



Economic and environmental characterization of an evolving Li-ion battery waste stream



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ABSTRACT

While disposal bans of lithium-ion batteries are gaining in popularity, the infrastructure required to recycle these batteries has not yet fully emerged and the economic motivation for this type of recycling system has not yet been quantified comprehensively. This study combines economic modeling and fundamental material characterization methods to quantify economic trade-offs for lithium ion batteries at their end-of-life. Results show that as chemistries transition from lithium-cobalt based cathodes to less costly chemistries, battery recovery value decreases along with the initial value of the raw materials used. For example, manganese-spinel and iron phosphate cathode batteries have potential material values 73% and 79% less than cobalt cathode batteries, respectively. A majority of the potentially recoverable value resides in the base metals contained in the cathode; this increases disassembly cost and time as this is the last portion of the battery taken apart. A great deal of compositional variability exists, even within the same cathode chemistry, due to differences between manufacturers with coefficient of variation up to 37% for some base metals. Cathode changes over time will result in a heavily co-mingled waste stream, further complicating waste management and recycling processes. These results aim to inform disposal, collection, and take-back policies being proposed currently that affect waste management infrastructure as well as guide future deployment of novel recycling techniques.

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1. Introduction

Development of renewable energy systems is contingent on concurrent technological development of energy storage systems, primarily batteries used for power grid load-leveling, renewable energy storage, and hybrid and all-electric vehicles. In this sector, lithium-ion batteries (LIBs) are emerging as a competitive technology due to higher power and energy densities compared to lead-acid and nickel-metal hydride chemistries (Howard and Spotnitz, 2007). These advantages have led to a dramatic shift to LIBs in a variety of devices including digital cameras, power tools, portables computers, cell phones, and electronic readers. To illustrate this shift, Fig. SI 1 in the supplementary information compares the chemistries of batteries in cell phones produced in 1996 (A) to those produced in 2005 (B). While older cell phones were powered by a variety of battery technologies, mobile phones produced in

2005 (and currently) use lithium chemistry almost exclusively. This shift can be seen for batteries in laptop computers as well (Fig. SI 1C and D), and a similar pattern holds true for camcorders, digital cameras, and other types of portable electronic applications (Wilburn, 2008). LIBs represent close to 20% of the total rechargeable battery market and 75% of the portable rechargeable battery market, which is the fastest growing segment of the rechargeable battery market, with a 20% global growth in 2008 (AE, 2009).

This surge in production of LIBs indicates a potentially looming waste problem: total waste from Li-ion batteries has likely grown substantially from the 500 metric tons estimated in 2003 (Lee and Rhee, 2003). Demand projections for electric vehicles also indicate a potentially exponentially increasing amount of waste Li-ion batteries (Gaines and Nelson, 2009, 2010; Gruber et al., 2011). California and New York state legislators have attempted to proactively address this waste challenge by issuing disposal bans on rechargeable batteries in their states (CACode, 2006; New York State Rechargeable Battery Law, 2010); similar bans exist as part of the Battery Directive in the European Union (EC, 2006). However, eliminating landfill as a disposal option means that alternative end-

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of-life management strategies must be developed, particularly infrastructure for recycling valuable metals contained within the battery. Research on efficient extraction of specific materials contained in LIBs has increased dramatically over the last decade; excellent reviews of such work are provided in [Bernardes et al. \(2004\)](#), [Xu et al. \(2008\)](#) and [Zeng et al. \(2013\)](#). However, ability to expand LIB recycling will be heavily dependent on the potential economic costs or revenues associated with material recovery from this poorly-characterized and continually evolving waste stream. Some work has been done on specifics of this issue such as costs of electric vehicle batteries ([Armand and Tarascon, 2008](#); [Scrosati and Garche, 2010](#)); however, a systematic review of potential value based on chemistry has not been conducted.

The wide variety of sizes, form factors, cathode chemistries, and morphologies indicate that the recycling infrastructure created will need to be responsive to a waste stream with diverse and continually changing materials. Introduction of large batteries for hybrid and all-electric vehicles will also complicate any forecasts in production volume by cathode, particularly given that each automotive company has chosen a different chemistry to pursue. For example, the Chevy Volt uses a Mn-spinel and mixed metal cathode ([Fletcher, 2011](#)), the Tesla Roadster and Model S use cobalt-based cathodes ([Schneider, 2007](#)), the Coda Sedan and the Tata Nano use LiFePO_4 ([Hernandez, 2011](#); [Lucas, 2012](#)), and the Nissan Leaf uses LiMn_2O_4 ([Hernandez, 2011](#)). Existing battery waste processors are, by necessity, largely backwards-looking. That is, they typically plan for recovery of materials from battery technologies that are several years behind those currently under development by manufacturers, as these will not appear in the waste stream until the lifespan of the products they are contained in has ended ([Dewulf et al., 2010](#); [Sloop, 2008](#)). As a consequence, battery recycling facilities are focused on recovery of high value, high volume materials contained in Li-ion batteries (e.g. cobalt, nickel, copper) and many are unable to economically recover other materials (e.g. lithium, electrolyte, mixed metals). As a result, current battery recycling rates are comparatively low (not including lead-acid), despite the valuable materials currently found in batteries. Beyond the potential economic benefit from increasing battery recycling ([Nan et al., 2005, 2006](#)), significant environmental gains could also be realized by recovering high embodied energy materials and offsetting future virgin material extraction demand ([Dewulf et al., 2010](#)).

