Recycling of spent lithium-ion battery with polyvinyl chloride by mechanochemical process

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Abstract

In the present study, cathode materials (C/LiCoO₂) of spent lithium-ion batteries (LIBs) and waste polyvinyl chloride (PVC) were co-processed via an innovative mechanochemical method, i.e. LiCoO₂/PVC/Fe was co-grinded followed by water-leaching. This procedure generated recoverable LiCl from Li by the dechlorination of PVC and also generated magnetic CoFe₄O₆ from Co. The effects of different additives (e.g. alkali metals, non-metal oxides, and zero-valent metals) on (i) the conversion rates of Li and Co and (ii) the dechlorination rate of PVC were investigated, and the reaction mechanisms were explored. It was found that the chlorine atoms in PVC were mechanochemically transformed into chloride ions that bound to the Li in LiCoO₂ to form LiCl. This resulted in reorganization of the Co and Fe crystals to form the magnetic material CoFe₄O₆. This study provides a more environmentally-friendly, economical, and straightforward approach for the recycling of spent LIBs and waste PVC compared to traditional processes.

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1. Introduction

Lithium-ion batteries (LIBs) are an attractive energy storage option due to their high energy density, high energy storage performance, long discharge period, and excellent circulation performance (Goodenough and Park, 2013). Recently, there has been a large increase in the number of spent LIBs due to the gradual upgrading of electrical and electronic products and the continued promotion of new energy automobiles. In China alone, the total quantity and weight of discarded LIBs are estimated to reach 25 billion units and 500 thousand tons, respectively by 2020 (Zeng et al., 2012b; Zeng et al., 2015a). Therefore, developing approaches to safely collect and dispose of spent LIBs has become a priority in the scientific community.

Owing to the lack of adequate policy and feasible technology, there are only limited recycling plants to recycle the spent LIBs in some countries such as China. As a consequence, the most wasted batteries including the spent LIBs entered into the approach of municipal solid waste treatment and disposal. LIBs are discarded at the end-of-life phase, and the recycling mainly rely on small workshops since lacking of specific collection system (Zeng et al., 2014). Some recyclers and scavengers only collect the most valuable parts of spent batteries and discard the rest, which not only results in a waste of resources, but also causes pollution (Chen et al., 2017). The European Union issued a battery directive in 2004 that requires all battery products to be recycled and producers and retailers to assume responsibility for the recycling and disposal of spent batteries (Directive, 2006). In China, “waste battery pollution control technology policy” has completed the document of the draft with the purpose to control the pollution prevention during the period of battery recycling and regeneration.

Prior work on recycling spent LIBs has focused on the recovery of valuable metals cobalt (Co) and lithium (Li) from the cathode materials using recovery processes such as pyrometallurgy, hydrometallurgy and biometallurgy (Zeng et al., 2014). Table 1 summarized recent works (2011–2016) focusing on the recovery of metals from spent LIBs. While these methods have been successful, there are practical limitations for their use in industry. Pyrometallurgy requires a large amount of energy and emits harmful fumes, while hydrometallurgy requires strong acids, organic acids, and expensive reagents, and produces an abundant amount of liquid waste (Ordoñez et al., 2016).

In recent years, the chlorination metallurgical process has been widely used for the recovery of metals from waste materials. Polyvinyl chloride (PVC) has been used in these processes because it contains 58% Cl by weight and is an excellent chlorine atom donor. Moreover, the disposal of waste PVC has become a major problem as the production of PVC rises steadily (Yu et al., 2016). In our previous work, a subcritical water process was proposed for the...
co-treatment of spent LIBs and waste PVC. Li and Co from C/LiCoO$_2$ powder can be successfully leached into subcritical water using PVC as an acid source. The disadvantages of this process include requirements of high temperature and pressure, generation of corrosive hydrochloric acid, and subsequent separation of Li and Co (Liu and Zhang, 2016).

Recently, many research groups have focused their efforts on the recovery of metals by mechanochemical methods (Lee et al., 2013; Yuan et al., 2012). During the milling process, mechanical forces act on reactants in order to induce changes in their structural and physiochemical properties, thereby facilitating chemical reactions (Day, 1984; James et al., 2012). Mechanochemical reactions are driven by mechanical energy rather than heat energy, thus they can occur at room temperature and atmospheric pressure. This could largely reduce the energy and cost. Moreover, the solid-solid reaction does not depend on water as a medium, hence there is no liquid waste produced by this process (Guo et al., 2016).

