,
$LiMn_2O_4$	4.1	120	596	Low-priced and environmentally friendly, rate capability, high Li^+ ion and electronic conductivity	Cycle life, low capacity, severe capacity fade at temperature (55 °C)
$LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$	3.7	170	600	Better safety and performance than LCO, high voltage	High-priced, harmful Co, Ni, Needs cell balancing and voltage protection
Graphite	0.1	370	–	Low-priced and environmentally friendly, cell voltage	High processing-cost, formation of solid electrolyte interfacial (SEI) layer, low energy density

Table 2
Chemical composition of some typical LIBs (Winslow et al., 2017).

Research group Metals	Complete Battery, $LiCoO_2$ cathode Conc. (mass %)	Complete Battery, $LiFePO_4$ cathode Conc. (mass %)	Complete Battery, $LiMn_2O_4$ cathode Conc. (mass %)	Complete Battery, $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ cathode Conc. (mass %)
Aluminum	5.2	6.5	21.7	22.72
Cobalt	17.3	0.0	0.0	8.45
Copper	7.3	8.2	13.5	16.6
Iron/Steel	16.5	43.2	0.1	8.79
Lithium	2.0	1.2	1.4	1.28
Manganese	0.0	0.0	10.7	5.86
Nickel	1.2	0.0	0.0	14.84
Binder	2.4	0.9	3.7	1.39
Electrolyte	14.0	14.9	11.8	11.66
Plastic	4.8	4.4	4.5	3.29

different battery management system. c) The lithium ion battery may explode during the disassemble process. d) In the process of dissolving and dissolution of the battery, harmful gases and toxic substances may be produced to pollute the environment; (Zeng et al., 2015a) e) Automation of disassemble process. Based on the work of first stage, chemical recycling process (Metallurgy processes include pyro-, hydro-, bio-metallurgy and combination methods) was carried out. The purpose of second stage is to recycle precious metals and raw materials from spent battery, especially cobalt and lithium (Wang and Wu, 2017; Wang et al., 2016). The main challenges in this stage includes: a) Energy consumption b) The comprehensiveness and diversity of the recovery methods c) Environmental impacts in terms of pollutant emissions; d) Investments and costs, influenced by economies of scale; (Heelan et al., 2016) e) Efficiency of recycling (Vezzini, 2014).In addition, recycling lithium ion battery from EVs also comprise the second use of lithium ion battery. According to the research of (Natkunarajah et al., 2015), the battery cannot be used in electric vehicles as the batteries have a residual capacity of 70–80%. A secondary application of these batteries can extend the life of battery more than ten years. More research about lithium ion battery second used can be found in (Saxena et al., 2015; Sathre et al., 2015; Heymans et al., 2014; Jiao and Evans, 2016; Rohr et al., 2016).

3.1. Mechanical recycling process

3.1.1. Process of returning spent lithium ion batteries out of EVs

The battery pack enclosure is usually located at the bottom of the electric vehicle, which consists of battery modules and battery management system (BMS). The battery module is connected in series and parallels with a lithium battery so as to guarantee the high voltage and capacity. Generally, a module includes more than several hundred cells. So, the total number of batteries in an electric car may exceed 1000 (Tesla electric car has more than 7000 batteries). In the mechanical recycle process, the battery pack is dismantled into a battery module, and the battery module is dismantled into a single cell. Finally, it is crushed into granular form and screening for further recycling. With the boom of EVs, a large number of battery pack with millions spent lithium ion batteries will be a big challenge for human beings in the near future (Swain, 2017).

3.1.1.1. Disassembly planning and assessment of automation potentials for spent lithium-ion batteries. Considering the impact on the environment and resources, ways to efficiently and safely remove the lithium battery in the battery pack during the recovery process become a key research problem. Thus, researches had put forward a series of conceptions to achieve this goal (Schmitt et al., 2011; Herrmann et al., 2012a; Wegener et al., 2015, 2014; Weyrich et al., 2013; Herrmann et al., 2012b). Jan et al. presented a flexible gripper system to show how the disassembly process can be executed by automation (Schmitt et al., 2011). According to the product analysis and a criteria catalogue, an integrated methodology was proposed by Christoph et al. (Herrmann et al., 2012b) that enables the assessment of automation potentials for disassembly operations for automotive traction batteries. Weyrich et al. set a more concrete conception of semi-automated disassembly steps on

