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# Targeting high value metals in lithium-ion battery recycling via shredding and size-based separation

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ABSTRACT

Development of lithium-ion battery recycling systems is a current focus of much research; however, significant research remains to optimize the process. One key area not studied is the utilization of mechanical pre-recycling steps to improve overall yield. This work proposes a pre-recycling process, including mechanical shredding and size-based sorting steps, with the goal of potential future scale-up to the industrial level. This pre-recycling process aims to achieve material segregation with a focus on the metallic portion and provide clear targets for subsequent recycling processes. The results show that contained metallic materials can be segregated into different size fractions at different levels. For example, for lithium cobalt oxide batteries, cobalt content has been improved from 35% by weight in the metallic portion before this pre-recycling process to 82% in the ultrafine (<0.5 mm) fraction and to 68% in the fine (0.5–1 mm) fraction, and been excluded in the larger pieces (>6 mm). However, size fractions across multiple battery chemistries showed significant variability in material concentration. This finding indicates that sorting by cathode before pre-treatment could reduce the uncertainty of input materials and therefore improve the purity of output streams. Thus, battery labeling systems may be an important step towards implementation of any pre-recycling process.



## 1. Introduction

### 1.1. Key challenges remain in LIB waste management

Lithium ion batteries (LIBs), as an emerging technology, currently dominate the power source market for portable consumer electronics. More recently, LIBs have started being used in electric vehicles (EVs) and are becoming more popular due to their high energy density, no memory effect, long cycle life, etc. In 2006, $1.1 billion of LIBs were consumed globally (BU, 2011a); according to Research and Markets, the global market for LIBs is expected to reach $25 billion by 2017 (Wood, 2013). After their use phase (ranging from 2 years for consumer electronics batteries to about 10 years for EV batteries), a large amount of end-of-life (EOL) LIBs will enter the waste stream (Richa et al., 2014). Even though LIBs contain less hazardous materials compared to lead acid batteries or nickel–cadmium batteries, there is still a potential for some toxic materials to leach and contaminate the ground water system when disposing of EOL LIBs into uncontrolled landfills (Majeau-Bettez et al., 2011). Additionally, many metallic materials in EOL LIBs still have economic value (Wang et al., 2014a) and may have associated criticality or scarcity concerns regarding their supply. Particularly for LIBs, niobium, cobalt, and manganese have been deemed critical by various organizations (Commission, 2014). Although the collection rate of EOL LIBs has been improving over the years, it is still extremely low. For example, only 0.5% of EOL LIBs were collected in the EU in 2002, and this figure only improved to 2.7% by 2007 (Weyhe, 2008). Currently, a few companies (e.g., Umicore and Toxco) process EOL LIBs; however their recycling technologies were not designed specifically for LIBs, usually processing multiple types of rechargeable batteries (e.g., nickel-metal hydride batteries) and/or non-battery scraps (e.g., metallic materials) at the same time, which results in lower recycling efficiency (Umicore; Olapiriyakul and Caudill, 2009). Therefore, development of a technology that can effectively recover more types of materials contained in LIBs is important.

A growing number of studies have been performed on EOL LIB recycling, with the focus on improving the recycling efficiency of cobalt, which is the most valuable material contained in LIBs from consumer electronics (Shin et al., 2005; Dorella and Mansur, 2007; Xia et al., 2008; Li et al., 2009). However, other materials contained in LIBs also show motivation to be recovered at a higher level, when considering both economic and environmental perspectives

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(Wang et al., 2014a). Copper, nickel, and lithium make a significant contribution to the potential recoverable value of EOL LIBs together, ranging from 27% of the total material value for $LiCoO_2$ cathode LIBs to 74% of the total material value for $LiFePO_4$ cathode LIBs. While the aluminum content is relatively low (ranging from 1% to 8% of the total mass for most LIB types), its recovery presents significant energy savings, since secondary production of aluminum saves up to 88% of the energy required during its primary production (Wang and Gaustad, 2012). With the consideration of the potential economic and environmental savings from recovering these contained materials, LIB recycling technologies must have a broader target than cobalt recovery alone (Table 1).

As LIB recycling efforts expand to target a broader spectrum of metals, the recycling technology must also be optimized to achieve higher efficiencies and selectivities of desired metals. LIBs come in different sizes, form factors, and cathode chemistries, indicating a highly co-mingled, uncertain waste stream if pre-sorting by cathode type cannot be done. A typical LIB consists of a cathode, an anode, separators and electrolyte, all of which contains a variety of metallic materials (e.g., copper, nickel, cobalt, lithium, aluminum, etc.), as well as plastics, carbon black, and organic liquids. A range of chemistries has been used as cathode materials as well, introducing further compositional uncertainty for unlabeled batteries. While lithium cobalt oxide battery dominates the market currently, LIB technology is transiting to low-cost cathode chemistries (e.g., $LiFePO_4$, $LiMn_2O_4$, and some mixed-metal cathodes). As these cathodes grow in market share, profits for recyclers will be greatly impacted (Wang et al., 2014a). Recovering cobalt alone will not make the LIB recycling process financially successful due to this transition; however, as states are already enacting landfill bans, recyclers may still need to process this mixed stream. Successful segregation of materials has the potential to enrich the constituent of targeted material(s) in a certain size fraction, which helps to improve the efficiency of subsequent recycling processes and improve the profit for recyclers. The goal of this work is to quantify the potential for material segregation via shredding and mechanical size separation for LIBs.