In this study, we developed an innovative mechanochemical method for the co-processing of cathode materials (C/LiCoO$_2$) from spent LIBs and waste PVC. Ferrous powder was used as a co-grinding reagent for the dechlorination of PVC. Li in LiCoO$_2$ was recovered by reaction with organic chloride in PVC. The residual Co was recombined with Fe and converted into magnetic material CoFe$_2$O$_4$ after calcination. The operating parameters were optimized, and the mechanochemical residues were characterized. The reaction mechanisms of the co-grinding of PVC/LiCoO$_2$ with different additives are discussed in detail.

2. Experimental

2.1. Materials and reagents

Spent LIBs were supplied by XIAMEN OASIS Sources Co., Ltd. The LIBs were completely discharged by addition of a 5 wt% NaCl.
solution and were subsequently washed with deionized water and dried. The LIBs were mechanically dismantled using pliers. The obtained cathode (C/LiCoO₂), which contained LiCoO₂, an acetylene black conductor, aluminum foil, and an organic adhesive, was dissolved in N-methylpyrrolidone to separate the adhesive PVDF. After digestion in aqua regia, the Co and Li contents of the C/LiCoO₂ powder were measured using an inductively-coupled plasma optical emission spectrometer (ICP-OES, Prodigy, Leeman, USA). Co and Li accounted for 51.8 wt% and 6.5 wt%, respectively, of the total cathode materials.

A commercially available PVC powder (Chemical Reagent Company of Beijing) was used in this study. The diameter of the PVC particles was approximately 50–100 µm. Other reagents were purchased from the Chemical Reagent Company of Beijing.

2.2. Recovery procedure

PVC and LiCoO₂ powders together with co-grinding additives were added at appropriate ratios to 100 mL ball milling pots. The pots were installed on the disk of planetary ball milling and rotated at the required rotation speed for a period of time. After grinding, the products were harvested. The milled products and milling balls were rinsed with 100 mL of deionized water and collected by vacuum filtration. After drying, the solid was calcined in a muffle at 800 °C for 2 h, and the resulting product was a magnetic material, CoFeₓOᵧ. Fig. 1 shows a flow chart for the recovery of materials by mechanochemical co-processing of spent LIBs and PVC. The conversion rates of Li, Co, and Cl were expressed as a percentage by the following formula:

\[
\text{Conversion rates rate (\%)} = \frac{C}{C_0} \times 100\%
\]

where \(C\) is the final concentrations of either Li, Co, or chloride ion in the filtrate and \(C_0\) is the original concentration of either Li, Co, or Cl from PVC in the sample before co-processing.

2.3. Analytical methods

The concentrations of metal ions in the filtrate were measured by ICP-OES (Prodigy, Leeman, USA), while the concentration of inorganic Cl⁻ was measured by IC. The crystal structures of the solid products were characterized by X-ray diffraction (XRD, Philips PW 1700) using Cu Kα radiation (\(\gamma = 1.5418\) Å) with 30 kV of voltage and 30 mA of current. Analysis of the XRD data was carried out by MDI Jade 6.0 software. The microstructures and relative contents of surface elements were analyzed on a scanning electron microscopy-energy dispersive spectrometer (SEM-EDS, JSM-7001F, Japan). The magnetic properties of the products were determined using a vibrating sample magnetometer (VSM, LakeShore-7307, USA) at room temperature.

3. Results and discussion

3.1. Effects of co-grinding reagents on the conversion rates of various elements

3.1.1. Alkaline compounds as co-grinding reagents

Fig. 2 shows the effects of alkali metallic oxides/hydroxides (CaO, NaOH and KOH) on the conversion rates of Li, Co, and Cl. When LiCoO₂ and PVC were co-grounded without additives, the conversion rates of Li, Co, and Cl were 27.5%, 1.4%, and 13%, respectively. Li has a relatively high charge density compared to Co and has a stable helium-type double shell, which makes Li more active.
than Co, suggesting that LiCl could be formed more easily than CoCl₂ during the ball milling process.

Previous work has shown that alkali oxides/hydroxides are efficient reagents for co-grinding dehalogenation (Cagnetta et al., 2016; Zhang et al., 2013). The addition of alkaline compounds such as CaO, NaOH, or KOH as co-grinding reagents increased the dechlorination rate of PVC to higher than 60%, but the conversion rates of Li and Co were not significantly increased. The Cl removed from PVC preferentially binds to alkali or alkaline earth metals M (K, Ca and Na) to form MClₓ because the ion binding force between M and Cl is stronger than that between Li and Co. Moreover, Cl removed from PVC is difficult to bind with Li or Co since lithium cobalt oxide has dense layered structure, leading to the low conversion rates of Li and Co. The foregoing indicates that alkali metal compounds are not suitable additives for the mechanochemical process.