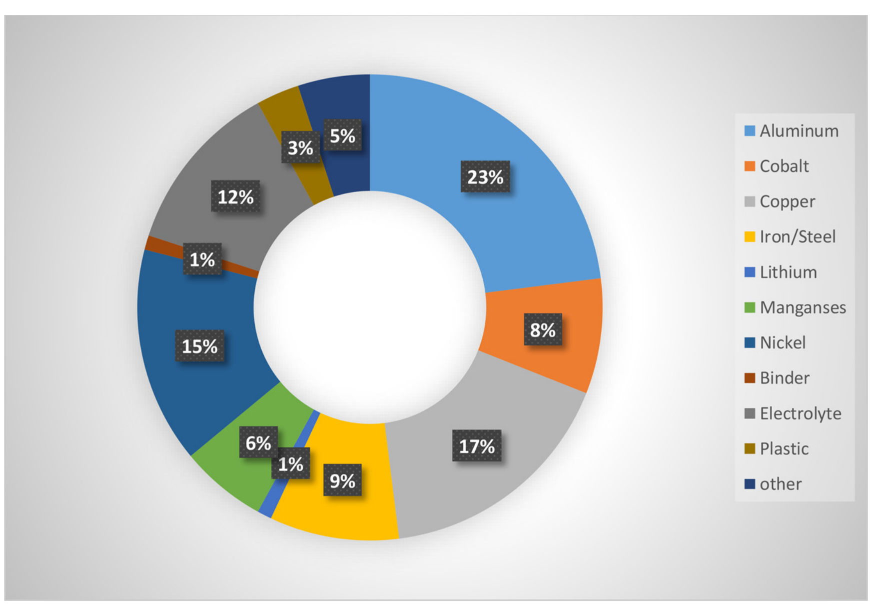

Fig. 1. Detailed composition of $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ battery.

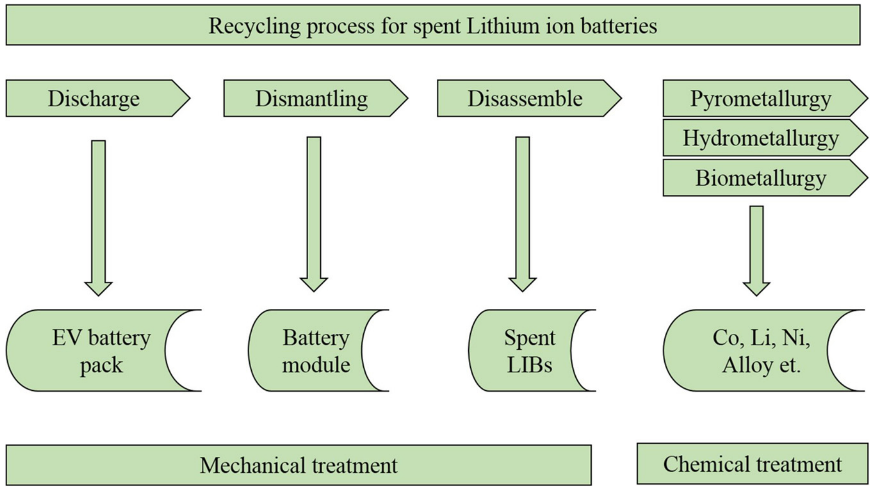

Fig. 2. A comprehensive process of recycling of lithium ion battery from EVs.

the automatically process of recycling lithium ion battery in Fig. 3 (Weyrich et al., 2013). The key point was that each components of spent battery (including battery pack, module and cell) should test its performance before dismounting. Several sensors for intelligent disassembly of pack are evaluated and a modular approach with separation-, detection-, control and sub modules is applied.

3.1.1.2. Intelligent integration of disassembly system. To improving the efficiency of recycling process, a properly disassemble sequence is necessary. There are some research had done for this topic (Xia et al., 2014; Nonomiya and Tanimizu, 2017; Hauschild et al., 2017; Yamazaki, 2017; Wang et al., 2017; Smith et al., 2016; Cong et al., 2017a; Smith et al., 2012). As is shown in Fig. 4, the methodical assessment approach was used for solving the complexity problem in dismounting EV battery pack (Herrmann et al., 2012b). To measure the parameters of battery pack, product analysis was first carried out. Second, the disassembly sequences were determined by a special software prodtect. Then the automation potential was analyzed with the monitoring of criteria checklist and the final disassembly sequences were determined in a bar chart. The advantages of this method are intuitive, simple visualization and possibility to visualize complex product structures. Furthermore, this integrated method can combine illustration of disassembly objects with joining techniques and display of disassembly times. Although some conceptions were concluded to solve the automation problem, the integrated system and intelligent workstation still play an important role in the recycling process (Song et al., 2017; Chang et al., 2017; Vongbunyong et al., 2017; Soo et al., 2017; Cong et al., 2017b; Barkmeyer et al., 2017; Bilge et al., 2017). Wegener et al. (2014) implement a new disassembly system and workstation to improve the efficiency of automation mechanical treatment base on the Audi Hybrid battery system. Disassembly matrix and disassembly priority graphs were made to assure a possible disassembly order. According to their analysis results, the efficiency of workstation will improve due to the cooperation of worker and intelligent robots. Meanwhile, the place of lithium battery recycling factory should consider the transportation cost based on the big data analysis results.

3.1.2. Further treatment of recovery processes

To recover the raw material more efficiently, the focus is on removing hazardous source and separating the components of spent LIBs. The process generally includes crashing, screening and separation. In general, the technology for secondary recycling process includes mechanical crushing and separation (Bertuol et al., 2015; He et al., 2017a; Zhang et al., 2014). Materials such as of anode, cathode, electrolyte and separator can be obtained after the proper mechanical conditioning treatment. Based on the research (Houlton, 2011), in mechanical conditioning, materials of the cells are separated by different combination,

