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Review and selection of recycling technology for lithium-ion batteries made for EV application - A life cycle perspective

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Abstract

The lithium-ion battery is the source of renewable energy and the battery-operated vehicles are gradually replacing fossil fuel-based automobiles. Though Electric Vehicles (EVs) do not produce emissions through tailpipes, yet the issues pertaining to recycling of the lithium-ion batteries employed in EVs lead to environmental burdens. The EVs sale in global market exceeds a million per year. It is estimated that there will be a huge amount of unprocessed waste of lithium-ion battery packs when these vehicles retire after the service life. The re-use of batteries can reduce the waste generation, however the cumulative burden of the battery wastes will be substantial considering the increasing trend of the electric-vehicle market. There are various methods employed for recycling of lithium-ion batteries. The existing literature reports that their environmental impact is significant. The challenge is to choose a method which causes minimal disruptions to the environment in terms of cost, pollution and energy consumption, which can be effectively addressed by a life cycle analysis based selection method. In this paper, a review is conducted on the current recycling technologies and WPM (Weighted Product Method) based Multi-Criteria Approach is employed to optimally choose the best recycling process for lithium-ion batteries from life cycle perspective.

Keywords: Electric Vehicles (EVs), Lithium-ion battery (LIB) recycling, life cycle analysis, WPM, MCDM

1. Introduction

The EV (Electric Vehicles) is the sustainable transportation technology that affects the environment with little to no effect. The EVs that run with electric motors using renewable chemical energy stored in rechargeable batteries contribute to a greater extent in curbing the greenhouse gas emissions as compared to those that are based on petrol or diesel. Though it is environmentally sustaining mobility, yet it has certain drawbacks in terms of managing the retired batteries after useful life. The upward sales of EVs as shown in Fig. 1 will lead to the exponential growth of accumulation of end-of-life LIBs (Lithium-ion battery) in 10 years down the road, consequently, recycling of these LIBs is crucial to actualize the environmental sustainability. A significant amount of recovered material from the used LIBs can be reused in the LIBs that are remanufactured [1].

In this regard, enhancing the life cycle performances of the LIBs, such as reliability issues, energy efficacy, etc. and the investigation of the effects of recycling on environmental burdens are to be examined. This work focuses on the latter. Several recycling processes of used Li-ion batteries are being proposed and each has its advantages and disadvantages [2,3] and several researches are performed to help in the development of improved recycling methods.

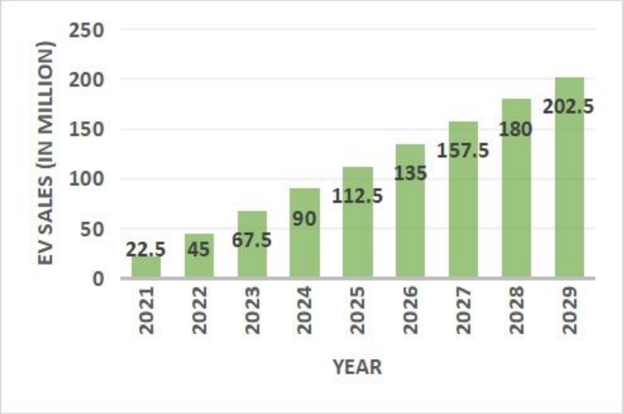

Fig. 1: EV sales trend [32]

One of the promising research on battery recycling is separation technology [4]. In this technology, the material is physically treated in order to obtain variety of waste materials to ensure that the separation process is efficient. Subsequently the metals or impurities from the waste streams are extracted by a selected metallurgic process thus recovering useful compounds by incorporating the principles of materials engineering. There are three major recycling methods based on separation technology, namely, Pyrometallurgical recovery, Hydrometallurgical reclamations, and Direct recycling. These are extensively discussed in the literature [5]. The hydrometallurgy recovers metals with high degree of purity with greater efficiency and limited energy input. Whereas, Pyro-metallurgical processes suffers poor Li recovery rate with high capital costs and energy consumption but it is a simpler process that can treat all LIBs with different material chemistries [6]. In a joint project supported by - ACCUREC Recycling and UVR-FIA, a recycling process was developed by combining a mechanical pretreatment with hydro- and pyrometallurgical process steps [7]. However, the recycling efficiency is found to be poor as the electrolyte is not recovered.