This work provides necessary first steps towards evaluating battery recycling infrastructure by quantifying the dynamic linkage between evolving LIB cathode chemistries and potential end-of-life material value and environmental impacts. Specifically, LIB cathode chemistries representing recent shifts toward low-cost materials were characterized based on materials they contained and analyzed to project potential economic value recoverable from an evolving LIB waste stream. This value was also assessed for sensitivity to compositional variability of a co-mingled waste stream and the expected yields associated with common recycling technologies. The aim of this work is to highlight the economic and environmental opportunities and tradeoffs of LIB recycling as a catalyst for research and development of novel recovery technologies as well as inform the development of proactive policies for LIB end of life management.

2.0. Methodology

2.1. Battery selection and scenario analysis

Battery compositions for a set of LIB cathode chemistries were determined using reported compositions from the literature, MSDS sheets for products containing Li-ion batteries, and bills of materials determined through physical disassembly. The base case

analysis focuses on LiCoO_2 cathode chemistry as this is the prevalent chemistry found in most consumer electronics which make up the majority of the current Li-ion battery waste stream. The “18650” cylindrical form factor (18 mm diameter, 65 mm length) was chosen, as this is a common form factor for laptops and power tools. The compositions of this same cathode chemistry and form factor were compared for seven different manufacturers.

In the cross-cathode comparison, the following chemistries were included: LiCoO_2 , LiFePO_4 , LiMn_2O_4 -spinel, and a mixed metal cathode $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3})_2$. These four cathode chemistries encompass the most relevant Li-based technologies for current consumer electronics and current and near-term electric vehicles as detailed in Section 3.2. For all of the cases, the battery form factor was held constant for comparison purposes. While electric vehicles will certainly make use of additional form factors (e.g. prismatic plate, pouch, etc.), the availability and quality of data with which to characterize the 18650 cells is much better, which provides a consistent basis to identify material-specific issues across chemistries. This analysis can then be expanded as other form factors become more prevalent.

Realistic recycling scenarios were also analyzed assuming a co-mingled stream and differing degrees of recycling rate and yield. For the current co-mingled case, it was assumed that the LiCoO_2 cathode chemistry would continue to dominate the waste stream (at 85 wt.%), with small amounts (5 wt.% each) of the other three prevalent cathode chemistries part of the overall mix. For the future co-mingled scenario, it is assumed that development of high performance cathode materials coupled with further penetration of electric vehicles will drive a weight percentage increase (to 10 wt.%) of non-cobalt based chemistries present in a mixed waste stream.

The base case, cross-cathode comparison, and co-mingled case assume that all of the materials within the battery can be successfully recovered implying a 100% yield rate. This provides policy-makers and waste management professionals with a “best” case scenario for recycling; however, it is likely that yield will be significantly lower. Yield for recycling Li-ion batteries at an industrial scale is currently unknown as the recycling infrastructure could take many forms: pyrometallurgical, hydrometallurgical, and a variety of mechanical/physical pre-sorting technologies are in development both at the lab-scale and industrially ([Dunn et al., 2012](#); [Espinosa et al., 2004](#); [Hart and Amarakoon, 2012](#); [Xu et al., 2008](#)). Three cases were analyzed to characterize changes in economic performance due to lower yields: a “medium” case of mid-value demonstrated yields from lab-scale pyrometallurgical and hydrometallurgical recycling technologies, a “high” case of the highest demonstrated lab-scale yield, and a “low case” of the current municipal solid waste (MSW) recycling rate (Equation (1)) for materials in the battery stream.

$$\text{RR} = \frac{\text{consumed old scrap} + \text{consumed new scrap}}{\text{apparent supply} + \text{imports} - \text{exports} + \text{adjustment}} \quad (1)$$

Table 1
Demonstrated lab-scale yields (both medium and high) for battery materials of interest.

	Mid-Value	High-Value
Cobalt	80% (Dorella and Mansur, 2007)	99% (Paulino et al., 2008)
Lithium	55% (Mantuano et al., 2006)	100% (Ferreira et al., 2009)
Aluminum	55% (Dorella and Mansur, 2007)	98% (Li et al., 2010)
Copper	10% (Li et al., 2009a)	90% (Nan et al., 2006)
Nickel	90% (Nan et al., 2006)	99% (Shin et al., 2005)
Manganese	92% (Paulino et al., 2008)	98% (Mantuano et al., 2006)

For the demonstrated yields, values for cobalt, lithium, aluminum, copper, nickel, and manganese are shown in Table 1; other contained materials were assumed to be at their MSW recycling rate (Table 2) as further recovery has not been successfully demonstrated yet. Each of the scenarios investigated and their associated parameters are summarized in Table 3, including those used for analysis of economic and environmental impacts, with additional methods described in subsequent sections.

