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Cost/Benefit Analysis of Adding A Pre-Treatment Step in the Lithium Ion Battery Recycling Process

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Introduction

A large quantity of batteries are used and disposed of due to a variety of portable devices such as cell phones, laptops, power tools, etc. [1]. A transition to an electrified transportation system will significantly increase the number of used batteries being disposed. This transition is already underway as can be seen from the development of hybrid electric vehicles (HEVs), plug-in hybrid electric vehicles (PHEVs), and purely electric vehicles (EVs). Rechargeable lithium-ion batteries (LIBs) are more prevalent in consumer electronics than nickel cadmium (NiCd) or nickel metal hydride (NiMH) batteries. [2] [3]. This is due to the higher energy density, lack of memory effect, small size, and light weight of LIBs [4].

LIBs accounted for $11.8 billion USD in sales in 2012 because of their high energy density, long lifespan, and light weight. This was 60% of the total portable battery sales and 37% of total battery sales [5]. The advantages of LIBs make them the ideal candidate for use in HEVs, PHEVs, and EVs. For HEVs sales were 1.5 billion USD as of 2011 and are expected to reach about 3.6 billion USD by 2015 [6]. The increased use of LIBs in the automotive industry can be attributed to the fact that CO_2 emission standards are getting stricter and the industry must keep up with the strict standards [7].

With such an increased use of LIBs, large quantities of LIB solid wastes are being generated every year.

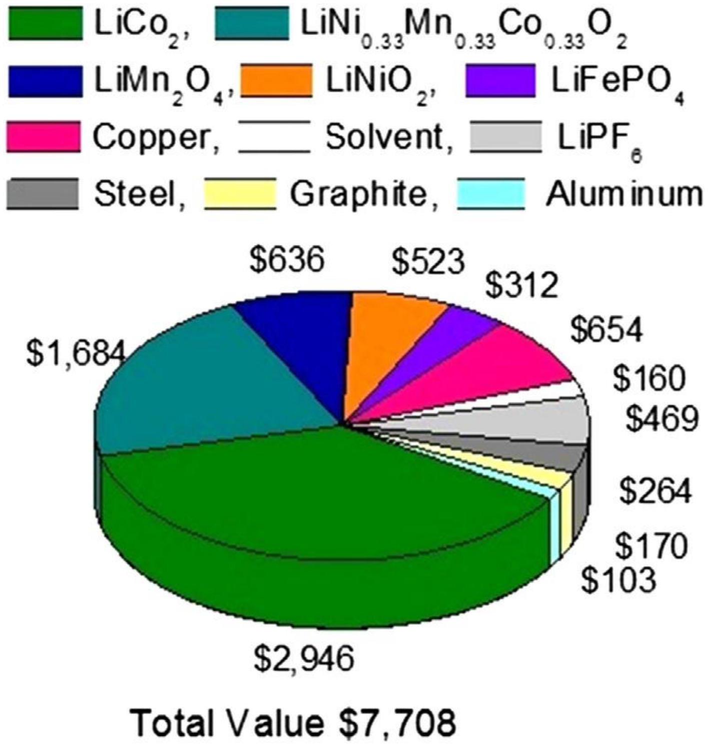

Figure 1: Cost breakdown of each component in 1 ton of Li-ion batteries [13]

LIBs used in consumer products have a lifespan of less than 3 years and those used in the automotive industry are projected to have a lifespan of roughly 10 years [8]. Spent LIBs will not only pollute the environment if they are not disposed of properly but also valuable resources will be wasted if they are not recycled and/or reused properly. This makes recycling valuable metals in spent LIBs imperative [5]. A study by Roland Berger Strategy Consultants shows that the global automotive LIB market is expected to reach over 9 billion USD by 2015 [9].

In HEVs, PHEVs, and EVs the battery shelf life and calendar life are very different since in many cases the battery reaches the end of its calendar life before reaching the end of its cycle life. Since batteries are not used until the end of their cycle life they may still be reused for stationary purposes [10]. The LIBs can then be recycled after reuse. The value for LIBs in such a case would be very similar to Pb-acid batteries. Adding this extra phase could improve the lifecycle economics of LIBs [11].

Recycling makes economic sense only if the revenues from recovered materials plus the avoided disposal costs are greater than the costs for collection and processing. R&D projects in battery recycling aim to develop processes that are feasible and economically viable by themselves.

Traditional Lithium ion battery recycling process.