In another study, the aging factors such as C-rate, DoD (Depth of Discharge), voltage and current were considered to analyse the remaining life of used batteries [8]. When the batteries after 2nd life are recycled, the recovery of materials may not be economically viable after extended re-use, and moreover, the burden on the environment is inevitable. Some research findings reported that the energy-intensive metallurgical methods such as electro-hydraulic fermentation are used to recycle end-of-life batteries [9], but their practical utility is far from reality. The material recovery from cathode material by breaking it into simple compounds is neither efficient nor economical [10]. The energy and environmental costs of the state-of-the-art lithium-ion batteries were evaluated based on various factors like elemental abundance, toxicity, synthetic methods and scalability as discussed in [11].

It is obvious that the virgin material supply could be restricted to a great extent by the production and supply of the recovered material. For example, it is possible to improve the yield of cobalt from the used Lithium-ion batteries by a modified hydrometallurgical process. On the other hand, the environmental impact of recycling processes should also be evaluated [12,13], and recycling of too many aged batteries could be little expensive[14]. The economies, material recovery, environmental impacts and operational efficiency of recycling also depend on factors like battery usage and its health condition, composition and type of electrode material, density, battery technology, etc. [15-18]. Considering the prevailing issues and challenges, it is crucial to choose a better recycling method that should have better life cycle performance. However, identifying suitable sustainable recycling methods for EV's lithium - ion batteries remains complex as the available methods are complex, and are not researched well in life cycle perspective. In this paper, an attempt is made to review and choose the best recycling technology based on various life cycle factors. A well- established WPM based MCDM (Multi-Criteria Decision Making) is employed to address this issue.

2. Recycling technologies

To begin the selection process, the three major methods normally adopted for Lithium-ion battery recycling are discussed under this section. The schematic of the process flow of all three methods, i.e., Pyrometallurgical recovery (Pyro.), Hydrometallurgical metals reclamation (Hydro.), and Direct recycling (Dir.) is represented in Fig. 2.

2.1 Pyrometallurgical recovery

In this process, a high-temperature furnace is employed to smelt the battery parts to reduce the metal oxides to alloys of metals including cobalt, copper, iron and nickel that are present in the cathode. This process produces metal alloy fractions, gases, and slag. The gases generated at lower temperatures are the organic compounds derived from the electrolyte and binder materials. Whereas, the polymers present in the battery will get disintegrated and burn off. The metal alloys separated in this process will not only be sent for re-manufacturing of battery but can also be used in other industries like the cement processing units. It is to be mentioned that certain level of safety hazard is associated with this process because of the exposure of the cell and modules to high temperature in the presence of reductant during metal reclamation.

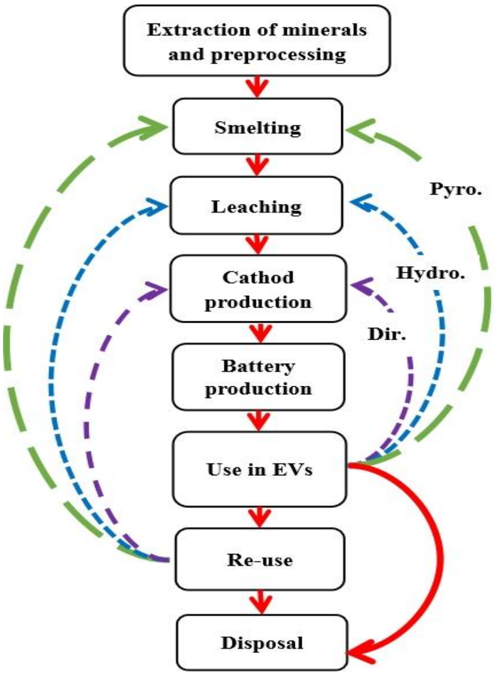

Fig. 2: Recycling technologies

Moreover, toxic gases are generated when the plastics and electrolytes are burnt. High energy consumption is the another issue with this process. Inspite of having environmentally bad effects like release of toxic gases, heavy energy costs, and poor material reclamation, this remains a widely employed method for recovering high valued elements like as cobalt and nickel due to the low production cost. Moreover, since the process is not so complex comparatively, it can be more reliable and easily maintainable. However, it scores poorly in energy usage, waste generation, and release of greenhouse gases which have a significant impact on the environment [2-5].