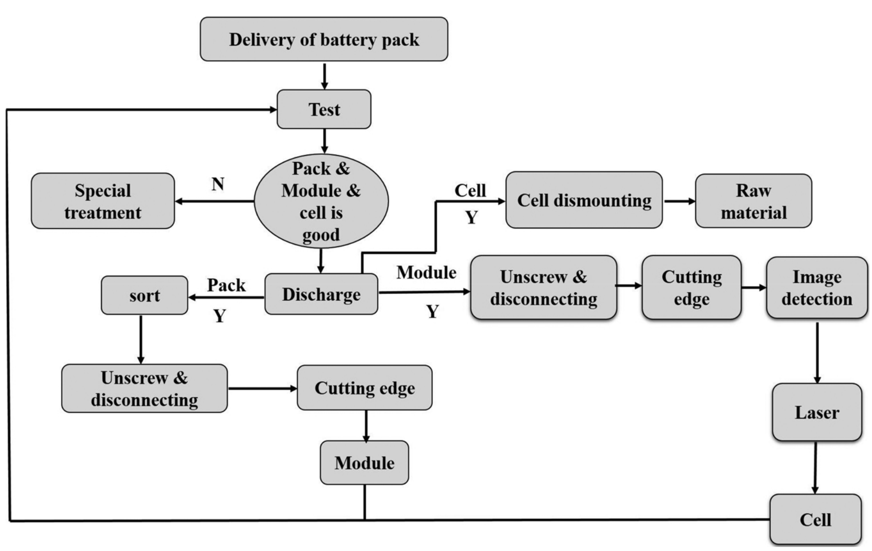

Fig. 3. Semi-automated disassembly concepts for recycling of battery pack.

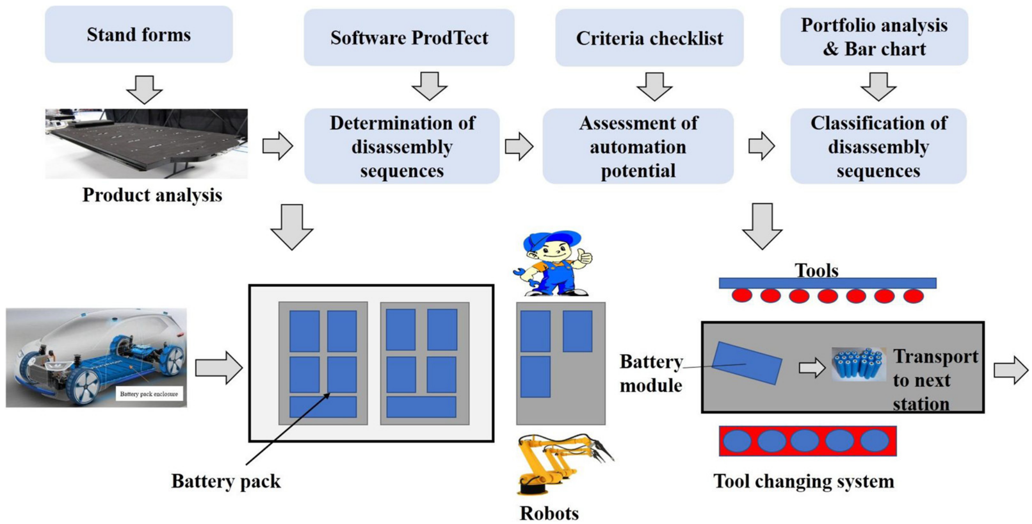

Fig. 4. Intelligent and Integrated disassembly system of Battery packs.

sizing, and concentration processes. Hanisch et al. (2015) proposed a novel approach to separate coating and foil of electrodes of spent lithium ion battery. The process was characterized by using thermogravimetric analysis, tape adhesion tests, atomic absorption spectroscopy, particle size analysis, and gravimetric sieve analysis. It can be found that 97.1% w/w of the electrode coating can be regained with aluminum impurities of only 0.1% w/ w, which is 30 times purer than the comparison process. The details about crushing step process and closed-loop Li-ion battery is given in (Diekmann et al., 2017; Gratz et al., 2014; Li et al., 2014a; Sa et al., 2015).

3.2. Metallurgy processes

After mechanical processes, the recovery methods of components on LIB cells, especially cathode materials, can be divided into hydrometallurgy, pyrometallurgy, biometallurgy and combination methods. The flow diagram of the typical recycling process is shown in Fig. 5.

3.2.1. Pyrometallurgy

Pyrometallurgy involves thermal treatment to decompose the components of spent LIBs. This process often includes two steps. Firstly, spent LIBs are provided a low temperature in a furnace in order to reduce the danger of burst and evaporate the electrolyte. Secondly, all plastics and solvents are burned at a higher temperature, followed by formation of slag and alloy. The alloy consists of cobalt, nickel, copper, and iron, which is often subjected to further processing using hydro- and bio-metallurgical processes. Currently, pyrometallurgical technologies have been yoked effectively to aqueous processes by some business such as Umicore, Accurec, Sony, Onto and Inmetco (Meshram