$LiCoO_2$ is the most frequently used cathode material used in LIBs because it has a high energy density, high operating voltage, and a good cycling performance [2]. The drawbacks of using this cathode are: high cost, limited cobalt reserves, and toxicity [6]. From an environmental and resource conservation perspective it makes recycling of cobalt, nickel, and lithium from spent LIBs important. If these materials are recycled and returned to new battery production, it has the potential to reduce the battery's life cycle impact by about 51%, when natural resource consumption from using only primary materials is considered. Recycling is important also because of the leaching potential of hazardous materials contained in LIBs during landfill disposals [12]. From an economic perspective it is a good opportunity to recover valuable materials used in LIB production, namely cobalt. Figure 1 shows the value of each component in a 1 ton of LIB. It is seen that the cathode material is the most expensive component of the LIB. The cathode material is constantly evolving to improve energy density and reduce cost of the LIB [13]. Since cobalt is an expensive metal, manufacturers are moving towards low cost cathode materials to reduce manufacturing cost [14]. This reduces economic incentives to recycle LIBs with cheap cathode material at their end of life.

Typical recycling processes include a combination of [15]:

1. Crushing, acid leaching, heat treatment, chemical precipitation, and a combination of mechanical, thermal, hydrometallurgical, and sol-gel steps.
2. Dismantling, acid leaching, chemical precipitation, solvent extraction, and a combination of mechanical dismantling and separation, electrochemical and thermal treatment
3. Dismantling, chemical deposition, and solvent extraction
4. Leaching, solvent extraction, electrode dissolution, and cobalt electrochemical reduction
5. Dissolution, heat treatment, acid leaching, and chemical precipitation
6. Crushing, ultrasonic washing, acid leaching, and chemical precipitation

The process under consideration in this paper is number 6. This process involves a number of pretreatment steps for recycling of spent LIBs. To prevent short circuit and self-ignition, LIBs are first discharged by placing in a salt solution. They are then frozen and either dismantled and/or crushed before being processed further.

We propose a recycling process consisting of the following steps:

1. Discharge the LIB
2. Freeze the LIB with liquid nitrogen and crush the discharged LIB in a 180/180 Econogrind granulator
3. Using rare earth magnets, remove magnetic impurities
4. Perform a density based separation of large materials (plastic/copper)
5. Use sieving to separate into different size fractions
6. Perform leaching on the fine fraction and middle fraction (rich in cobalt and gold)
7. Chemical precipitation to recover the high value metals

The process aims to increase the value of metals recovered, lower the operating costs, as well as lower the emissions and wastes to landfill.

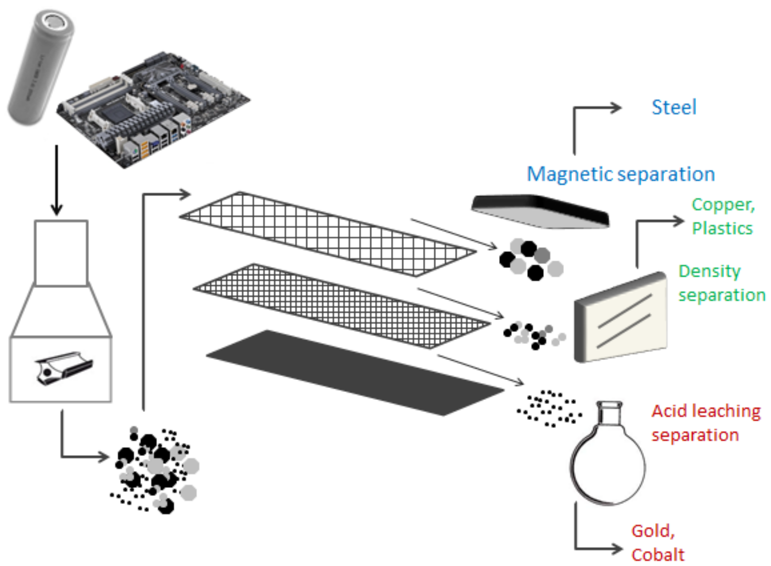

Figure 2: Conceptual schematic of the proposed system

The objective of this report is to verify the claims that have been made about the process and compare it to a traditional LIB recycling processes to see whether there are economic benefits of adding the extra pre-processing steps.

Methodology

For a robust end-of-life battery infrastructure, a better understanding is needed about how to maximize profitability whilst mitigating uncertainties associated with a widely variable waste stream. The optimization model developed by Wang *et al.* will be used to determine the profitability of recycling LIBs based on their cathode chemistry. This model gives an insight about the economies of scale for LIB recycling relative to different cathode chemistries [16]. For the sake of simplicity, this paper will assume all the LIBs in the recycling stream to have a cathode chemistry based only on $LiCoO_2$.