2.2. Battery compositions from disassembly

The total cell mass was recorded before disassembly, and any losses in the total mass before and after disassembly were assumed to be evaporated electrolyte. Three 1/2-inch diameter circular samples were punched from areas in each electrode with adhered coatings to the metal current collector and from the separator as well. Electrolyte contained in these samples was evaporated by drying in a vacuum oven at 100 °C. The adhered coatings were removed from the dried electrode, leaving the metal current collector (Al for cathode and Cu for anode), the mass of which was directly measured. The coating mass was then calculated as the difference between the masses of the total dry electrode and the current collector. The mass percentages of electrode coating and current collector in the samples were then multiplied by the total electrode mass to scale up findings for the total electrode coatings and current collector masses in the cell. The electrode coating contains an active material, polymer binder, and carbon conductive additive. The mass of the active material was calculated by dividing the cell capacity (mAh) by the specific active material capacity (mAh/g). The carbon conductive additive and polymer binder (PVDF) were then assumed to be of equal percentage of the remaining electrode coating mass. This methodology follows the lab-scale disassembly of others (Contestabile et al., 2001; Li et al., 2009b).

2.3. Potential value

For each material category identified through disassembly, representative commodity values were obtained to estimate the maximum economic value of an EOL LIB stream. This estimation assumes both primary commodity pricing for materials and no material losses due to recycling inefficiencies. This is referred to as the “theoretical maximum value” throughout the remainder of the paper and represents the upper limit to the economic value associated with the waste stream, and would be reduced in reality once real secondary values and processing yields are taken into account. Although yield and secondary stream material values are unknown at this time, sensitivity analysis was performed in Section 3.4 to investigate how these might impact the economic return.

Current commodity prices have been significantly volatile, with large day-to-day swings. Regardless, to reflect current value, average spot prices for metals and plastics in March 2012 were collected from the London Metals Exchange (LME), American Metal

Table 3

Summary of parameters for analyzed scenarios.

	Sec.	Cathode chemistries (# manufacturers)	Yield
Base Case	3.1	LiCoO ₂ (7)	100%
Cross-Cathode Comparison	3.2	LiCoO ₂ (7) LiFePO ₄ (3) LiMn ₂ O ₄ -spinel (3) Li(Ni _{1/3} Mn _{1/3} Co _{1/3}) ₂ (4)	100%
Co-mingled Case	3.3	Current: 85% LiCoO ₂ , 5% LiFePO ₄ , 5% LiMn ₂ O ₄ -spinel, 5% Li(Ni _{1/3} Mn _{1/3} Co _{1/3}) ₂	100%
Variable recovery cases	3.4	LiCoO ₂ (7) LiFePO ₄ (3) LiMn ₂ O ₄ -spinel (3) Li(Ni _{1/3} Mn _{1/3} Co _{1/3}) ₂ (4)	US recycling rates High and average lab yields
Environmental impacts	3.5	LiCoO ₂ (7) LiFePO ₄ (3) LiMn ₂ O ₄ -spinel (3) Li(Ni _{1/3} Mn _{1/3} Co _{1/3}) ₂ (4)	100%

Market (AMM), and a scrap trading website, GlobalScrap. These values were averaged both by geographic area and over the month time span and are available in the supporting information (Table SI 1). This variability is particularly relevant for lithium, as the United States Geological Survey (USGS) reports a significantly low value, while lithium spot prices have increased exponentially over the last year. Prices were taken from Alfa Aesar assuming bulk discount for the electrolyte and binder materials that may be recoverable. The range of types of plastics used in Li-ion batteries make it challenging to select a specific value. Some high quality plastics have end-of-life value as high as \$0.35/kg, however, plastics recovered from batteries will likely be co-mingled thermosets and therefore a lower, more typical, average value was used. As with the other materials, contamination and co-mingling may decrease this value significantly.

2.4. Environmental considerations

A variety of environmental metrics are available to evaluate end-of-life LIB impacts such as greenhouse gas emissions, eco-toxicity, and human health effects. However, comprehensive life-cycle inventory and impact assessment data for LIBs have yet to be quantified (Sullivan and Gaines, 2012). Therefore, cumulative energy demand (CED) was selected as a representative metric of the environmental impact of materials contained in the LIBs (Huijbregts et al., 2005, 2010); these values are reported in the supporting information Table SI 2. CED includes all direct and upstream energy inputs associated with mining, refining, and processing LIB materials from “cradle-to-gate,” but does not take into account the assembly and transportation of the LIBs once they have been fabricated from the supply materials. Refining has been included in this cumulative energy but should be considered a minimum and therefore conservative estimate for the CED as inventory data does not exist for many of the extremely high purity materials included. There is a particularly high degree of uncertainty in the magnitude of CED for the advanced anode and electrolyte materials. However, a considerable amount of energy is required for mining, manufacturing, and transporting primary metals. Recycling provides an opportunity to recapture some of this energy albeit with its own set of environmental impacts. Energy data was taken from life-cycle assessment (LCA) software databases (ecoinvent v2.2 within SimaPro 7.2) and calculated according to the impact assessment methodology “cumulative energy demand v1.07” (Hischier et al., 2010).

Table 2

US 2010 recycling rates (low value) from USGS for battery materials of interest (Jaskula, 2010; USGS, 2013; Wilburn, 2008).

