

Study on Recycling of Lithium-Ion Batteries: A future aspects of Sustainable Development

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ABSTRACT

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Technology must be created to enable a resource-efficient and financially viable recycling system for lithium-ion batteries in order to secure the availability of the component materials in the future. Planning for future recovery is more difficult because lithium-ion batteries are complicated goods whose designs and materials are still in development. There are several recycling methods under consideration or in use, and each has benefits and drawbacks. This study contrasts different procedures on a technical and financial level, highlighting variations in advantages as a result of cathode composition. There are proposed research fields because none of the current procedures are perfect and could lead to the improvement of recycling techniques. Separation technologies are among the most promising research fields.

Keywords: techniques, Separation, technologies

1 Introduction

1.1 Background

The supply of lithium was first the focus of worries regarding material restrictions on the manufacture of Li-ion batteries [1 – 3]. Even a very aggressive rollout of electric vehicles into the automotive sector was not projected to put a strain on lithium resources until the year 2050, according to a comprehensive analysis of the global production base and the physical availability of the resource [4].

Table 1: Projected cumulative world battery material demand to 2025 (1000 tons).

S. No.	Element	Project Demand		USGS Reserves
		If all NMC is low-Co (811)	If all NMC is high-Co (111)	
1	Lithium	230	230	16000
2	Cobalt	790	910	7100
3	Nickel	580	340	74000

Production facilities would need to expand at the same rate as the other newly necessary components. Short-term problems would arise if expansion did not keep up until balance was achieved, but a frenzy of interest prompted proposed capacity to vastly exceed probable demand. Cobalt, on the contrary hand, was recognised right away as a factor to be concerned about. Batteries might absorb

around 10% of the cobalt reserves now in existence, according to estimates of the total global demand for batteries merely (ignoring additional cobalt industries) by 2025. Table 1 estimates based on cathode estimates by Christophe Pillot [5].

The cost of cobalt, which as of this posting is roughly \$79,000/tonne, is already reflecting the effects of this significant demand [6]. The quick price increase and worries about child labour in the Democratic Republic of the Congo, in which the majority of cobalt is mined, have prompted cathode formulations for electric vehicle (EV) batteries to change to ones that rely more on nickel and less on cobalt. A kilogramme of high-cobalt cathode (LCO) contains \$55 value of lithium, cobalt, and nickel at the present price, but just \$17 worth of cobalt in a new lower-cobalt formulation (NMC811). Cobalt worries have led to a rise in interest in reprocessing as a potential supply of materials.

However, it is simple to demonstrate that recycling automobile Li-ion batteries for the purpose of supplying materials is a long-term tactic. Batteries should last approximately 10 years for mobility and a further 5–10 years for second-life uses like utility load management. As a result, significant quantities of batteries won't be ready for recycling for 10–20 years after they become widely available.

In the meantime, demand is predicted to continue growing quickly, requiring far more material than recycling could provide. Recycling can only provide a sizable portion of basic necessities once demand drops. However, recycling also has additional advantages, such as lowering the cost of disposal and other negative effects, and reducing the need for imported resources. The effectiveness of recycling through decreased energy use and emissions in the production of EV batteries has been shown through life-cycle analyses of battery production and recycling procedures [10].

1.2 Brief Li-Ion Battery Description

The battery pack for the Chevrolet Bolt car is depicted in Fig. 1. It has a very complicated construction and is made up of a lot of pouch cells that have been combined together into sections and then put into the huge pack housing. Each cell is wired up to electronics, and an electronic battery management system regulates the pack (BMS). Manufacturers, and even individual models within manufacturers, vary in the configuration, size, and design of the cells, module, and packs.



Fig. 1. Chevrolet Bolt battery pack.

Tesla uses cylindrical shape cells, which are comparable to those used in many smart appliances, but the majority of original equipment suppliers use pouch cells.

The interior parts of the cells may also differ in makeup. In example, any one of the numerous lithium transition metal oxides, such as lithium cobalt oxide (LCO), lithium nickel/manganese/cobalt oxide (NMC), or lithium iron phosphate, can be used as the cathode material (LFP).

To lessen their reliance on cobalt, many battery makers are switching to other nickel-rich compositions. $\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$ is represented by NMCs, which are typically notated as NMC_{abc} (e.g., NMC_{111} or NMC_{811}), where $x+y+z=1$ and $x=a/(a+b+c)$, $y=b/(a+b+c)$, and $z=c/(a+b+c)$.