The optimization model that was developed, identifies the minimum amount of spent LIBs for a recycling facility to be profitable based on the total revenue and operating costs. This assumes that all the metallic materials contained in the LIBs are recovered and recycled. [16]

First, the results of the model will be used to determine the minimum amount of LIBs required for a recycling facility to be profitable based on costs and revenues. The revenue stream will come from selling off the materials recovered at the prices specified in Table 1. The costs fall into two major categories which are variable and fixed costs. Variable costs are classified as the costs that scale proportionally with the volume of the outputs [17]. Fixed costs are the costs which are independent of the volume of the outputs. They include salaries, rents, etc. [18].

The model assumes that all the metallic materials contained in the LIBs will be recovered at recycling efficiencies determined from literature. Recycling efficiencies are scaled up from lab scale studies and are not indicative of real world efficiencies. The costs include the variable cost and the annual fixed costs. The minimum amount of LIBs for a recycling facility to be profitable can be identified by calculating the break-even point through the model.

Base Materials	**Prices ($/kg material)**	**Composition (kg/ton LIBs)**	**Recycling Efficiency %**
Cobalt	46.30	173	89
Nickel	21.71	12	62
Lithium	62.26	20	80
Iron/Steel	0.05	165	52
Aluminum	2.25	52	42
Copper	7.54	73	90

Table 1: Metal prices and recycling efficiencies for $LiCoO_2$ based batteries [16]

Using the pre-processing steps outlined in this paper we expect the recycling efficiencies of the materials outlined in Table 1 to increase thereby increasing the revenue stream, and reducing the amount of LIBs needed to reach the break-even point for recycling facilities. Another advantage of using the pre-processing step would be a reduction in variable costs. Since the crushed LIBs are separated into different size fractions, different metal compositions get enriched in different size fractions. This would enable a material specific recovery process which would imply less material being processed to obtain similar results. Processing a lower amount of material would decrease the amount of raw material (acid) needed to carry out the recycling process which would in-turn reduce the variable cost (material and disposal cost).

According to Wang *et al.* improving the efficiency of recycling of Cobalt from the $LiCoO_2$ chemistry can increase the revenue by up to 9%. Table 2 summarizes the findings of the paper.

Base Material	**Improved Recycling efficiency %**	**% increase in revenue**
Cobalt	99%	9%
Nickel	72%	<1%
Lithium	90%	1%
Iron/Steel	62%	<<1%
Aluminum	52%	<1%
Copper	100%	<1%

Table 2: Increase in unit revenue through higher recycling efficiency [16]

It can be seen that ignoring operating and raw material cost but increasing recycling efficiency an increase in revenue of 10% can be achieved. The pre-processing step that was proposed in the paper will be tested at lab scale to determine the increase in recycling efficiency and reduction in operation and raw material cost. These results will then be scaled to the level specified in the model to determine the percentage increase in revenue. The scaling factors will be determined from literature and assumptions regarding the operating and maintenance cost of the pre-processing steps.

The base case assumed in this scenario consists of only $LiCoO_2$ batteries since they currently dominate the battery market for consumer electronics. They are the most commonly used in electronics. Different manufacturers use different cathode chemistries thus the material composition in LIBs would vary in their bill of materials (BOM). The BOM from several manufacturers (Panasonic, Sony, AT&T, etc.) were studied and an average material composition was calculated and used as the BOM for the base case LIBs. Table 3 summarizes the BOM that was calculated and used for the base case. The potential value of materials that can be recovered from spent LIBs was calculated using yearly average commodity metals prices from United States Geological Survey (USGS) for 2012. For the base case, from literature, the fixed cost was assumed to be $1,000,000/year, the variable cost was assumed to be $2,800/ton, and the maximum recycling capacity was assumed to be 34,000 tons/year. The initial investment was considered to be $25,000,000.