Recycling rate (Equation (1))			
Aluminum	46%	Iron	41%
Cobalt	0%	Graphite	0%
Copper	30%	Carbon	0%
Lithium	0%	LiPF ₆	0%
Manganese	33%	PVDF	0%
Nickel	41%	Binders	0%
Steel	61%	Plastic	30%

Eco-toxicity is used as a representative metric of the environmental impact of releasing LIB materials into the environment, and measures the potential for pollutants (both natural and synthetic) to cause stress to ecosystems (including plants, animals, and humans). A variety of eco-toxicity metrics exist and have been widely used to identify chemical hazards (Bascietto et al., 1990). For this study, the eco-toxicity metric used is based on the 2007 Priority List of Hazardous Substances from the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) (ATSDR, 2007); reported in supporting information Table SI 2. CERCLA provides comprehensive information about eco-toxicity of hazardous substances, taking into account the frequency of occurrence of substances at national priorities list (NPL) hazardous waste sites and facilities, the Environmental Protection Agency's (EPA) reportable quantity ranking, EPA toxicity score, and the potential for human exposure; detailed information on how this score is calculated is available in the CERCLA support document (ATSDR, 2011). This comprehensive metric avoids a narrow view of eco-toxicity that a single indicator metric such as LC50 (the median lethal concentration) or TD50 (the median toxic dose) may provide. While CERCLA eco-toxicity points are not normalized by a mass or volume metric, a points per kilogram extrapolation was performed in order to weight the compositional differences of materials within LIBs as detailed in the supplemental information.

3.0. Results and discussion

3.1. Base case: LiCoO₂

The base case analysis uses the average compositional values from both literature and physical disassembly for sixteen different LiCoO₂ cathode LIBs representing seven different manufacturers. Even though this set of sixteen batteries shares the same cathode chemistry and form factor (18650), significant variability can be seen in Fig. 1. Coefficient of variation (CV), the standard deviation normalized by the mean, ranges from 21% for binders to 126% for carbon black. The base metals have relatively lower variability ranging from 21% for steel to 37% for aluminum; however, this degree of variation is still a significant source of concern for recyclers. As cobalt is one of the key materials targeted for recycling and recovery, it is interesting to note that the standard deviation of ± 1.8 g Co could result in a range of secondary values between

\$0.15–\$0.42 per 18650 cell (based on March 2012 spot prices). This difference is quite extreme when extrapolated to volumes of spent batteries that may be processed by a typical recycler. For example, a current lead-acid battery recycling facility in the US processes between 132,000–176,000 metric tons per year (Roell, 2011). Spot prices for commodity metals have significant volatility, however, even 2011 average USGS prices show an even larger range in value of \$0.18–\$0.50 per 18650 LiCoO₂ cell.

It is again emphasized that this is the maximum theoretical value for recyclers, as the calculation assumes that high yield recovery of all materials from the LIB is possible. These results show that there is significant potential for valuable resource recovery; given an average weight of 40.5 g per 18650 cell, one metric ton of this scrap could be worth \$4400–\$10,400. The cost to collect and process these scraps would make the profit margin significantly less than this total, however, compared to other scrap materials this is still quite valuable. For example, mixed electronic scrap sells in the range of \$1000 per metric ton and up to \$8000 per metric ton for high grade sorted printed circuit boards (MW, 2012). Ferrous scrap from shredded automotive hulks averages around \$525 per metric ton (USGS, 2012). This strong economic incentive may be a likely driver for many electronic waste processors moving toward processing batteries as well.

The compositional uncertainty seen for the LiCoO₂ cathode chemistry LIBs is present for other cathode chemistries as well. Detailed results for LiFePO₄ 18650 cells from three different manufacturers (two each from Tenergy, A123, and Sony) are shown in the supporting information Fig. SI 2.

3.2. Technology trajectory: cross-cathode comparison

While LiCoO₂ cathode chemistries dominate in terms of current manufactured volume, significant progress in the rechargeable battery field has been made through research and development of late. Increases in energy density and reduction in costs have been found through exploration of other cathode chemistries as well as changes in anodes, cans, and processing routes. A variety of chemistries are currently being used commercially, with many more being actively developed for future high volume applications, namely transportation. Fig. SI 3 in the supplementary information shows an approximate chronological progression of explored cathode chemistries, illustrating a slight improving trend in energy

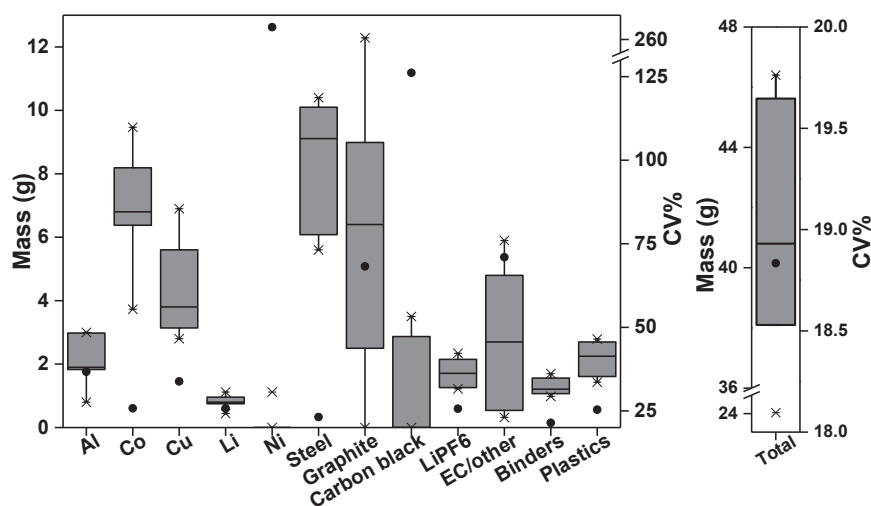


Fig. 1. a) Statistical analysis of sixteen LiCoO₂ LIBs for various material categories, b) analysis for the total weight, the gray box represents the 25th–75th percentile with the line being the median, x's present the 1st–99th percentile, and the black dot is the coefficient of variation. Detailed legend for statistics presented shown in Fig. SI 3.