3.1.2. Oxides as co-grinding reagents

The effects of oxides (Fe₂O₃, SiO₂, or Al₂O₃) addition on the conversion rates of Li, Co, and Cl are shown in Fig. 3. The addition of each oxide led to an increase in the conversion rates of Li, Co and Cl in LiCoO₂/PVC, but the conversion rates of Li was relatively low when co-ground with Fe₂O₃. The conversion rates of Cl increased in the following order Al₂O₃ > SiO₂ > Fe₂O₃. When Al₂O₃ was added, the mechanical force promoted the generation of friction plasma on the surface of Al₂O₃ to induce the conversion of the organic Cl in PVC into inorganic chloride ion. In the presence of SiO₂, the dechlorination was accelerated by the formation of Si—Cl and Si—O—Cl (Yuan et al., 2012). The addition of Al₂O₃ or SiO₂ enhanced the conversion rates of Li and Co by converting Cl in PVC into chloride ion. However, the conversion rates were still low, thus oxides are not ideal co-processing reagents.

3.1.3. Zero-valent metals as co-grinding reagents

Fig. 4 shows the effects of zero-valent metals (Ni, Al, and Fe) on the conversion rates of Li, Co, and Cl in the co-grinding process. The addition of Ni converted 78.5% of Li, 6.7% of Co, and 70.3% of Cl. The addition of Al converted 89.8% of Li, 38.2% of Co, and 91.5% of Cl. The addition of Fe converted 98.7% of Li, 18.5% of Co, and 92.5% of Cl. Zero-valent metals promote the dehalogenation of PVC via reductive dehalogenation, whereby they abstract a halogen from a halogen-containing organic molecule (Cagnetta et al., 2016; Ye et al., 2011). Zero-valent metals can also enhance the chlorination of Li. When PVC is dechlorinated by ball milling, the chloride binds to the zero-valent metal (Ni, Al, or Fe) to produce MClₓ. These covalent compounds bind to the Li and Co in LiCoO₂. Because Co is less active than Li, the zero-valent metals favor the formation of LiCl and exhibit a low conversion rate for Co. Compared with alkaline compounds and oxides, zero-valent metals are better additives for this mechanochemical co-grinding process. Therefore, zero-valent Fe was chosen as the optimal additive for the mechanical co-processing of LiCoO₂/PVC.

3.2. Effects of co-grinding parameters on the conversion rates of various elements

3.2.1. PVC dosage

The effects of PVC dosage on the conversion rates of Li, Co, and Cl are shown in Fig. 5. With the LiCoO₂/PVC/Fe mass ratio of 1:1:1, the conversion rates of Li, Co, and Cl were 98.7%, 18.5%, and 92.5%, respectively. As the mass ratio of PVC increased, the conversion rates of Li, Co, and Cl all significantly decreased. This could be attributed to the reduction of contact chance between PVC and the milling balls, which would eliminate PVC dechlorination effect. The decrease in dechlorination would result in a lower amount of chloride ion and, in turn, lower conversion rates of Li and Co were obtained. This experiment indicated that one equivalent of PVC to LiCoO₂ was the optimal mass ratio.
3.2.2. Fe dosage

As can be seen from Fig. 6, an increase in Fe dosage above the 1:1:1 LiCoO₂/PVC/Fe ratio could decrease Co conversion rate and dramatically increase Cl conversion rate. Adding Fe powder to the co-grinding system increased the number of dechlorination sites for the organic Cl in PVC, thus accelerated Cl conversion rate. With a LiCoO₂/PVC/Fe mass ratio of 1:1:2, the conversion rates of Li, Cl, and Co were 100%, 96.4%, and 8.1%, respectively. The optimal mass ratio of LiCoO₂/PVC/Fe for mechanochemical co-processing was found to be 1:1:2.

3.2.3. Co-grinding time

Fig. 7 shows the effect of various co-grinding time on the conversion rates of Li, Co, and Cl. All conversion rates increased with increasing co-grinding time. The conversion rates increased significantly from 71.6% to 100% for Li, from 1.2% to 8.1% for Co, and from 68.1% to 96.4% for Cl when the co-grinding time was increased from 6 h to 12 h. Longer reaction time provides more energy to the system and allows for the crystal structure of the ball milling powder to be broken down more thoroughly (Balaz et al., 2013). Therefore, prolonging the co-grinding time accelerates PVC dechlorination, which increases the conversion rates of Li and Co. The data in Fig. 7 also indicate that Li was successfully converted to LiCl, Co largely remained in the co-grinding residues, and PVC was dechlorinated. The optimal milling time was determined to be 12 h based on energy consumption and conversion rates.