Material	**Grams**	**Weight %**
Aluminum	2.4	5.2
Cobalt	8	17.3
Copper	3.4	7.3
Lithium	0.9	2
Nickel	0.6	1.2
Steel/Iron	7.6	16.5
Graphite	10.6	23.1
Carbon black	2.8	6
$LiPF_6$	1.7	3.7
Other	4.7	10.3
Binder	1.1	2.4
Plastic	2.2	4.8

Table 3: BOM for a typical LIB based on $LiCoO_2$ [16]

Intuitively, the higher the maximum capacity, the higher the fixed costs; however, this relationship is likely not linear, usually being shown by Equation 1:

$$\frac{I_2}{I_1} = \left(\frac{Q_2}{Q_1}\right)^x$$

Where I_1 refers to the known investment for capacity Q_1; I_2 refers to the investment desired for capacity Q_1; and x is the investment capacity factor. The value of this investment-capacity factor is empirically

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derived, usually falls between 0 and 1, and varies depending on the type of industry or products. Since LIB recycling is still in its infancy, there is not enough data to make a meaningful estimation of this factor from a statistical perspective. Therefore, the six-tenths factor rule ("0.6 rule") has been assumed. The "0.6 rule," which says the capital investment typically increases along with production capacity to the power of 0.6, was adduced initially based on the relationship between individual equipment and their capacities and has been extended to complete e-waste recycling plants [19]. Based on literature review a fixed cost of $1,000,000/year was selected.

Results and discussion

1. Base Case

According to the parameters detailed above, an input stream of at least 170 tons per year of spent LIBs is required for the base case facility to cover all associated costs. This breakeven amount is the minimum steady-state supply required; higher volumes would result in a net profit. Achieving this target will be determined by a number of factors, including service area, population density, usage patterns, available transportation infrastructure, potential capacity, and collection rate.

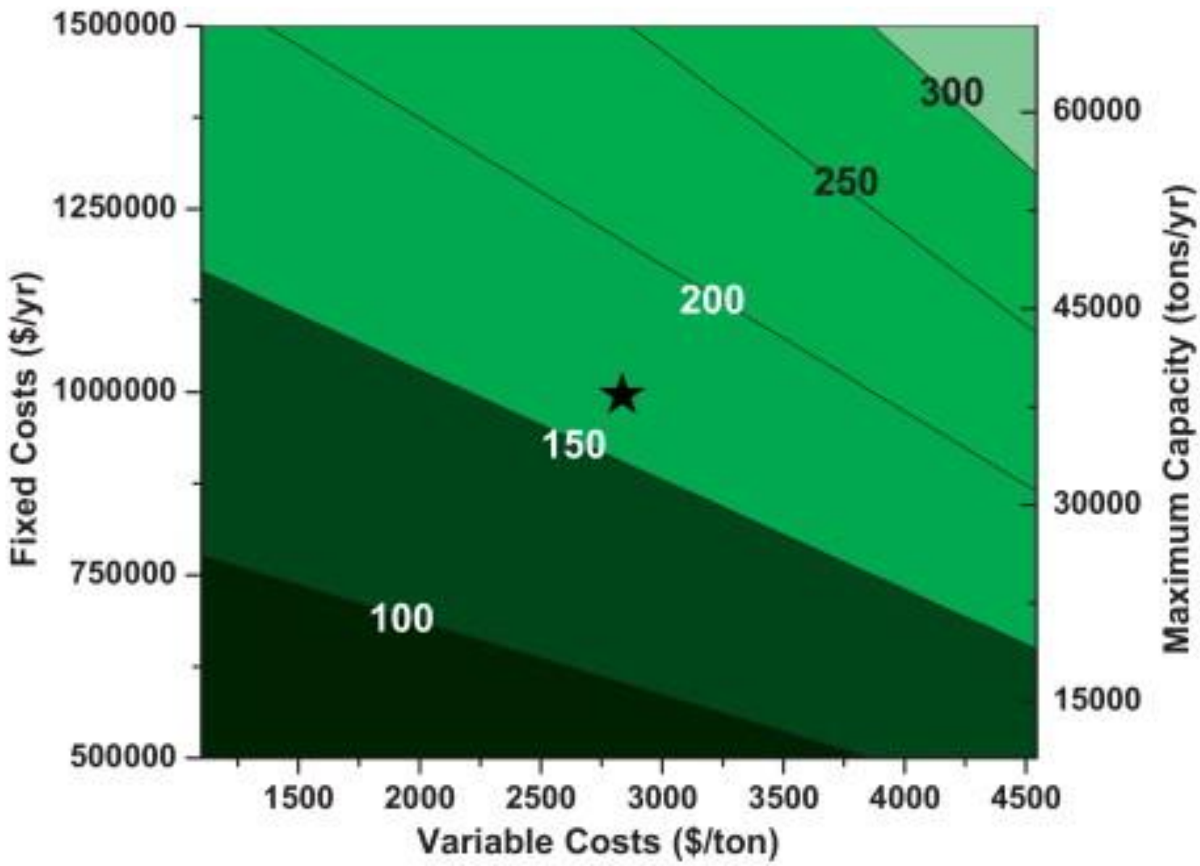

Figure 3: Minimum volumes of spent LIBs for a recycling facility to cover all expenses for different fixed and variable costs. Star refers to the base case. Darker color refers to lower break-even levels; lighter color refers to higher levels.