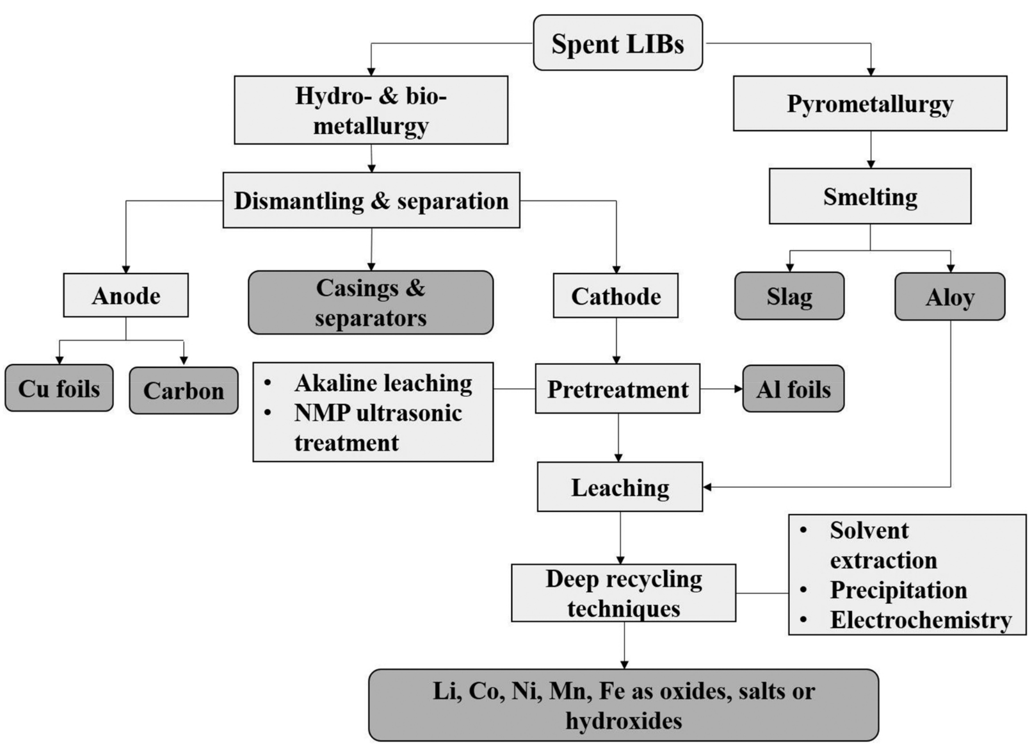

Fig. 5. The flow-chart showing the typical recycling process.

et al., 2015a). A slag including aluminum, manganese, and lithium is obtained and used for construction materials (Xiao et al., 2017).

In this method, copper, nickel and cobalt could be recycled effectively, resulting in purified and gathered alloys. Other harmful chemicals such as: solvents, plastics are burned that serve as an important role to provide a lot of process-energy and remove their toxicity. In addition, many spent LIBs can be treated at the same time, making a significant contribution to large-scale production costs and simplified operation. Pyromatallurgy have been utilized widely in manufacturing industries. However, this technology has the main disadvantage of energy-consumption due to high temperature operation and associated to extra instruments for the control of toxic gases. Moreover, all organic electrolytes, binders, acetylene black, and some others are mostly burned off. Lithium and manganese cannot be recycled because it is trapped in the slag of complex materials. The economic is not beneficial for batteries without containing Ni or Co metals, leaving an open-question to the recycling procedure for LIBs in the future (Meshram et al., 2015a).

3.2.2. Hydrometallurgy and biometallurgy

Initially all spent LIBs are discharged, dismantled, and separated into outer casing, cathode, anode, and separator (Xu et al., 2008). Currently, the main objective of these processes is to regain the valuable metals from cathode; it consists of pre-treatment, leaching, deep recycling techniques. Pre-treatment including NMP-dissolving, ultrasonic treatment, alkaline-leaching is first carried out to separate active material from Al foil. Then, leaching treatment including acid-leaching and bioleaching is used as important technique to bring valuable metals into solution. Finally, the metals in solution are more easily separated by deep recycling techniques such as solvent extraction, precipitation or electrochemistry (Xu et al., 2008; Yang et al., 2015).

3.2.2.1. Pre-treatment. Due to adhering tightly between active materials and Al foil in cathode by binders like PVdF, PTFE, many methods have been tested to effectively separate active materials from Al foil, including alkaline leaching and NMP dissolution often assisted by ultrasonic treatment and solvent extraction (Yang et al., 2015; Li et al., 2017).

In the alkaline leaching method, the cathodes are immersed in Sodium Hydroxide (NaOH) solution to dissolve Al foils into solution. After washing with water and filtration, Al foils and active materials are effectively separated (Li et al., 2017). The reaction of Al foils in NaOH is as following:

$$2Al + 2NaOH + 2H_2O \rightarrow 2NaAlO_2 + 3H_2 \quad (1)$$

Currently, NMP ultrasonic treatment have been widely employed to take advantage of both NMP dissolving and ultrasonic treatment. Yang et al. carried out an experiment at room temperature using an ultrasonic system with NMP as ultrasonic solvent. According to the experimental results, this method not only has the benefit of decreasing the separation time, but also is considered as the beneficial way on environmental health because the evaporation of the NMP at 100 °C is extremely toxic and unhealthy for the community (Yang et al., 2015). However, this method has drawback of the NMP being too expensive and is impractical for scale-up industry. Therefore, a variety of solvents have been investigated comprising dimethyl sulfoxide (DMSO), N, N-dimethylacetamide (DMAC), and N, N-dimethylformamide (DMF). To date, the DMSO solvent employed at 60 °C in 85 min seems to be the effective one because of its economic benefits and environmental health (Kumar et al., 2017).