2 Recycling Processes for Li-ion Batteries

Due to their complexity, Li-ion batteries can be recycled in a variety of ways. Fig. 2 illustrates the interconnections among process types and maps out some of these routes beginning only after pack has indeed been disassembled. Leftward pathways produce more valuable goods. There are three fundamental process types: direct recycling, hydrometallurgy (leaching), and pyrometallurgy (smelting) (physical processes). Depending on variables including the availability and properties of the raw material as well as the volume and cost of the substances that can be recovered, process elements can be integrated in a variety of ways.

High temperatures are used in pyrometallurgy to speed up the oxidation and reduction processes that turn transition metals like Co and Ni from oxides to metals so they may be recovered in mixed metal alloys. After being separated (via hydrometallurgy), the metals can be utilised to create fresh cathode material. In the smelter, other materials like aluminium, anode, and electrolyte are oxidised, providing the majority of the process energy. Lithium and aluminium oxides are lost in the slag and are rarely recovered. By using acids to dissolve the ions out of a solid, such as the cathode, hydrometallurgy creates a variety of ionic species in solution. These can be extracted using solvents or precipitation, then combined with other recovered components to create new cathode material.

Membrane separation is one of the additional methods that have been suggested [11]. By using physical processes, such as gravity separation, to recover separated materials without causing chemical changes, direct recycling separates the various parts of the black mass (active material powder from cell shredding) and makes it possible to recover cathode material that is reusable with little processing. (See [12], for instance. Due to its high value, recovering cobalt has historically been the principal goal of recycling Li-ion batteries.

The other things have been secondary. However, interest in recovering more materials has increased as cobalt concentration in batteries declines and obligatory recycling laws demanding recovery of more than 50% of the materials take effect in the European Union [13]. There is a significant opportunity to recover cathode material because materials account for more than half of the initial cell cost (Fig. 3). Cathode material is the main contributor to material cost.

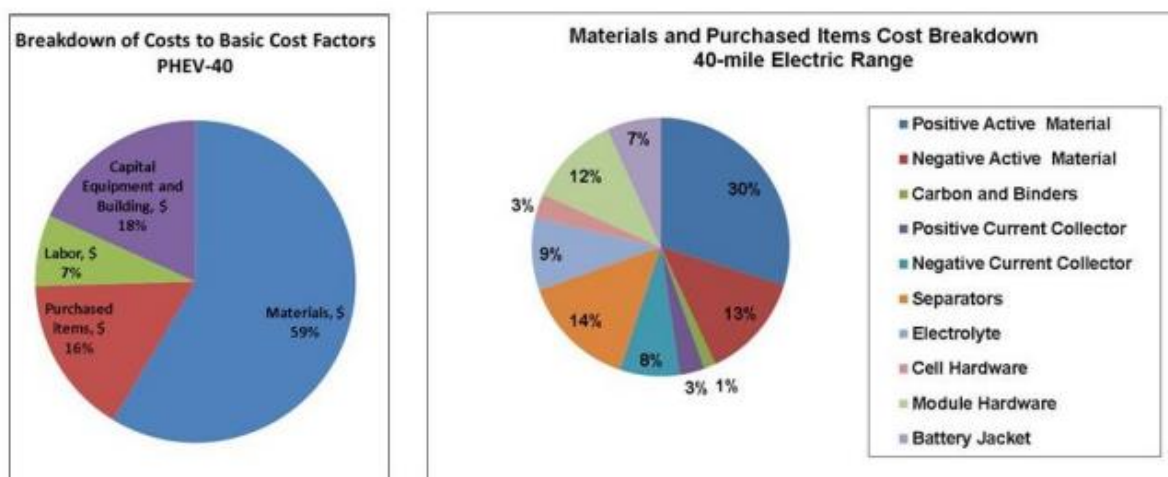


Fig. 3. Cost breakdowns for Li-ion batteries showing materials as part of total costs, (left), and components of material cost (right). Data from K. Gallagher [14].

Additionally, because the cost of cathode material exceeds the value of its component parts, recovering reusable cathode generates more income than recovering its component parts. Fig. 4 compares the cost of cathodes to the utility of recoverable elements or important precursors. About 70% of the cathode potential may be recovered from LCO cathodes by smelting or leaching, a percentage that dramatically decreases for other cathode chemistries with less cobalt.

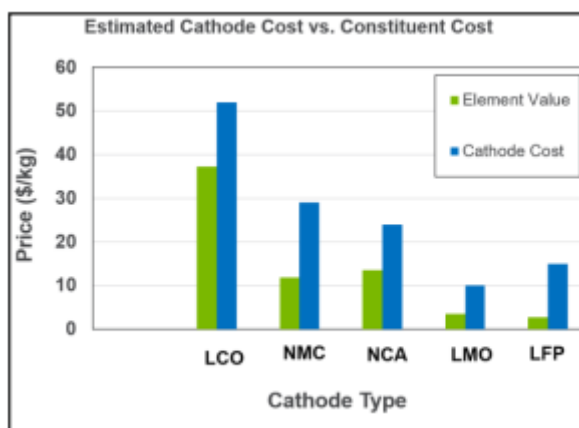


Fig. 4. Comparison of cathode cost to constituent element cost [10].