According to the statistics in their annual report, approximately 10 million pounds (4500 tons) of rechargeable batteries were collected in 2012. Assuming 20% of this stream is made up of LIBs [20], approximately 2 million pounds (910 tons) of spent LIBs were collected by Call2Recycle® in 2012. The calculated economic break-even point in the base case (170 tons per year) is about one quarter of the total spent LIBs collected by Call2Recycle; this equates to four LIB recycling facilities like the one assumed in the base case in viable operation.

Base Materials	Prices ($/kg material)	Composition (kg/ton LIBs)	Recycling Efficiency %
Cobalt	46.30	173	89
Nickel	21.71	12	62
Lithium	62.26	20	80
Iron/Steel	0.05	165	52
Aluminum	2.25	52	42
Copper	7.54	73	90
Total revenue per ton = $8900			

Table 4: Total revenue per ton for LIBs based on $LiCoO_2$

	Base Case
Total Investment	$25,000,000
Fixed Cost ($/per year)	$1,000,000
Variable Cost ($/ton)	$2,800
Maximum Capacity (ton/year)	34,000
Break Even Tonnage (ton/year)	170

Table 5: Base case results

2. Proposed Case

It can be seen from Figure 3 that a reduction in the variable cost can reduce the break-even tonnage required to make the recycling facility profitable. Our proposed recycling process aims to do just that. The three additions made to the original recycling process include:

- **Magnetic Separation**

Magnetic separation can effectively get rid of all the magnetic impurities in the shredded LIBs before processing. Using electromagnets on a conveyer belt, impurities like Iron and Nickel can be successfully removed from the shredded batteries. From Table 3 it can be seen that magnetic separation would remove about 17.7% of the total shredded LIB waste stream thereby reducing the amount of material that needs to be processed further.

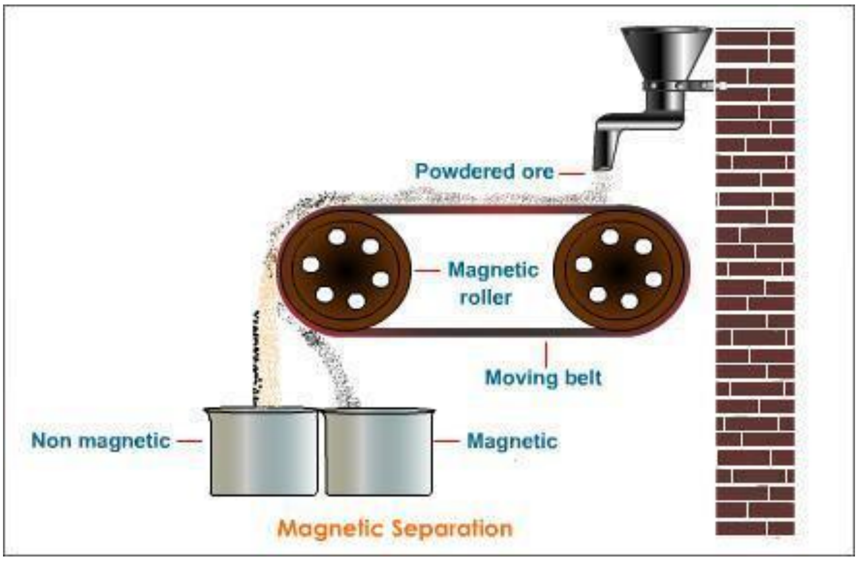

Figure 4: Schematic of Magnetic Separation

- **Density based separation**

Density based separation would ensure that metals are separated from plastics and other lightweight impurities that do not have any significant resale value. These plastics that have been recovered can be used as an input source for a waste-to-energy plant but it is beyond the scope of this project to look into it further. A typical ionic solution of $NaAlO_2$ can be used to accomplish this. $NaAlO_2$ has a specific gravity of about 1.5 g/cm^3 which makes it the ideal candidate to differentiate between plastics that have a lower specific gravity and metals having a much higher specific gravity. This step would remove an additional 5% of the shredded LIB waste stream thereby reducing the material to be processed even further.