density. While it is not clear which cathode types may be the next generation in high volume production, LiFePO_4 , LiMn_2O_4 -spinel, and mixed metal type cathodes have emerged as clear contenders, particularly for electric vehicles which may require a significant volume of batteries as their penetration increases. Examples of EVs with these battery types were detailed in the introduction. Regardless of which batteries dominate production, these changes in battery composition in all demand sectors will have significant impacts on the stability and profit of recycling infrastructure.

Not surprisingly, batteries with varying cathode chemistries have significantly different overall material compositions. Fig. 2A shows the compositional breakdown by weight for three cathode chemistries chosen to represent batteries with high likelihood of gaining considerable market share compared to LiCoO_2 . All four are taken from 18650 form factor cells with a minimum sample size of four per chemistry. While base metals as a whole make up the largest fraction of total mass, thus driving metrics of interest (value, embodied energy, eco-toxicity, etc.), there is little consistency in composition or amount of specific metals (Fig. 2B). Steel makes up a significant portion of the base metal weight for most of the battery types due to its use in the “can”, the outer packaging of the cell, however some cells use aluminum as the can such as the mixed $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3})_2$ cathode (labeled as LiMO_2). Regardless of the remainder of the composition, cobalt has the most impact on recoverable value. Fig. 3 compares the values of these four cathode chemistry types given commodity market prices for different years. Mn-spinel and iron phosphate cathode batteries have potential material values 73% and 79% less than cobalt cathode batteries, respectively.

This analysis assumes lithium can be economically extracted at scale, implying that the spot price of lithium would have to reach over 600% its present value before these two chemistries had EOL values comparable to cobalt based chemistries. Even for the mixed metal cathodes, which contain a large proportion of cobalt, reaching an EOL value on par with wholly cobalt based chemistries would require high purity extraction of both copper and nickel; technology which has not been successfully developed or scaled-up yet. This emphasizes the need to develop a recycling infrastructure

for LIBs that is robust enough to handle processing materials beyond cobalt in order to ensure profitability for waste managers. Fig. 3 highlights the need to recover lithium for future cathode chemistries such as LiMn_2O_4 and LiFePO_4 as it makes up a significant portion of the total value. For these cathode chemistries, copper and steel (included in “other”) also become significant portions of the total value which would incentivize prioritizing their recovery as well. As stated previously, these are also theoretical maximum values assuming a high degree of purity and consistency in the LIB waste stream; actual recycling rates as explored in Section 3.4 will lower the profitability as well. Waste managers may need further investment in sorting and separation technologies to achieve a higher purity stream as explored more fully in the following section.

3.3. Managing a mixed stream: co-mingled case

Compositional uncertainty that arises from having a co-mingled scrap stream can create a barrier to recycling by raising processing costs, increasing the likelihood for off-specification products, and complicating batching management (Gaustad et al., 2007; Olivetti et al., 2011). Two key mechanisms for co-mingling for Li-ion batteries at EOL are a) being mixed in with other electronic waste as they are often not removed from laptops, cell phones, etc. upon disposal, and b) the wide variety of form factors and cathode chemistries mentioned previously. For most consumer electronic LIBs, the former is most likely as these scraps are often shredded; a pre-processing step performed by most studies researching hydrometallurgically based recovery technologies and being done by many industrial recyclers. For electric vehicles, the latter is the most probable mechanism as H/EV batteries will likely have a dedicated collection and processing infrastructure. To illustrate the degree of uncertainty that one of these aspects, cathode chemistry, would contribute to overall composition of the mixed stream, a hypothetical mixture of different cathodes and manufacturers (all 18650 cells) was created. This mixed scenario was dominated by LiCoO_2 cells reflecting the product make-up of the current EOL stream, however, other cathode chemistries were present, thus