2.2 Hydrometallurgical metals reclamation

This method involves crushing or shredding of battery cells and dissolving them in an aqueous solution to leach and reclaim the metal elements from cathode material. After leaching, series of precipitation reactions are induced by adjusting pH value of the solution to help recover the metals. Cobalt is separated as carbonate, sulphate, oxalate or hydroxide, and then and the lithium is dissociated through precipitation. The major issues associated with hydrometallurgical process are: expensive neutralization process, low speed of delamination, contamination of the extracted materials, and high solvents consumption. Mechanical shredding is performed as the preliminary process requirement before sending the battery for the hydrometallurgical process. This will end up with generation of recovered material which has reduced particle size, material's passivation, low grade of purity, difficulty in separating cathode elements from anode material. Moreover, the cell- binding material, polyvinylidene fluoride needs further processing. Also, it is understood that the cell design currently in practice renders the recycling more complex. In this process, a huge amount of wastewater, waste solid, and off-gases are generated.

Energy consumption along with processing costs is too high. The quantity of the recovered material is reasonable but with compromising quality[2-5].

2.3 Direct recycling

In direct recycling method, the materials are removed from the electrodes of used LIB, and reused in the batteries to be remanufactured. Basically, metal oxide- cathode materials are added in the new cathode material with little morphological changes. Also, cathode strips collected from the dismantled batteries are usually soaked in NMP (N- Methyl-2-Pyrrolidone) before undergoing sonication, a process which uses sound energy to separate particles from the mixture of multiple compounds. The direct recycling method does not involve extensive and expensive purification process, and hence it is suitable for remanufacturing of low valued cathode where production process for the cathode oxide contributes to high cathode costs, more energy consumption and emission of carbon dioxide. However, the quantity of recovered material is high compared to other methods. Despite the fact that the direct recycling has many potential benefits, some of the obstacles remain intractable. The process efficiency determines the health condition of the battery and making it unsuitable for the batteries with low state of charges. Sometimes the battery after 10 years of service life re-used in a second- hand application presents challenges in an industry where the rapid evolution of battery materials is taking place. There are some quality and process-related issues in direct recycling due to direct mixing of all cathode and anode materials. If the quality of the recycled material is poor, then it will be critical to sort out the materials into goods and bads, and therefore, an efficient and effective sorting should be advised here. This will not only make the process complex but also increase life cycle costs as the factors like failures and the energy consumption become multiplicative. In addition, pyrolysis process that is used to remove the electrode binder material pose further challenges. This includes the formation of hazardous by-products from PVDF (Polyvinylidene fluoride) binder through pyrolysis or from the application NMP (N-Methyl-2-pyrrolidone) for dissolution of the material [2-5].

From the above, it can be seen that each method has strengths and weaknesses, and it is thus difficult to recognize the best method to recycle the used LIBs. In this work, MCDM is applied explicitly to resolve this issue by taking into account various factors that impact the life cycle of the recycling process.

3. Selection of battery recycling method

In this section the factors, which affect the life cycle performance of the recycling method are considered for choosing best recycling methods using WPM (Weighted Product Method) approach. The LCA (Life Cycle Analysis) is the most important process for any product or process. The schematic of typical life cycle analysis for the battery recycling process is developed and shown in Fig. 3.

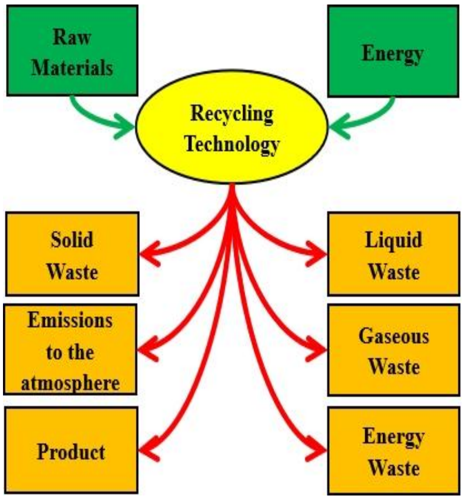

Fig. 3: A typical LCA model for battery recycling process

Based on the literature [18-20], an MCDM method is suggested to choose the best method for recycling based on the life cycle performance. In this method, various factors associated with the recycling technology are picked up, evaluated, and weighted based on the degree of importance and merged to get a performance score that is subsequently used to rank the recycling technologies from life cycle perspective. The following factors are considered in this context:

A. Capital cost (C): This cost involves the initial expenditures that are incurred in establishing the technology of recycling. It represents the cost of the machinery, facility location planning, material handling and other resources. Though the capital cost does not directly affect the life cycle performance, yet, it is worth considering this factor as it is all about capital investment and return on investment can be realized only there is a marginal initial investment on establishing the process. The process that attracts more capital investment will not be normally preferred.