- **Sieving**

Shredding and sorting has widely been used in other products' recycling processes to increase the surface area, liberate the component materials, achieve material segregation, and improve the efficiency of subsequent recycling processes, all at relatively low cost and environmental impact [21].

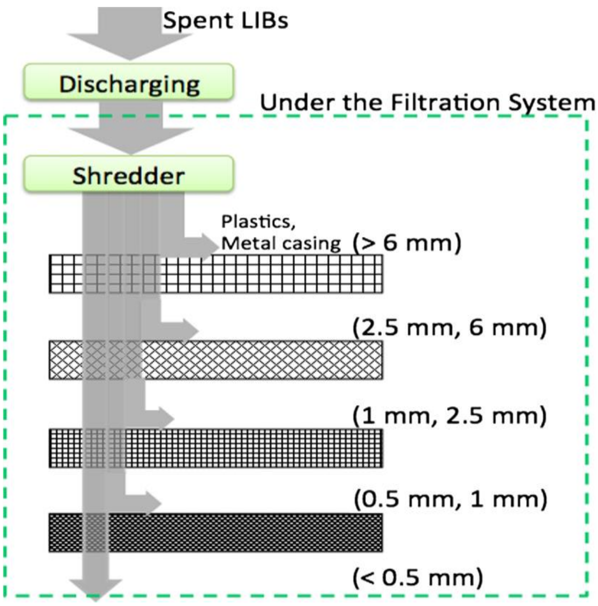

Figure 5: Schematic of sieving process [22]

Experiments carried out by Wang *et al.* demonstrate that the pre-recycling process enabled comparisons of the separated size fractions on the basis of metal content, economic value of that content, and variability across multiple battery chemistries and manufacturers. The sorted fractions of shredded cells showed clear visible differentiation, particularly in the accumulation of poorly-shredded battery housing material in the largest size fraction. The larger pieces (>6 mm) are mostly battery casings and plastic separators. Copper pieces can be visibly detected in the coarse (2.5–6 mm) and mid (1–2.5 mm) fraction. Fine black powder, likely comprised of graphite from the anode and the active materials from the cathode, dominates the ultrafine (<0.5 mm) fraction. The X-ray fluorescence (XRF) analysis of different size fractions are shown in Figure 6. The results shown are very positive since we can now successfully target high value metals in different size fractions. It is important to note that these separations do not include magnetic separation and density separation. If magnetic and density separation are carried out before the sorting we can expect the size fractions to be free of Nickel and Iron. This would completely eliminate the >6mm size fraction.

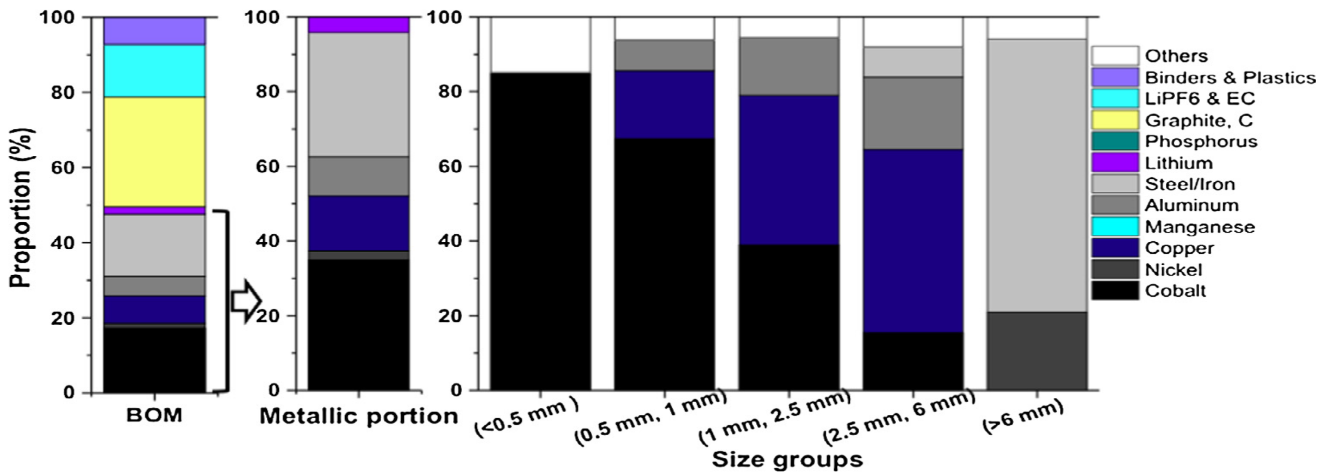

Figure 6: a) BOM for $LiCoO_2$ batteries. b) Metallic portion of the LIBs c) Metals in different size fractions [22]