3. Recycling Process Comparison

Recovery of cathode material, notably LCO, is crucial from an energy consumption and greenhouse gas perspective because it is the second-largest energy and greenhouse gas contributor after recovery of aluminium [2]. The transition metals, which are typically made from sulphide ores, are the main cause of SO_x emissions, which recycling practically completely eliminates. Additionally, the estimated production implications of the cells can indeed be lessened the more materials that are recovered in a form that is as close to its ultimate, useable state as possible. Fig. 5 illustrates how using more recycled materials can lower the energy required to produce a cell. Recycling procedures that recover materials in forms more suited for direct usage in fresh cell manufacture make this possible.

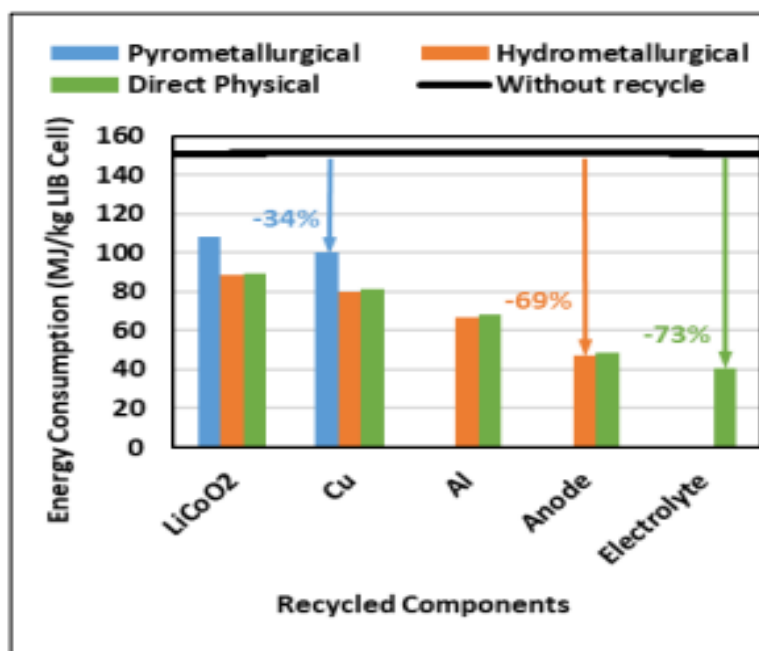


Fig. 5. Reduction in cell production energy by increasing use of recycled material [10].

The cells are disassembled or destroyed at the beginning of the hydrometallurgical and direct recycling processes, respectively. The copper and aluminium foils can now be recovered as metals right away, albeit they still need to be separated from one another. In contrast, pyrometallurgy feeds complete cells into a furnace and sends the aluminium and lithium to the slag while sending the copper to a mixed alloy product (where it is typically recovered by hydrometallurgy). Leaching might be used to recover these, however in most cases it is not practical due to the expense and energy required. Many of the

lithium is sent to the furnace dust when an electrical discharge furnace is used, where it may be easily collected [15]. Prior to smelting, the cells can be crushed (enabling extraction of the copper and aluminium foils), although doing so would not only incur additional costs but also eliminate a key source of smelter energy. The furnace uses less bought fuel since the aluminium is oxidised and acts as a reductant. Aluminum cans were reportedly used for fuel in one research [16].

Retriev, the only U.S. lithium recycler, shreds the cells but does not process the resulting black mass (a mixture of active components left after removing the foils); instead, the business offers that intermediate product for melting using transition-metal ores. Both hydrometallurgical processing and direct recycling, in contrast to pyrometallurgical processing, are low-temperature, low-energy procedures that don't require a huge scale and may thus be utilised locally or for household scrap, preventing the need to transport the material.

Direct recycling maintains the cathode crystal's shape, while hydrometallurgical procedures employ strong acid to dissolve the cathode into its individual ions, which are then introduced into an aqueous solution. Strong acids increase the cost and complexity of processes, which has led to various studies suggesting the application of organic acids [17][18]. The dissolved components can be segregated from one another and used again to create new cathode material after leaching with acid. Cobalt and nickel ions have highly similar features and are consequently challenging to separate from one another, necessitating multiple steps of solvent extraction.