B. Processing cost (P): The production cost is the cost that includes the expenses of labour, raw materials, consumable manufacturing supplies etc. in order to carry on a particular recycling technology. This is one of the most important factors that stumbles across the widespread use of electric vehicles. Hence the production cost of the recycling process for EV batteries must not be too much.

C. Energy utilization (E): Energy usage refers to all the energy used to perform a particular operation. Appropriate utilization and conservation of energy would make the engineering processes more sustainable [22,23]. The energy usage must be efficient in any engineering process and the recycling technology is no exception. Efficient energy usage means using less energy to provide the same level of energy.

D. Waste produced (W): Circular economy emphasizes the continual use of resources by eliminating wastes. Waste generation from any engineering process helps to measure its sustainability [24-27]. It includes all the materials discarded into the landfills while carrying on the recycling technology. Our aim is to obtain a better sustainable environment, so lower the generation of waste, better the recycling technology.

E. Reliability (R): The degree of failure of any production process clearly reflects the extent of reliability and sustainability of the process[28,29,33]. The complexity of the recycling technology will render the process vulnerable to failures. More failure, poor reliability, and thus high operational costs.

F. Maintainability (M): Maintainability is the ability of an engineering system or process to be able to be maintained or repaired [30,31]. At times, the poor repairability of the technology may result in heavy maintenance cost. If the recycling technology is complex, the repair rate will be very high and will lead to increase in operational cost.

The selected factors are provided with suitable rating [Refer Table 1]. The rating may vary from Low (L) to High (H) and in between the codes are given as "Fairly Low" (FL), Medium (M) and "Fairly High" (FH). In order to convert these ratings into quantifiable data, a standard scale with a range of 0 - 10 has been selected, and appropriate numerical value within this range is assigned to these ratings. The similar procedure has been widely employed for solving several multi-criterion selection problems [17-19]. According to this, each rating is given a performance code on the scale of 0-10. The performance code is to be normalized on the basis of the type of factor, be it beneficial or non-beneficial. The performance code represents the performance of each battery recycling technology and will vary based on the value of each factor. When the performance of the selected factor is 'Low' or 'Fairly Low', then performance code is assigned as 1 (Low) or 3 (Fairly Low), and if it is 'Medium' or 'Fairly High', then the code is assigned as 5 (Medium) or 7 (Fairly High) respectively. The performance code for 'High' is assigned as 9. The selected range of scale can effectively show the performance of the selected factors. Table 1 presents the performance rating for the factors of each recycling technology, and Table 2 presents the performance code of the factors for each recycling technology.

The performance ratings for the recycling processes are selected based on the literature (Refer to section I and II) and extensive interaction with technical managers from the established battery recyclers.

Table 1: Performance rating

Recycling technology	**Factors**					
	C	**P**	**E**	**W**	**R**	**M**
Pyrometallurgy	**H**	**L**	**H**	**FH**	**FH**	**FH**
Hydrometallurgy	**M**	**FL**	**M**	**M**	**FH**	**FH**
Direct recycling	**M**	**FH**	**M**	**FL**	**M**	**FL**

Table 2: Performance code

Recycling technology	**Factors**					
	C	**P**	**E**	**W**	**R**	**M**
Pyrometallurgy	9	1	9	7	7	7
Hydrometallurgy	5	3	5	5	7	7
Direct recycling	5	7	5	3	5	3

Table 2 shows performance code of various factors. Here, let 'A_{ij}' be the performance code of 'i^{th}' recycling technology with 'j^{th}' factor. Refer to the earlier discussion. The factors are grouped into two categories such as beneficial and non-beneficial and the quantified values of beneficial and non-beneficial factors are normalized using Aij/Max(Aij) and Min(Aij)/Aij respectively to obtain normalized performance code. It is observed that the parameters, C, P, E, W (Highlighted in red) are found to be non-beneficial, while the parameters, R, and M are considered as beneficial parameters. This means that the larger value of beneficial parameter represents better recycling technology. Using the above expressions, the performance code of all factors are normalized and presented in Table 3. Subsequently, suitable weights are assigned to determine the performance codes of each factor considering their degree of importance, performance code of all parameters, i.e., performance code of i^{th} recycling technology, which is having j = 1,2,…,n parameters, is evaluated using the expression below;

$$A_i^{wpm} = \prod_{j=1}^{n} A_{ij}^{w_{ij}}$$

In this case, all the factors are assigned equal weights, i.e., w = 0.25), as all are equally important. The ranking of the calculated performance codes are done to help to select the best-performing battery recycling technology. Table 3 and 4 represent the normalized performance code, and performance score / ranking respectively.