Since we have assumed that all the revenue comes from selling the recovered metals, sieving and sorting the shredded LIBs actually helps reduce the amount of material that needs to be processed. This could potentially help reduce the variable cost associated with processing of the LIBs. Initially it was assumed that the variable cost of recycling per ton of LIB was $2,800. Adding these pre-processing steps, reduces the amount of material needed to be processes by half. This does not mean that the variable cost also reduces by half. Using Equation 1 we calculate the new variable cost to be around $1,867/ton. According to the U.S. Department of Energy the additional cost of adding sieving, density separation, and magnetic separation equipment according to the sizes that we need would be around $5,000,000. This would bring the initial investment cost up from $25,000,000 to $30,000,000.

Adding these pre-processing steps will not only reduce the variable cost but also increase recycling efficiency since now specific metals can be targeted in specific size fractions [22]. The increase in revenue that can be achieved by the increased recycling efficiencies is shown in Table 6. Based on this increase in revenue the break-even tonnage was calculated. Two cases were created; the first one assumes that the variable cost reduces to $1,867/ton and the revenue increased to $9,900/ton (best case); the second scenario assumes that the variable cost reduces to $1,867/ton but there is no increase in revenue i.e. it stays at $8,900/ton (worst case). These results are represented in Table 7.

Base Material	Improved Recycling efficiency %	% increase in revenue	$/ton increase
Cobalt	99%	9%	$800
Nickel	72%	<1%	$30
Lithium	90%	1%	$130
Iron/Steel	62%	<<1%	$0
Aluminum	52%	<1%	$10
Copper	100%	<1%	$50
Total Revenue per ton = $9900			

Table 6: Increase in revenue due to higher recycling efficiencies [22]

	Base Case	Proposed Case (best)	Proposed Case (worst)
Total Investment	$25,000,000	$30,000,000	$30,000,000
Fixed Cost ($/per year)	$1,000,000	$1,000,000	$1,000,000
Variable Cost ($/ton)	$2,800	$1,867	$1,867
Maximum Capacity (ton/year)	34,000	34,000	34,000
Break Even Tonnage (ton/year)	164	125	143
Revenue ($/ton)	$8900	$9900	$8900

Table 7: Comparison of base case to proposed case

Table 7 shows that even when there is no increase in recycling efficiency, the proposed case reduces the break-even tonnage. When there is an increase in revenue the break-even tonnage reduces even further. The increase in investment can be justified due to this reduction in break-even tonnage. This result is beneficial as there can now be more facilities to recycle batteries in locations close to collection centers which would further reduce the cost of transportation of the end of life LIBs.

Richa *et al.* developed a future oriented material flow analysis (MFA) used to estimate the volume of LIB wastes to be potentially generated in the United States due to EV deployment in the near and long term future. They concluded that when considering the range from the most conservative to most extreme estimates, a cumulative outflow between 0.33 million metric tons and 4 million metric tons of lithium-ion cells could be generated between 2015 and 2040. By 2040, projected annual waste flows can reach up to 340,000 tons/year [23]. Assuming that 10% of these waste flows arise from LIBs only based on the $LiCoO_2$ cathode chemistry, we can calculate the projected cash flows in the year 2040 for the base case and our proposed cases.

For Year 2040	Base Case	Proposed Case (best case scenario)	Proposed Case (worst case scenario)
Tonnage (ton/year)	34,000	34,000	34,000
Cost of processing ($/year)	$96,200,000	$64,466,667	$64,466,667
Revenue obtained ($/year)	$302,600,000	$336,600,000	$302,600,000
Profit ($/year)	$206,400,000	$272,133,333	$238,133,333

Table 8: Projections for year 2040

From Table 8 it is evident that our proposed case is better than the base case in terms of profit that can be

obtained when the plant is operated at capacity. Thus, we can conclude that there is enough economic incentive to implement the proposed recycling system over the traditional process.
As battery recyclers are strongly driven by economic incentives, this paper takes the potential recoverable value of battery materials into consideration, resulting in a more clear recovery priority. The results suggest that pre-sorting by cathode type has the potential to further improve the segregation efficiency of battery waste streams. While this might be expensive to perform in certain locales, emerging economies might be an ideal place for labor intensive pre-processing [16]. Particularly, while lithium cobalt oxide is the most common cathode type at present, cobalt is not even contained in several projected next-generation LIBs (e.g., lithium iron phosphate, lithium manganese spinel, and lithium polymer). If LIB recycling is carried out without any presorting by cathode chemistry, a significant uncertainty would be involved, and recycling yields will likely be diminished. This is demonstrated by a simple calculation involving $LiFePO_4$ and $LiMn_2O_4$ which are the two most commonly used cathode chemistries apart from $LiCoO_2$.