The mixture is used in studies at Massachusetts Institute of Technology without separation, with enough virgin materials added to provide the relative proportions needed to produce the required remanufactured cathode formulation [19]. The solution's components typically also had some intrinsic value, making their recovery after leaching economically sensible for most cathode chemistries. However, the component value of some cathode materials, such as lithium manganese oxide (LMO) and lithium iron phosphate (LFP), is so low that hydrometallurgy is not profitable. Direct recovery of LFP could be cost-effective, and BYD Co. representatives claimed that this was achieved in China, but they were unable to elaborate [20]. In the literature, direct regeneration of LFP has been described [21].

The benefit of hydro metallurgically disassembling the structure is that even the outputs are universal products that can be utilised as inputs to create a range of new products because they are not unique to a particular cathode structure. However, it is believed that direct recycling's outputs will still have a distinct, well-defined structure. Although the conservation of shape can be viewed as a benefit, it does present a limitation because inputs to direct recycling must have been separated by cathode type or the result will be a mixture with significantly lower value (unless cathode mixtures are proven to be advantageous). Although a magnetic separation technique has been patented [22], no efficient and cost-effective cathode material separation technology has been demonstrated for usage before or after recycling.

As a result, the majority of companies that enter the recycling market today provide variations on hydrometallurgical methods. Because of the retention of structure, the formulation that was recovered at the conclusion of a vehicle's life and after a battery's second life is probably around 15 years old and could have been replaced with more recent materials. Although the restoration of cathode material or metal foils has received the majority of the attention so far, low-temperature techniques can also be used to recover anode and even electrolyte.

Of course, both are consumed in a smelter to provide some process fuel. In a smelter, separators are also burned. Separators' usefulness resides in their specific form factor (thin porous film), and they would be destroyed in any processing, hence no method of recovering separators has been proposed. Although the raw materials for polymers may be reclaimed, they have little worth.

It has been shown that anode material may be recovered using straightforward physical procedures as part of direct recycling; although anode material is less valuable than cathode, it must be separated in order to obtain usable cathode material. There are a variety of techniques for separating the black mass into the anode and cathode fractions, including froth flotation and gravity separation with dense liquids [23][24].

Hydrometallurgical procedures could also be used to recover anode material. Graphite would stay a solid as the cathode degrades since acids do not dissolve it. Filtration might then be used to recover the material, but now there is a problem with the quality. It is possible to see planar flaws and particle separation. The cell that contained the ancient particle was extracted had a 33% capacity decline, but some performance might be recovered with therapy.

It has been proven that electrolyte recovery, including the recovery of lithium salts, is possible when breached cells are extracted using supercritical CO₂ [25]. Even while the recovered electrolyte worked well in regenerated cells, the procedure is not thought to be economically viable. The possibility of solvent extraction has limited research into electrolyte restoration due to its poor value and anticipated contamination by oxidation products. When significant amounts of material are really being processed, interest might rise.

4 Research to Enable Li-ion Battery Recycling

Li-ion battery recycling research aims to recover as many materials as possible, in as usable a condition as possible, and in a way that is both sensible for the environment and practical economically. Inadequacies of current procedures have been emphasised throughout this study; we now group them together as areas where research could advance practice. There are various types of research fields. The first is separation processes, which are notable.

4.1 Separation Methods

The necessity for material segregation affects every step of the recycling process, from product recovery on through collection. Separating different battery kinds from one another is the first step in order to send them for the proper procedure. If Ni-MH and Li-ion batteries are differentiated, along with LFP batteries, even smelting results in a higher-value output. Large packs probably only need labels for identification, which either humans or machines can read. Both manual and automatic dismantling of the packs and modules down to the nano scale is an option. Depending on the recycling procedure to be employed, further separating individual cells by chemical and form factor may be necessary when processing a mixture of them.

The design of the apparatus to sort and process these would be substantially simpler if they were consolidated to only a few varieties. Here's one illustration of how product designers could incorporate recycling (DFR). Other DFR recommendations include employing reversible connection techniques (such as nuts and bolts in place of welding) to enable pack disassembly. Similarly, DFR would require employing an adhesive substance that might be easily removed, maybe with a common solvent, if cells had to adhere to the module. Even if this way of thinking isn't strictly based on scientific research, its application would nonetheless aid promote economic recycling. Additional separation procedures are necessary to extract materials from the cells for all but the simplest pyrometallurgical processing.