The optimal co-grinding parameters for the mechanochemical co-processing of LiCoO₂/PVC/Fe were determined to be a mass ratio of 1:1:2, a co-grinding time of 12 h, and a ball-to-powder mass ratio of 50:1. Under these conditions, the conversion rates of Li, Co, and Cl were 100%, 8.1% and 96.4%, respectively. 91.9% of the Co remained in the residue and was rearranged to form the magnetic material CoFeₓOᵧ.

3.3. Characterization of the co-grinding products

3.3.1. XRD analysis

Fig. 8 shows the XRD spectra of samples (LiCoO₂/PVC/Fe = 1:1:2) (a) before and (b) after mechanochemical co-processing. The peaks corresponding to LiCoO₂ are no longer present after the mechanochemical co-processing, and the peak intensity of Fe is largely weakened (Fig. 8a and b). According to valence theory, LiCoO₂ can be considered as Li₂O and Co₂O₃. According to the ICP-OES results, in the presence of Fe, Li₂O in LiCoO₂ could
bound to chlorine in PVC to form water-soluble LiCl. In comparison, the Co-O in Co$_2$O$_3$ remained tight and stable, and 91.9% of Co remained in the residue after water-leaching. Under the co-grinding solid-solid reaction, only some of Fe entered Co$_2$O$_3$ crystals, resulting in diffraction peaks only for Co$_2$O$_3$ and Fe in the co-grinding products (Fig. 8b). After calcination of the water-leaching residues at 800 °C for 2 h, diffraction peaks corresponding to CoFe$_2$O$_4$ and α-Fe$_2$O$_3$ were observed (Fig. 8c). This indicates that Co and Fe compounds were oxidized under air and converted to CoFe$_{x}$O$_{y}$ after high-temperature calcination while the Li in LiCoO$_2$/PVC/Fe was recovered as LiCl after the water-leaching process.

3.3.2. SEM-EDS analysis

Solid products were analyzed by SEM-EDS under different operating conditions (Fig. 9). Before co-grinding, the LiCoO$_2$/PVC/Fe sample consisted of irregular PVC particles, ~100 μm in diameter, and C/LiCoO$_2$ and Fe powders scattered around the PVC grains (Fig. 9a). After co-grinding, no PVC particles could be seen in the LiCoO$_2$/PVC/Fe sample, and the obtained products were homogeneous (Fig. 9b). This indicates that after mechanochemical co-processing, the structures of the starting materials were destroyed. The calcined product CoFe$_{x}$O$_{y}$ had a uniform grain size, and there was a small degree of crystal aggregation due to the high-temperature calcination (Fig. 9c). The magnified image in Fig. 9d shows that the CoFe$_{x}$O$_{y}$ crystal formed irregular octahedrons. Elemental analysis of CoFe$_{x}$O$_{y}$ shows that C found in the water-leaching residues disappeared after calcination (Fig. 9e). The molar ratio of Co/Fe/O in the resulting CoFe$_{x}$O$_{y}$ was 1:4:6, suggesting that the product is CoFe$_4$O$_6$.

3.4. Assessment of the technical, economic, and environmental impact of different processes on spent LIBs recovery

We assessed the impact of different processes used to recover materials from spent LIBs (Table 2). Compared to traditional processes (e.g., pyrometallurgy, hydrometallurgy, and biometallurgy), the proposed mechanochemical co-processing method has several technical advantages. It can be performed at room temperature, which reduces energy consumption. In addition, the LiCoO$_2$/PVC/Fe solid-solid reaction eliminates the use of water as a medium thus no acidic wastewater was generated. The proposed mechanochemical method also has several economic advantages over other processes. While hydrometallurgy and biometallurgy require chemicals to promote the leaching of Li/Co, the mechanochemical method uses waste PVC as a chlorine donor. Moreover, this mechanochemical method has a simpler recovery procedure and significantly lower separation costs compared to traditional processes. Furthermore, this method is also more environmentally friendly. Pyrometallurgy generates toxic waste gases, and both hydrometallurgy and biometallurgy produce waste liquids (Xu et al., 2008; Zeng et al., 2014). In comparison, this mechanochemical method does not require corrosive acids or strong oxidants and does not require water as a solvent so no liquid waste is generated. The limitations of the mechanochemical method include long reaction time for optimized results (12 h) and the noise generated by the operation of ball grinder. Overall, this study proposes a mechanochemical method for the recovery of metals from spent LIBs that significantly outperforms traditional processes in terms of being more environmentally friendly, more economical, and technically easier to perform.