3.2.2.2. Acid-leaching. In recent years, acid leaching is the most widely utilized method in both academic research and industrial application (Hu et al., 2017). This method benefits from the chemical properties of metals in aqueous solution to isolate and recover a marketable product. In this method, valuable metals from cathode materials are leached out, utilizing either inorganic acids, e.g. H_2SO_4, HNO_3, HCl, or various organic acids, e.g. succinic, ascorbic, aspartic, malic, oxalic and citric as leaching agents, often supported by reducing agents (e.g., H_2O_2, $NaHSO_3$, Na_2SO_3) which make the various metal-forms easily dissolve in the acid solutions through oxidizing them to higher oxidation states (Li et al., 2013; Gratz et al., 2014).

Joulié et al. studied the leaching of $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ cathode by sulfuric, nitric and hydrochloric acids. It has been presented that the leaching-efficiency is highest in HCl solution because the presence of chloride ions decreases stability of surface-layer formation, giving rise to the dissolution process. According to experiments, the ideal conditions for leaching out all valuable metals were 4 M solution of HCl with at 90 °C and the leaching time of 18 h (Jouli et al., 2014). Meshram et al. studied the leaching of $LiCoO_2$, $Li_2CoMn_3O_8$ and $(Li_{0.85}Ni_{0.05})(NiO_2)$ cathodes by H_2SO_4 assisted by $NaHSO_3$ as reducing agent. This experiment indicated that 87.9% of Mn, 96.4% of Ni, 91.6% of Co and 96.7% Li could be gained from cathodes under the optimum conditions with 0.075 M solution of $NaHSO_3$ and 1 M solution of H_2SO_4 at 95 °C and the leaching time of 4 h (Meshram et al., 2015b). However, compared to organic acids, using inorganic acids for leaching are not beneficial for economic point view. In addition to that, it releases some toxic gases (eg. Cl_2, NO_x and SO_3), maybe resulting in serious environmental and human-health problems if special equipment installed to treat these gases is not available.

For handling these issues, organic acids are employed as alternative choices. Zeng et al. investigated utilizing oxalic acid for the process of leaching $LiCoO_2$ without any supports from reducing agents. The optimum conditions were controlled at 15 g L^{-1} solid–liquid ratio at 90 °C, leaching time of 150 min and rotation rate of 400 rpm, the recycling proportion of cobalt and lithium may reach to 97% and 98%, respectively (Meshram et al., 2015b). The leaching reaction of $LiCoO_2$ in oxalic acid is as following:

$$2LiCoO_2 + 4H_2C_2O_4 \rightarrow LiHC_2O_4 + 2CoC_2O_4 + 2CO_2 + 4H_2O \quad (2)$$

The experiment carried out by Li et al. indicated that around 90% of Co and nearly 100% of Li were recovered using citric or malic acids with H_2O_2 as reducing agent. Aspartic acid was fairly less efficient because of lower solubility in water and its weak acidity (Li et al., 2013).

3.2.2.3. Bioleaching. Bioleaching have been considered as a hopeful technique to supplant traditional acid-leaching because of its environmentally friendly and low-cost procedure and higher efficiency. This process takes advantage of interaction between low-grade waste or ores and living microorganisms (involving fungi and bacteria) to transform insoluble metals into solution (Horeh et al., 2016; Chen et al., 2015a).

In bacterial bioleaching, the autotrophic-cells involving iron(II)-oxidizing bacteria and sulfur compound oxidation are often utilized to generate a number of metabolites like ferric ions and acid sulfuric. Xin et al. studied this method for the first time to regain some precious metals from cathode materials for LIBs (eg. $LiFePO_4$, $LiMn_2O_4$ and $LiNi_xCo_yMn_{1-x-y}O_2$). According to experiment, 98% of Li from $LiFePO_4$, 96% of Mn and 95% of Li from $LiMnO_2$, and around 95% of four metals from $LiNi_xCo_yMn_{1-x-y}O_2$ were regained (Xin et al., 2015).

In comparison with bacterial leaching, fungal leaching has more ability to tolerate toxic materials. To date, a variety of fungi (eg. Penicillium chrysogenum, Aspergillus niger, and Penicillium simplicissimum) could employed to excrete metabolites as organic acids that play a role as dissolving and chelating agents with metals from a variety of waste materials and ores. Hored et al. investigated the fungal bioleaching of spent LIBs using Aspergillus niger in the wide-ranging conditions to recycle Ni, Co, Al, Mn, Li, and Cu metals. The experimental results indicated that 38% of Ni, 45% of Co, 65% of Al, 70% of Mn, 95% of Li and 100% of Cu were recovered (Horeh et al., 2016).

3.2.2.4. Deep recycling techniques. After a number of pre-treatment and leaching steps, the valuable metals could be selectively separated from mixtures of leach liquor by a variety of methods (eg. solvent extraction, precipitation, electrochemistry, and combination methods). Solvent extraction method is usually employed to extract and separate many metals from filtrate by using many extractants like di-(2-ethylhexyl) phosphoric acid (D2EHPA), 2-hydroxy-5-nonylbenzaldoxime (ACORGA-M5640), bis-(2,4,4-tri-methyl-pentyl)phosphinic acid (Cyanex 272), Ionquest 801 (2-ethyl-hexylphosphonic acid mono-2-ethylhexyl ester), 2-ethylhexyl phosphonic acid mono-2- ethylhexyl ester (PC-88A) or diethylhexyl phosphoric acid (DEHPA) (Kumar et al., 2017; Liang et al., 2017; Xiao et al., 2017). The chemical precipitation method is also utilized to precipitate precious metals. Lithium can be precipitated by treating the leaching solution with oxalic acid ($H_2C_2O_4$), sodium carbonate (Na_2CO_3) or phosphoric acid (H_3PO_4) (Xu et al., 2008; Yang et al., 2015; Li et al., 2017; Hu et al., 2017; Li et al., 2013; Jouli et al., 2014; Meshram et al., 2015b; He et al., 2017b). Other metals such as Co, Ni, Mn are often precipitated with NaOH, ammonium hydroxide (NH_4OH), dimethylglyoxime & potassium permanganate ($KMnO_4$) (He et al., 2017b; Li et al., 2014b; Sun and Qiu, 2012; Zeng et al., 2015b; Li et al., 2012, 2015; Nayaka et al., 2016a,b; Ku et al., 2016; Xin et al., 2015; Horeh et al., 2016; Chen et al., 2015a; Freitas et al., 2010). In fact, both methods are usually combined to gain highly effective separation.