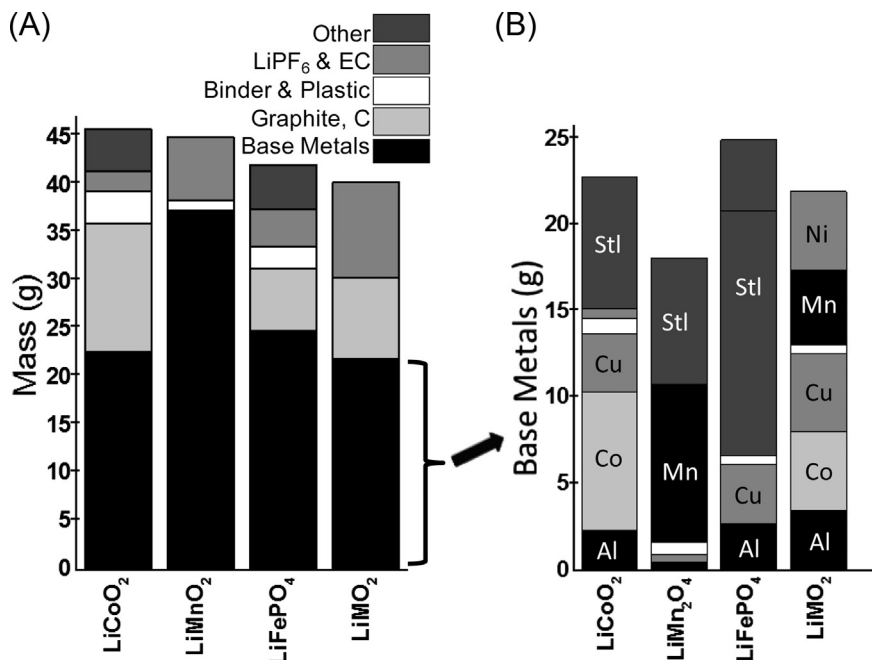


Fig. 2. A and B. Compositional breakdown in total (A) and for base metals (B) for 18650 cells of varying cathode chemistry.

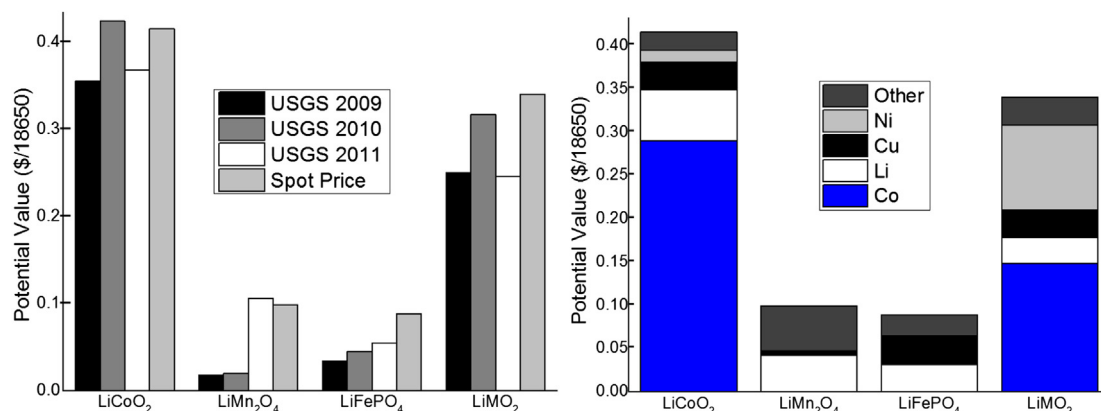


Fig. 3. Cross cathode comparison of potential value per 18650 form factor battery.

contributing to higher overall uncertainty. Fig. SI 4 in the supplemental shows that the coefficient of variation for the base metals ranges from 70% (Li) to 175% (Al) with accompanying significant ranges in weight percent. It is assumed that batteries mixed with other electronic waste would have an even higher coefficient of variation.

3.4. Variable recovery cases: yield, recycling rate, and disassembly effects

Up to this point, the discussion of economic attributes of a LIB waste stream has centered on the maximum economic content of the waste stream itself, rather than the actual recoverable value, once recycling process inefficiencies are accounted for. This consideration is treated separately here, due to the different end-of-life avenues potentially available to batteries. As highlighted in Table 1, a number of hydrometallurgical studies have demonstrated yields above 90% at the lab-scale for a variety of battery materials. However, many of these recycling technologies involve a multitude of steps, are still in development, and reported efficiencies at the lab scale may not be reproducible once the recycling infrastructure is scaled up and complicated by the compositional and co-mingling issues raised earlier in this paper. Some companies (e.g. Toxco, Umicore) have begun successfully scaling up battery recovery facilities, however the focus has been on high yield of cobalt with other materials being recovered at significantly lower rates.

Here, we examine how the potential recoverable economic value changes based on three scenarios of recycling efficiencies: highest known (lab scale) efficiency (high), average lab-scale

efficiency (medium), and US cumulative recycling rate (low). Fig. 4 shows that relying on current recycling rates results in significantly less recoverable value of the Li-ion battery scrap stream ranging from 23% for iron-phosphate cathodes to 5% for cobalt based cathodes. It should be noted that on an absolute basis, the value is still much higher for cobalt based cathodes even at 5% compared to others due to its overall higher value (cf. Fig. 3). More surprising is that current demonstrated lab-scale maximums can achieve quite close to the maximum economic value ranging from 89% for Mn-spinel cathodes to 96% for both mixed metal and cobalt based cathodes. Each set of technologies used to achieve these yields will have cost trade-offs as well, for example, it could be assumed that the low case would not incur much additional cost as the yield depends on the recycling infrastructure already in place for each of those materials. The medium and high yield cases would incur much higher costs and both capital investment as well as scale-up research and development would need to be implemented in order to achieve those higher yield rates. None of the studies cited in Table 1 have conducted cost or economic related analysis for their recovery technologies.