Base Materials	Prices ($/kg material)	Composition (kg/ton LIBs)	Recycling Efficiency %
Cobalt	46.3	0	89
Nickel	21.71	0	62
Lithium	62.26	12	80
Iron/Steel	0.05	432	52
Aluminum	2.25	65	42
Copper	7.54	82	90
Manganese	0.01	0	53
Revenue for $LiFePO_4$ = $1230/ton			

Table 9: Revenue from $LiFePO_4$ BOM

Base Materials	Prices ($/kg material)	Composition (kg/ton LIBs)	Recycling Efficiency %
Cobalt	46.3	0	89
Nickel	21.71	0	62
Lithium	62.26	15	80
Iron/Steel	0.05	164	52
Aluminum	2.25	11	42
Copper	7.54	11	90
Manganese	0.01	204	53
Revenue for $LiMn_2O_4$ = $840/ton			

Table 10: Revenue from $LiMn_2O_4$

Assuming the proposed worst case is applied to a stream consisting of LIB cathode chemistry only based on $LiFePO_4$ and $LiMn_2O_4$. Profit for year 2040 only based on waste flows from EV waste is calculated and presented in Table 11.
Table 11 clearly shows that if the LIB waste stream is only made up of either $LiFePO_4$ or $LiMn_2O_4$ there will be no chance to make a profit. Wang *et al.* performed a similar analysis and determined that if the $LiCoO_2$ content of the LIB waste stream drops below 21% then it will not be economically viable to recycle end of life LIBs anymore.

Proposed case (worst)	$LiCoO_2$	$LiFePO_4$	$LiMn_2O_4$
Total Investment	$30,000,000	$30,000,000	$30,000,000
Fixed Cost ($/per year)	$1,000,000	$1,000,000	$1,000,000
Variable Cost ($/ton)	$1,867	$1,867	$1,867
Maximum Capacity (ton/year)	34,000	34,000	34,000
Break Even Tonnage (ton/year)	143	∞	∞
Revenue ($/ton)	$8900	$1230	$840
Year 2040 tonnage of end of life battery packs just from EVs = 34,000 tons/year			
Profit for year 2040	$238,133,333	- $22,646,667	- $35,906,667

Table 11: Profit calculations for alternate cathode chemistries

Conclusions

An efficient collection and recycling infrastructure must be put into place to minimize environmental impacts due to EOL LIB collection and transportation. Very few companies currently process LIBs. In the U.S. recycled LIBs only account for a minimal portion of the total number of EOL LIBs entering the waste stream per year. The base case in the model developed by Wang *et al.* shows that current battery collection rates would only generate enough EOL LIBs to enable four recycling facilities to operate with profit in the U.S. The pre-processing steps that were added in this study would probably enable a couple more to operate but it will not be a significant increase. A comprehensive LIB recycling network must be developed by performing analysis based on LIB recycling potential, the risks, as well as the uncertainties associated with recycling.

A large volume of EOL LIBs will be entering the waste stream in the near future due to the rapid adoption of EVs. To accommodate this flow a feasible, automated, and low=cost recycling process should be developed. The pre-recycling process that is proposed in this study requires only a few pieces of additional equipment, has a low energy consumption, and has the potential for scale-up quite easily.

LIB technology is moving toward less expensive cathode materials. When that happens, incentives to recycle those batteries will diminish. If the fraction of $LiCoO_2$ batteries will fall below 21% of the total EOL LIB stream, a recycling facility will not be profitable anymore (when the fixed and variable cost of the base case are assumed). In the future if prices or recycling rates of valuable metals (Co, Cu, Li, etc.) increase the economic incentive to recycle batteries will increase. This pre-recycling step aims to increase efficiency of recycling to make it more economically attractive to recycle EOL LIBs.

In any case extended producer responsibility (EPR) laws must be implemented to supplement market-based recycling initiatives. Recycling-oriented policies must be implemented which make it mandatory to recycle EOL LIBs. This would ensure that EOL LIBs are properly recycled and/or reused.

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