Chen et al. (2015a) investigated hydrometallurgy to recover some valuable metals from sulfuric acid leaching liquor of cathodes from spent LIBs. Firstly, nickel ions were selectively precipitated and recovered utilizing dimethylglyoxime after purification operation. Secondly, Mn and Co was separated utilizing Co-loaded D2EHPA through solvent-extraction process. Finally, In order to regain Li and Co, sodium carbonate and ammonium oxalate were employed to precipitate them in the forms of Li_2CO_3 and $CoC_2O_4.2H_2O$, respectively. The proportion of recycling efficiency under optimum conditions gained as following: 81% for Li, 98% for Co, 99% for Ni, 97% for Mn. Meshram et al. (2015b) studied the precipitation of Li, Mn, Co, Ni, from leaching solution. According to experiments, over 98% cobalt was separated by oxalic acid, 98% Li was precipitated and regained as carbonates by Na_2CO_3, followed by separating Ni and Mn at pH 9.0 and 7.5, respectively (Table 3).

Electrochemistry is applied to regain and separate metals from leaching solution. Nevertheless, technique tends to utilize too much electricity, resulting to economic efficiency aspects (Freitas et al., 2010). Table 4 show some separation techniques by previous studies.

4. Critical gaps and new directions of research

4.1. Challenges in the process of mechanical condition

(1) Automatic and intelligent recovery system

There are many EVs manufacturers, but most of them own different design of battery pack. Some even use the battery made by themselves. (Tesla and BYD, for instance). It will lead to that many different screw types that have been used for the joints of battery pack of EVs and not all screws are accessible from the same direction. Besides, flexible components such as cables and joints that are difficult to access such as the plug-in connectors of the BMS. Therefore, the automatic and intelligent recovery system are necessary for the disassemble process. Government should make industrial standards for electrical vehicles.

(2) Efficiency and safety disassemble of battery pack

Efficiency in the recycle process mostly depends on the recover methods and tools. While disassemble in the industry still rely on workers. Its will be a big challenge due to the boom of EVs in 2020. Furthermore, the surplus capacity of spent lithium ion battery can cause fire easily; especially the due to amounts of batteries being stacked together. To avoid short circuit inside of battery and explosion during the dismantling process, the battery first undergoes discharging considering its safety.

(3) Adjustment of chaos in recycling market

Recycle problems exist throughout the whole world due to the high cost of recycling lithium ion battery and lack of effective recycling technology. The recycling policies supplied by the government are different from each other. Even with the boom of electrical vehicles market attracting attention from the world (Hoyer et al., 2013), recycling lithium ion battery still lack strong polices to be supported in most of fields. The recycling conception of government should be redesigned and recycling technology of raw material from waste batteries need to be improved in the future.

4.2. Critical gaps in the metallurgy processes

(1) The lack of recovery processes for slag, electrolyte, and anode

Table 3
Summarizes some relevant literatures (2012–2017) used to recover metals from spent LIBs.

Types	Reagents	Temp (°C)	Time (h)	Metal recovery rates (%)	Ref
$LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$	1–4 M H_2SO_4/HNO_3/HCl	25–90	3–18	> 80% Li, 100% Co, 99.99% Ni	Jouli et al. (2014)
$LiCoO_2$, $Li_2CoMn_3O_8$ and $(Li_{0.85}Ni_{0.05})(NiO_2)$	1M H_2SO_4 + 0.075M $NaHSO_3$	95	4	96.7% Li, 91.6% Co, 96.4% Ni, 87.9% Mn.	Meshram et al. (2015b)
$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	1M H_2SO_4, 1 vol% H_2O_2	40	1	Co, Li, Mn, and Ni > 99.7%	He et al. (2017b)
$LiCoO_2$	H_2SO_4/ HCl/ citric acid + 0.55 M H_2O_2	60	5	100% Li, 96% Co	Li et al. (2014b)
$LiCoO_2$	1.0 M Oxalate + H_2O_2	80	2	Li/Co > 98%	Sun and Qiu (2012)
$LiCoO_2$	1.0 M Oxalate	95	2.5	98% Li, 97% Co	Zeng et al. (2015b)
$LiCoO_2$	(0.5–2.0 M) Citric acid/Malic acid/Aspartic acid + (1.0–6.0) vol.% H_2O_2	90	2	100% Li, 90% Co	Li et al. (2013)
$LiCoO_2$	1.25 M Ascorbic acid + H_2O_2	70	1/3	98.5% Li, 94.8% Co	Li et al. (2012)
$LiCoO_2$	1.5 M Succinic acid + 1.0 vol.%H_2O_2	70	2/3	96% Li, 100% Co	Li et al. (2015)
$LiCoO_2$	1 M iminodiacetic acid + 0.02 M maleic acid + ascorbic acid	80	6	99% Li, 91% Co	Nayaka et al. (2016a)
$LiCoO_2$	1.5 M phosphoric acid 0.02 M glucose	80	2	98% Co, 100% Li	Nayaka et al. (2016b)
$LiCoO_2$	0.5 M Glycine + 0.02 M ascorbic acid	80	6	95% Co	Nayaka et al. (2016b)
$LiMn_2O_4$, $LiCo_xMn_yNi_zO_2$ and Al_2O_3	NH3 + $(NH_4)_2SO_3$ + $(NH_4)_2CO_3$	80	1	80%Co, 100%Cu 2%Al, 1% Mn, 25%Ni	Ku et al. (2016)