3.4. VSM of CoFe$_{x}$O$_{y}$

VSM was performed to obtain the magnetic hysteresis loop of the resulting magnetic material CoFe$_{x}$O$_{y}$ (Fig. 10). The experiment was conducted at room temperature. The saturation magnetization ($M_s$), residual magnetization ($M_r$), and coercivity ($H_c$) were 54.1 emu g$^{-1}$, 22.9 emu g$^{-1}$, and 934.9 Oe, respectively. These values indicate that the magnetic material CoFe$_{x}$O$_{y}$ had excellent magnetic performance.

The measured magnetic properties of CoFe$_{x}$O$_{y}$ are consistent with its crystalline structure and magnetic behavior. The high $M_s$ and $H_c$ values suggest that CoFe$_{x}$O$_{y}$ is a hard magnetic material, which is desirable for applications requiring high magnetic fields. The VSM analysis corroborates the excellent magnetic performance of CoFe$_{x}$O$_{y}$, highlighting its potential for use in magnetic devices and separations.

3.5. Assessment of the economic benefit and profit

We assessed the economic benefits of the mechanochemical co-processing method in terms of equipment investment, raw mate-
Technical, economic and environmental impact assessments of different spent LIBs recovery processes.

| Process | Pyrometallurgy | Hydrometallurgy | Biometallurgy | Mechanochemical method |
|---------|----------------|-----------------|---------------|-----------------------|
| Equipment Reagent | Pyrolysis furnace | None | None | Ball grinder |
| Temperature (°C) | 100 | 60–80 | Normal temperature | Waste PVC + Fe powder |
| Reaction time (h) | T < 1 | 2–6 | 72–336 | 12 |
| Recovery rate (%) | Li/Co > 95 | Li/Co > 95 | Li/Co > 95 | Li/Co = 100 |
| Merits | Simple, easy-to-use | Low energy consumption, high metal recovery rate | Low energy consumption, high metal leaching rate, gentle conditions | Use waste to treat waste, low cost, no water in solid-solid reaction, Li/Co separate conversion avoids subsequent separation |
| Limitations | High-temperature and high energy consumption, generation of waste gases | Generation of waste liquid; solvent extraction, ion adsorption or precipitation method is needed by subsequent Li/Co separation | Long reaction period; heavy metals are toxic to microorganisms; subsequent Li/Co separation is tedious | Medium reaction time |
| Environmental impact | Waste gases | Waste water and waste acids | Waste water | Noise |

Table 2

Economic benefit assessment of the proposed mechanochemical co-disposal process.

| Parameter | Mechnochemical co-treatment process |
|-----------|-----------------------------------|
| I | Equipment input II | Material input III | Product output |
| Equipment cost ($) | 2000 | PVC ($) | 0 | Metal case | 0.085S (250 g) |
| Power (kW) | 0.75 | Spent LIBs ($/kg) | 1.05 | Al foil | 0.14S (70 g) |
| Temperature (°C) | 25 | Fe | 1. (140 g) | Cu foil | 1.0S (150 g) |
| Time (h) | 12 | – | – | LiCl | 1.50S (18.2 g) |
| Power cost ($) | 0.69 | – | – | CoFeOx | – |
| Sum1 ($) | 0.69 | Sum2 ($) | 1.06 | Sum3 ($) | 2.82 |
| a Profit ($) | 1.07 |

a Profit ($) = Sum3 – Sum1 – Sum2.

4. Conclusions

A novel mechanochemical process was successfully established for the recycling of spent LIBs and waste PVC, where PVC was dechlorinated, Li was recovered as LiCl and Co was converted into a separable magnetic material (CoFeOx). The optimal co-grinding additive was zero-valent Fe, which yielded nearly 100% conversion of Li into water-soluble LiCl, 96.4% conversion of organic chlorine into inorganic chloride ion as a ligand, and the transformation of Co into CoFeOx. This LiClO2/PVC/Fe mechanochemical co-processing approach is more cost effective and more environmentally friendly than traditional processes, which therefore can be scaled up to reduce the environmental loads from spent LIBs recycling.

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