One other potential recycling avenue is the partial or full disassembly of Li-ion batteries for recycling given safety concerns of shredding and sorting processes (Herrmann et al., 2012; Lain, 2001; Schneider et al., 2009). While some of these studies performed such disassembly by hand for their work, a quantitative assessment based on resulting compositional yield and purity has not been conducted. Here, we compare the relative mass and

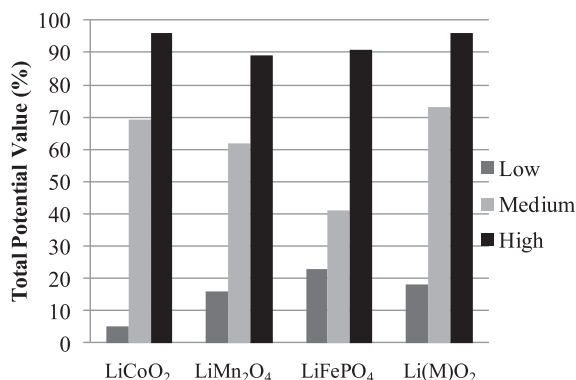


Fig. 4. Percentage of the total recoverable value (spot price value in Fig. 2) achieved with potential yield scenarios.

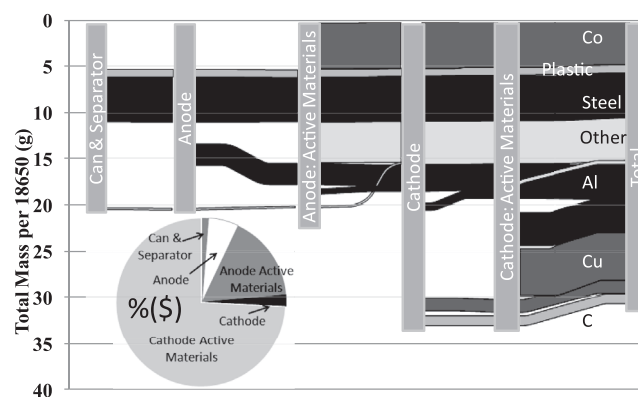


Fig. 5. Sankey diagram of disassembly showing portion of total weight by component, in order of disassembly for a LiCoO₂ cathode chemistry 18650; inset pie shows the breakdown by percent value.

economic value recoverable at each stage of the disassembly process, if a manual recovery system was employed. One can see that a majority of the mass is contained in the anode and cathode active materials. If one were to extrapolate from the value results in Fig. 3, it is clear that the majority of the value is in the cathode active materials which are accessed at the last stage of disassembly shown in Fig. 5. This would indicate that partial or component-level disassembly would not be economically viable; however, the economics of disassembly compared to shredding combined with a hydrometallurgical or pyrometallurgical process have not been explored.

3.5. Environmental considerations

To assess environmental impacts, the cross-cathode comparison was revisited (Table 3) and average mass was used for each of the cathode chemistries. For this analysis, yield is not a relevant factor as the environmental impacts are assessed for the total magnitude of materials contained within a battery. Similar to the economic results, the base metals dominate as major contributors to the life-cycle cumulative energy demand (CED) of all battery materials. While only making up a small portion of the weight percent (cf. Fig. 2), aluminum makes up a large percentage of the total CED due to the energy intensity of Hall-Heroult process. Particularly, it accounts for 25% of the base case LiCoO₂ cathode, 45% of the LiFePO₄ cathode, and 25% of the mixed metal cathode (Fig. 6A). Diverging from economic results, a key contributor for all four battery types is the carbon black and graphite contained in the cells, ranging from 19% for LiFePO₄ to 42% for the LiMn₂O₄ cathode (but less than 2% by value for these same chemistries). Battery grade carbon and graphite require additional purification steps that add to their overall energy impact. For the mixed metal cathode, the other category is dominated by manganese (6%), steel and lithium (5% each), and copper (3%). The steel casing dominates the other category for the LiFePO₄ cathode at 14%, followed by lithium (8%) and copper (5%). Not surprisingly, manganese is the main contributor in the other category for LiMn₂O₄ cathode battery at 22%, followed by lithium (11%) and the steel can (8%). For the cobalt base case, lithium is the largest of the other category at 10% of the total embodied energy with steel accounting for 5%.

The difference in CED between the four cathode chemistries is not as dramatic as the difference in potential economic value. The iron phosphate and manganese based cathode chemistries have roughly 35% less lifecycle CED compared to the cobalt based chemistry (compared to nearly 80% less economic value). For the

most part, these results indicate an alignment of economic and energy incentives regarding prioritization of material recovery: the base metals are clear priorities. Thus, additional policy is likely unnecessary to ensure optimization of recycling from an energy perspective for the profit based infrastructure in place currently. One key difference, however, is the importance of carbon black and graphite from an energy perspective, whereas these materials are low recycling priorities when considering value or potential recovery infrastructure.

In considering eco-toxicity of materials contained in the battery, should they be released during recycling, the CERCLA point system was adapted by multiplying material-specific points by the mass of each material in the battery. Following the economic and energy results, the base metals are again the major contributors to potential eco-hazard. Nickel, cobalt, manganese, and copper have the highest potential according to CERCLA, and even though their weight percent is comparatively low, they become the key contributors to overall eco-toxicity risk. Not surprisingly, cobalt and manganese are respectively highest for chemistries in which they are dominant metals (Fig. 6B). The mixed metal cathode has total potential risk roughly evenly divided among its mix of included metals and actually has a 10% higher potential risk compared to the cobalt base case, mainly due to the inclusion of nickel. LiMn₂O₄ cathode batteries have 30% less potential risk compared to the cobalt base case and LiFePO₄ has a 62% reduction in potential eco-toxicity risk. While phosphorus has relatively high CERCLA points, its inclusion as an oxide poses little risk; however, it should be noted that certain acid-based hydrometallurgical recycling routes have the potential to reduce and therefore release contained phosphorus.