Table 4
Separation techniques from past studies.

Separation method	Sample	Additive chemicals	References
Solvent extraction	$LiCoO_2$	Cyanex272	Swain et al. (2008)
	Mixed types of LIBs	Cyanex272	Kang et al. (2010)
	$LiCoO_2$	Acorga M5640, Ionquest 801, etc.	Pranolo et al. (2010)
	Mixed types of LIBs	Cyanex 272	Provazi et al. (2011)
	Mixed types of LIBs	Cyanex272 and PC-88A	Zhao et al. (2011)
	$LiCoO_2$	PC-88A	Chen et al. (2011)
	$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$, $LiCoO_2$ and $LiMnO_2$	D2EHPA	Chen et al. (2015a)
Precipitation	$LiCoO_2$	NH_4OH	Dorella and Mansur (2007)
	$LiCoO_2$	NaOH	Li et al. (2009)
	Mixed types of LIBs	NaOH and Na_3PO_3	Chen et al. (2015b)
	Mixed types of LIBs	Dimethylglyoxime, $(NH_4)_2C_2O_4$, Na_2CO_3	Chen et al. (2015a)
Electrochemistry	Mixed types of LIBs	–	Lupi et al. (2005)
	Mixed types of LIBs	–	Freitas et al. (2010)

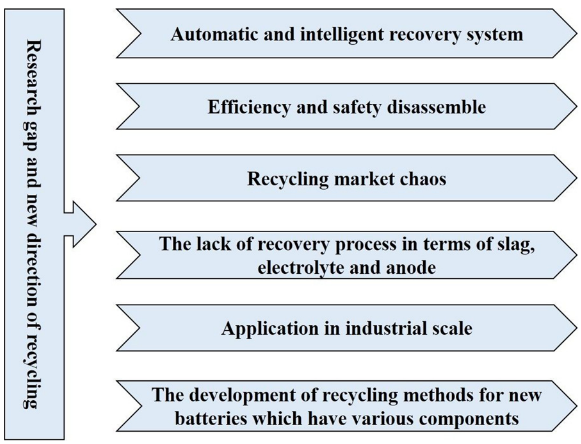

Fig. 6. Critical gaps with new research directions for recycling of battery pack.

Recycling spent LIBs in the long term is mainly for legislation about environmental aspects, because the profits from selling recovered metals is less economically attractive. Presently, many studies have been focused on recycling of precious metals from spent LIBs such as cobalt, nickel or a small part of lithium. Thus, recycling lithium and other toxic components from slag (in pyrometallurgy process), electrolyte and anode active materials (in hyro- and bio-metallurgy processes) are significantly important in an environmental point of view (Fig. 6). The research in this aspect needs thorough attention.

(2) Application in industrial scale

Most of the researches in field of hydro- and bio- metallurgy have been carried out in pilot scale and are extremely complex with a series of long processes, followed by high-cost operation (Kaya, 2016). More research needs to be conducted on optimizing procedure with eco-efficiency, to apply to industrial scale.

(3) Development of recycling methods for new batteries having various components

Manufacturing technologies for LIBs is evolving in future such as the altering of components of LIBs. This shall also result in conducting further research on redesign of recycling framework for Li-ion batteries.

5. Framework for recycling of battery pack

Higher degree of automation and intelligence in the mechanical dismantling process were the main challenges for recycling of lithium ion battery pack from EVs. Most of the solutions proposed by researchers were at the stage of conception. From Fig. 7, we can draw a conclusion that both government incentive polices and effective recycling technology can contribute to pushing the process of conception to practicability. Once the mechanical automatically system and intelligent database exchange system enter the practical stage, the environment of recycling spent battery will improve and achieve the recycle goal with lower cost. To reduce the pollution of the waste battery