Although manual disassembly of individual cells is sometimes done for experimental purposes, processing small cells industrially would be prohibitively expensive. Although it would be difficult due to the wide range of cell designs, robotic disassembly is still a possibility. Therefore, before materials may be recovered, the majority of processing plans for small Li-ion cells propose shredding. Separating as many different components as you can is the next step after shredding. Typically, screening is used to separate the electrode active elements from the aluminium and copper foils containing them. Part process schemes now add a step before or after screen to drive off volatile organics because some of the active ingredient may still adhere to the foils. In addition to evaporating the electrolyte, this procedure eliminates the binder and minimises the amount of active material adhering to the foils. At this point, the electrolyte may be recovered, but processing would be necessary to get rid of the breakdown by-products. To meet the objectives of the EU Battery Directive's material recovery, electrolyte recovery might be required [15]. Eventually, the copper and aluminium must also be separated from one another, a task made challenging by the enormous surface area of the tiny, asymmetrical particles. If one desires to recover anode or cathode materials for reuse, little aluminium and copper foil fragments might well be compressed in the black mass that is left after filtering out the foils.

This is a far more serious hazard. Ionic components need to be separated from one another once they are in solution for the hydrometallurgical approach. Precipitation, solvent extraction, electrochemical processing, and membrane processes are a few potential techniques. There have also been suggested biological solutions. At this point, recovering anode material might also be taken into consideration. The smelted mixed alloy result also goes through leaching and needs to be separated. Hydrometallurgy reagent and process parameters can be optimised to increase selectivity and yields while decreasing overall costs.

The cathode and anode must be physically separated from one another for direct recycling using procedures that don't alter the particle morphology. Both froth flotation and heavy liquid separation have been used and are protected by patents. It is necessary to ascertain the most effective techniques for separating cathode and anode from one another and for eradicating all traces of foils from them. The discussion in this article assumes a single cathode formulation, however the shredded cells may have actually been part of a mixed batch.

In that instance, to acquire the best possible product value, the cathode materials must be separated from one another. Even though magnetic separation is patentable, it may not be practical, and it is not known whether identical formulations (such as two distinct NMCs) can be separate from one another. Direct recovery of high-value cathode products might become possible with techniques for isolating cathodes from one another or for utilising a mixed product. Three different process types have been discussed thus far. But between direct recycling and hydrometallurgy, wherever cathode is handled under increasingly harsh conditions, there is essentially a continuum of potential processes that may be studied in order to fully understand the underlying mechanisms in degrading the cathode. It is permissible to use solutions other than acids and bases.

This might result in the creation of a hybrid recycling approach that eliminates impurities and flaws in the crystalline structure without compromising its integrity. Creating recycling techniques that might turn mixed input directly into high-value outputs, such cathode material, without destroying the structure seems to be another method for dealing with the range of materials. One would wonder, for example, if a combination of NMC electrochemical devices with varying proportions of nickel, manganese, and cobalt can be treated with more of these elements to make the mixture into a product with the requisite uniform composition.

4.2 Process Design

Other research concepts are not easily categorised. Process optimization is a possibility for all of the processes mentioned. The effects of pre-treatment options, reagents, reaction periods and temperatures, unit process order, time duration (or throughput, if run continuously), and batch size on specific products and process economics will all vary depending on the battery chemistry. Utilizing varying feedstock would be possible with flexible process design. It is crucial to confirm that all hazardous substances, even in minute amounts, have been tracked before expanding any procedure. Fluorine in electrolyte salts and binder in particular needs to be taken into consideration. Even if workable recycling procedures are created, a market for the product is still necessary. Therefore, it is necessary to guarantee the dependability of recycled materials.

4.3 Modeling of Recycling Processes

Process modelling allows for the estimation of process behaviour and costs without the need to construct a plant, which saves both time and money when developing new facilities. To offer financial and environmental implications throughout all stages and processing processes of the life cycle of a Li-ion battery, Argonne is developing a high-level, closed-loop battery process model dubbed ReCell. With the use of this tool, stakeholders will be able to simulate and analyse the relative costs and environmental effects of various battery recycling pathways. It may also make DFR analyses easier. When specific process information is entered, the model will give a high-level preview of the effects of suggested process and chemical adjustments. The tool's purpose is to assist in the improvement of battery recycling procedures and ensure the recovery of vital materials.

5 Conclusions

Li-ion battery recycling is not an easy process. These are a wide range of intricate products whose design is always changing. Although recycling them would improve the environment and, one would think, the economy, none of the available techniques is perfect; each has advantages and disadvantages of its own. We have outlined some potential research directions in this study that could result in improved recycling procedures so that, if and when a significant number of electric vehicles are produced, we will be able to dispose of their batteries properly. Material separation technology appears to be the most productive research field at various sizes. Another key development that could enhance the prospects for recycling is design for recycling.

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