Table 3: Normalized Performance code

Recycling technology	**Factors**					
	C	**P**	**E**	**W**	**R**	**M**
Pyrometallurgy	**0.55**	**1**	**0.55**	**0.43**	**1**	**1**
Hydro- Metallurgy	**1**	**0.34**	**1**	**0.6**	**1**	**1**
Direct recycling	**1**	**0.14**	**1**	**1**	**0.71**	**0.43**

Fig. 4 represents the performance code of various factors of all three recycling technologies.

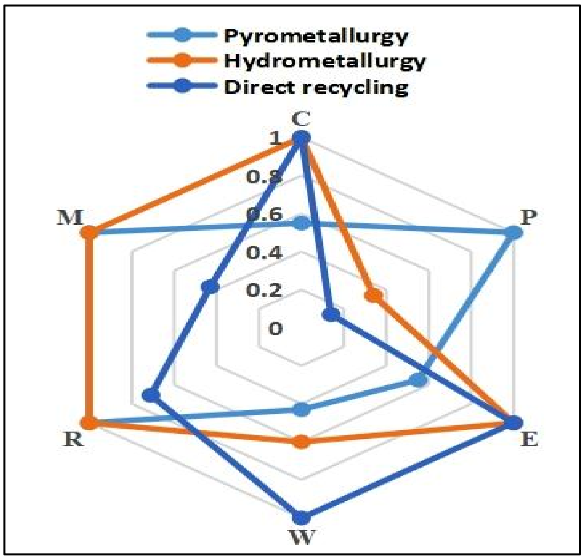

Fig. 4: Performance code of various factors

Table 4: Performance score and ranking

Recycling technology	**Pyrometallurgy**	**Hydrometallurgy**	**Direct recycling**
Performance score	**0.5990**	**0.6680**	**0.4545**
Ranking	**2**	**1**	**3**

From the above, it is clear that the technology, Hydrometallurgical metal reclamations is ranked as '1', while Pyrometallurgy and Direct recycling adjust ranking '2' and '3' respectively. The overall performance score of the various recycling technologies is represented in Fig. 5.

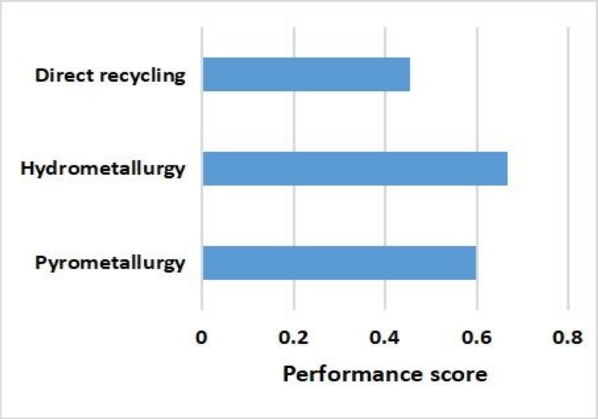

Fig. 5: Overall Performance score of recycling technologies

Though the Pyrometallurgy scores good in reliability and maintainability, yet it does not show better performance in costs, energy consumption and waste generation, while Hydrometallurgy scores better in all factors except production cost, and the Direct recycling consumes less energy with low capital cost and produces minimal wastes, but it has high production cost and poor maintainability.

4. Conclusion

Based on the findings, it is concluded that the method, Hydrometallurgy, is identified as the better option for LIB recycling as it performs better throughout the life cycle. The factors, which affect the life cycle performance of the recycling technology are considered in this selection method, which is based on WPM. The Hydrometallurgy method, which is selected to be the best one with overall performance score of **0.6680**, has not been implemented extensively due to its cost factor, however, it may gradually replace the Pyrometallurgy with overall performance score of **0.5990** which is currently employed by the most of the recyclers world-wide. The Direct recycling with a low score of **0.4545** needs more technological improvement before it becomes practically possible. However, this multi-criterion selection method has not considered material recovery in terms of quality and quantity. This aspects will be studied in future research work.

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