### 1.2. Shredding and size segregation as LIB waste management strategy

Shredding or 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 (Khoo, 2009). For LIBs specifically, while a few studies have included some type of pre-recycling steps into their proposed recycling process, the possibility of scaling these steps up may be limited and is usually not considered. For example, cutting battery cases is the first step of the laboratory-scale LIB recycling process proposed in many studies (Contestabile et al., 2001; Li et al., 2010b; Chen et al., 2011). While these authors recommend cryogenic treatment on an industrial scale according to their experimental experience (i.e., heat caused by the internal short-circuit of the cell during cutting), the feasibility of manually extracting the active materials has not been addressed for process scale-up. In (Nan et al., 2006), EOL LIBs were first dismantled to separate the outer steel cans from the contained materials using a custom dismantling machine; however, specific details on this process were not clearly presented. Li et al. used ultrasonic washing to separate cathode materials from the aluminum foils and separate carbon powder from the copper foil before the leaching process. However, their sorting process is limited by the low concentration (28% by weight) of cobalt in the targeted fraction (Li et al., 2009). Yamaji et al. proposed a novel method of under-water explosion to dissemble EOL LIBs (Yamaji et al., 2011). While this method can successfully prevent fires

**Table 1**
The price of materials (USGS, 2014).

| Metals | Prices ($/kg) | Metals | Prices ($/kg) |
|---|---|---|---|
| Cobalt | 28.44 | Aluminum | 2.09 |
| Nickel | 15.02 | Iron | 0.73 |
| Copper | 7.50 | Phosphate rock | 0.09 |
| Manganese[a] | 2.30 | Niobium | 44.00 |

[a] The price on the Infomine website (http://www.infomine.com/) (July, 2014).

during the crushing process, its associated environmental safety issues (such as the water treatment after the explosion) need to be further analyzed. These studies lay an important foundation for understanding the feasibility and potential for several pre-recycling processes such as manual cutting, ultrasonic washing, and dismantling.

However, a key challenge still remains: development of pre-recycling process that can be easily scaled up, requires low initial and operating cost, reduces energy and materials input, and at the same time can efficiently achieve material segregation. Therefore, the aim of this paper is to determine if material segregation can be achieved via pre-processing steps, particularly through shredding and sorting technologies that are frequently already in place at waste processing centers. In addition, one of the obstacles that LIB recyclers are facing is little information on LIB composition due to non-disclosed cathode chemistries and casing materials among different battery manufacturers. The effectiveness of this proposed process is examined for current market-dominate (i.e., $LiCoO_2$ cathode LIBs) as well as three future popular cathode batteries (i.e., $LiFePO_4$, $LiMn_2O_4$, and mixed-metal cathode LIBs), from perspectives on both material distribution and economic contribution.

## 2. Materials and methodology

To evaluate the efficacy of this proposed pre-recycling process when applied to batteries of differing cathode chemistries, a mixed stream of scrap LIB cells were used in this study, including 64 battery cells removed from 10 end-of-life laptop battery packs and 49 cells purchased and cycled to end-of-life; information about laptop brand and battery manufacturer for each battery pack is shown in Table 2 using indices to preserve confidentiality. These chemistries were compared to the average material content of four popular cathode chemistry types, i.e., $LiCoO_2$, $LiFePO_4$, $LiMn_2O_4$, and a mixed-metal cathode (i.e., $Li_{1.05}(Ni_{4/9}Mn_{4/9}Co_{1/9})_{0.95}O_2$), taken from the literature, manufacturers documentation, and previous work by the authors (composition provided in Table 3 with details in the supplemental material). All sample batteries used in this study are 18650[1] cells except for $LiMn_2O_4$ cathodes that are only present in 26650 cells as revealed by X-ray fluorescence.

The material flow through the proposed pre-recycling process for EOL LIBs is shown in Fig. 1. LIB packs removed from laptops were disassembled to separate the digital circuit and LIB cells. Next, LIB cells were discharged and immersed in liquid nitrogen to reduce the risk of fire and then mechanically shredded by a commercial granulator (i.e., EconoGrind 180/180[2]) into small pieces (less than 7.5 mm). To eliminate the risk of exposure to electrolyte, the shredding process was performed under a fume hood. Shredded

[1] The 18650 form indicates the battery is cylindrical, having a diameter of 18 mm, and length of 65 mm. The 26650 form indicates the batter is cylindrical, having a diameter of 26 mm and length of 65 mm.

[2] Physical parameters of Model 180/180, Economizer, USA are as following: 200 rpm rotor speed, 13.8 in. * 13.8 in. feed opening, max. 50 kg of material throughput per hour and 3 Kw drive capacity. It is assumed shear is the dominant acting force.