4. Conclusions and implications

Results shown here suggest that environmental and economic perspectives lead to generally consistent prioritization of materials to be recovered from a LIB waste stream. However, the actual recovery system will need to take into account geographically based regulations on emissions, disposal bans, and producer responsibility. The metrics presented here are somewhat narrowly scoped for a first approximation; but can be expanded into a more holistic life-cycle assessment as additional data on recycling processes and the upstream battery manufacturing processes become available.

Further consideration of EOL battery management must also take into account recent trends toward use of nano-scale materials

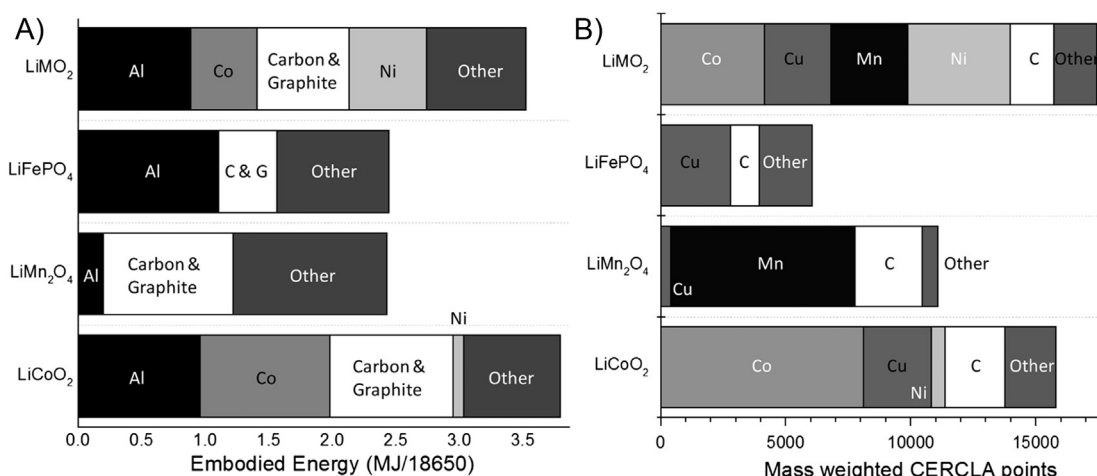


Fig. 6. A) Estimated embodied energy for four selected case study cathode chemistries showing key contributors, B) EPA CERCLA points weighted by mass.

in next-generation cathode chemistries. There are considerable environmental uncertainties and concerns surrounding nanoparticles contained in products, particularly at end of life. These concerns include fate of nano-particles in landfills (Reinhart et al., 2010), in waste water (Brar et al., 2010; Hull et al., 2009), soil leaching (Lowry and Casman, 2009), and disposal in general (Breggin and Pendergrass, 2007; Gao et al., 2008). To this end, a number of efforts are underway within the research and regulatory community to better quantify battery system impacts due to contained nanomaterials. For example, the U.S. EPA has recently launched a Lithium-ion Batteries and Nanotechnology Partnership involving industry, government and academia (including several of the authors) to conduct a life-cycle assessment (LCA) of LIBs used in electric vehicles, including those using single-wall carbon nanotube (SWNT) technology in the battery anode (Hart and Amarakoon, 2012). More studies of this nature must be conducted to produce data that can be used to expand environmental impact assessment methods and metrics discussed here. Work to quantify the risk of particulate exposure during end-of-life processing as well as leaching risk during disposal is also necessary to understand the comprehensive life-cycle impacts of lithium ion batteries.

Given a profit-based waste management system, results presented here help stakeholders comprehend what materials within lithium-ion batteries should be prioritized for recovery. Particularly, results show which materials may require additional policy intervention to overcome economic limitations. As automotive batteries for hybrid and all-electric transportation applications shift towards different form factors, sizes, and configurations, the relative mass contributions to battery components will change. For example, prismatic cells will have a higher ratio of cathode and anode materials to packaging (can and plastics). The choices that automotive companies make in cathode chemistry coupled with speed of adoption will greatly impact the value of the stream seen by recyclers. While it is clear that economics of recovery and recycling will be impacted by a transition away from cobalt based chemistries, the rapidity and magnitude of this shift are still unclear.

The preceding results suggest policy or other incentives may be necessary to promote a robust recycling infrastructure as the economic incentives will likely decrease as the battery stream changes. For example, widespread battery disposal bans will likely be needed to ensure collection and recovery of Li-based batteries of many next-generation cathode chemistries that exclude high-value cobalt in their composition. Regulation concerning energy savings and eco-toxicity will also favor cathode chemistries without cobalt included. Design for disassembly as a strategy to encourage enhanced recycling efforts will likely not have a significant impact, at least on a single cell basis. Large packs for hybrid and all-electric vehicles may still benefit from such proactive engineering design. It also appears that any collection or recycling infrastructure will likely need the capacity to process a co-mingled LIB scrap stream. Future work aims to model the future flows of the entire waste stream, taking into account significant changes in cathode chemistries, volume, and form factor. These results will inform both incentives for build-out of collection infrastructure as well as economically efficient recycling strategies.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.jenvman.2014.01.021>.

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