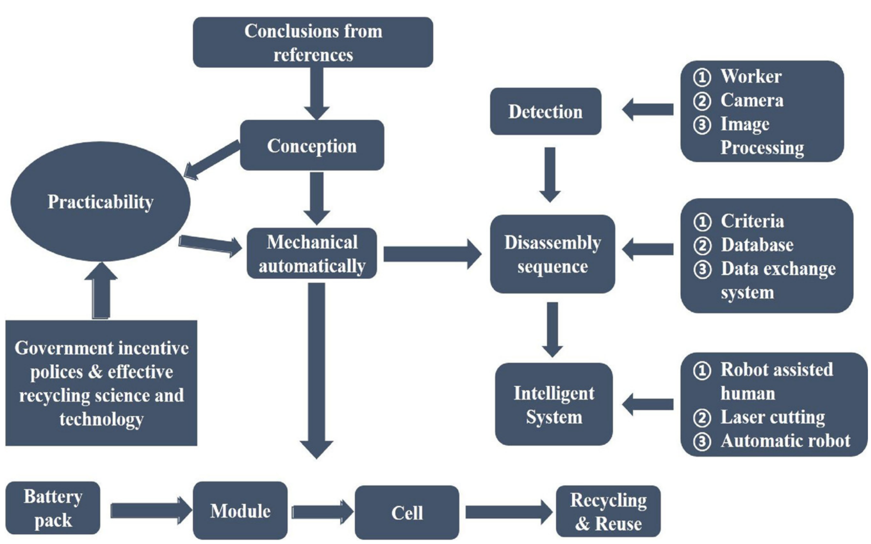

Fig. 7. A Proposed Intelligent and integrated framework for the recycling of battery packs from EVs.

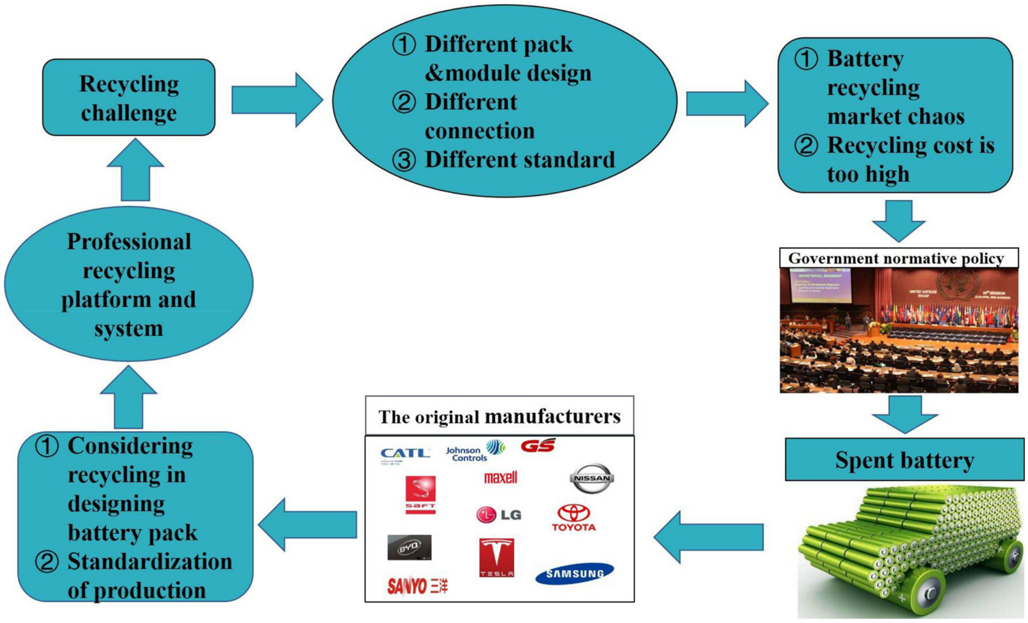

Fig. 8. A conception of virtuous circle in recycling of lithium ion battery.

to the environment and recycle the raw material more efficiently, the government should put more efforts on the recycling market with a series of polices and regulations. At the same time, the industrial sector should make strict industry standards to adjust the chaotic recycling market (G.O. INDIA, 2016). It would ensure that recycled products meet the same high-quality standards as virgin materials and thereby be accepted for reuse.

Practicability solutions suggest that spent batteries could be returned to the original manufacturers. (In Europe this may be a requirement but most countries still lack these kind of policies) Such policies in Fig. 8 could bring some new design-for-recycling guidelines to the battery manufactures so that all batteries will be design with recycling in mind (Gaines, 2014). It is obvious that the same infrastructure could easily be used for recycling of lithium ion batteries. All the efforts will bring a virtuous circle to recycling lithium ion battery in the future.

6. Conclusions

With the boom of EVs in the world and shortage of raw material of lithium ion battery, the ways to solve recycling problem will be a big challenge in the near future. Higher cost and lack of efficient and intelligent technology are the main challenges in the process of recycling of lithium ion batteries from battery packs of electric vehicle. Most of the research work about automation of dismantling EV battery pack to modules and cells are still at the conception level. In this review paper, we propose a new procedure by combining the mechanical procedures and metallurgical processes involving recycling of battery pack. The critical gaps from the study were concluded and six research directions of recycling of lithium ion battery pack were as follows: (i) automatic and intelligent recovery system, (ii) efficiency and safety disassemble of battery pack (iii) Adjustment of Chaos in recycling market (iv) Recovery processes for slag, electrolyte and anode, (v) Application in industrial scale, and (vi) development of recycling methods for new batteries having components with different properties. Further research work could focus on validating the proposed framework to realize its practicability. In addition, artificial intelligence (AI) methods such as genetic programming variants as mentioned in (Garg et al., 2017a,b,c) can be studied with the integration of other AI methods (for instance, artificial neural networks (Huang et al., 2018) and support vector machines) for obtaining higher reliability, accuracy and robustness of the proposed framework.

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