International Journal of Mineral Processing 169 (2017) 91–98

Contents lists available at ScienceDirect

International Journal of Mineral Processing

journal homepage: www.elsevier.com/locate/ijminpro

Recovery of valuable materials from spent lithium ion batteries using electrostatic separation

A.V.M. Silveira, M.P. Santana, E.H. Tanabe, D.A. Bertuol *

Environmental Processes Laboratory (LAPAM), Chemical Engineering Department, Universidade Federal de Santa Maria – UFSM, Avenida Roraima 1000, 97105-900 Santa Maria, RS, Brazil

A R T I C L E I N F O

Article history:
Received 9 February 2017
Received in revised form 23 October 2017
Accepted 7 November 2017
Available online 07 November 2017

Keywords:
Electrostatic separation
Lithium ion batteries
Recycling
Mechanical processing

A B S T R A C T

Continuing industrial development results in ever greater consumption of products and materials. These include electrical and electronic equipment (EEE) such as mobile phones and, consequently, lithium ion batteries (LIBs). Therefore, an efficient and environmentally friendly recycling technology is vital for the recovery of valuable materials from spent LIBs. This work describes an alternative process for the recovery of these materials, using mechanical processing and electrostatic separation. Firstly, the batteries are dismantled and their components are characterized. This is followed by comminution, drying (to remove the organic electrolyte), separation according to particle size, and electrostatic separation of the conductive and nonconductive parts of the LIBs. Parameters evaluated in the electrostatic separation were the electrode voltage, roll rotation speed, distance of the electrostatic electrode, and the inclination angle of the deflector. The results showed recovery of a conductive fraction containing 98.98% of metals and a nonconductive fraction containing 99.6% of polymers, demonstrating that electrostatic separation is a promising and efficient method for the recovery of high purity materials from spent LIBs.

1. Introduction

One of the consequences of technological development is the great demand for electrical and electronic equipment (EEE). These devices are rapidly discarded, because every day there are new models on the market, making the products become obsolete faster. The composition of this waste electrical and electronic equipment (WEEE) includes many materials that can be recovered, avoiding the unnecessary use of natural resources (Kasper et al., 2011). Cell phones are among the most significant EEEs, and their useful lifetimes have continuously decreased, with substitution now typically within three years (Kasper et al., 2011; Jing-Ying et al., 2012).

The increased demand for portable devices such as mobile phones, computers, and digital cameras directly increases the consumption of lithium ion batteries (LIBs). As a result, the disposal of spent LIBs has become an environmental concern, although this type of battery can be considered a rich source of hazardous but valuable metals with considerable commercial potential (Saeki et al., 2004; Zhang et al., 2014a).

A typical LIB consists of a cathode, an anode, separators, and electrolyte, all of which contain a variety of metallic materials as well as plastics, black carbon, and organic liquids (Zhang et al., 2014b; Wang et al., 2016). The electrolyte is lithium hexafluorophosphate ($LiPF_6$), together with an organic solvent such as ethylene carbonate (EC), diethyl carbonate (DEC), dimethyl carbonate (DMC), or their mixture. The separator is made of polypropylene (PP) or polyethylene (PE) (Gratz et al., 2014).

The anode contains graphite (the active anodic material), adhesives, and a current collector consisting of a copper foil. The cathode is composed of a lithium compound (the active cathodic material), adhesives, and an aluminium foil current collector (Zou et al., 2013).

A wide variety of lithium compounds are used as active cathodic materials, including $LiCoO_2$, $LiMn_2O_4$, $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$, and $LiFePO_4$ (Gratz et al., 2014). The most commonly used is lithium cobalt oxide ($LiCoO_2$) (Mohan et al., 2009; Mukherjee et al., 2012).

There are different technologies and processes for recycling LIBs. These methods are based on essentially two types of recycling: physical and chemical processes. Among the former, mechanical processing, magnetic separation, and heat treatment are widely employed (Xu et al., 2008; Bertuol et al., 2015; Costa et al., 2015). Mechanical separation involves the reduction and homogenization of particle size, and separation of different components according to their physical properties (Zhang et al., 2014b; Georgi-Maschler et al., 2012; Shin et al., 2005).

Chemical processes include hydrometallurgical, electrometallurgical, and pyrometallurgical techniques (Dorella and Mansur, 2007; Jha et al., 2013; Li et al., 2013; Zhang et al., 1998). Mechanical separation is usually applied as a pretreatment in the recycling of batteries, removing the external casing and releasing the metallic fraction. The metallic fraction is then subjected to a chemical process (Ito et al., 2010; Lupi et al., 2005).

Despite the advances in LIB recycling technologies, the development of efficient methods for the recovery of valuable materials, such as cobalt, copper, and aluminium, remains an important challenge for a

* Corresponding author at: Chemical Engineering Department, Federal University of Santa Maria – UFSM, Avenida Roraima 1000, 97105-900 Santa Maria, RS, Brazil.
E-mail address: daniel.bertuol@ufsm.br (D.A. Bertuol).

https://doi.org/10.1016/j.minpro.2017.11.003
0301-7516/

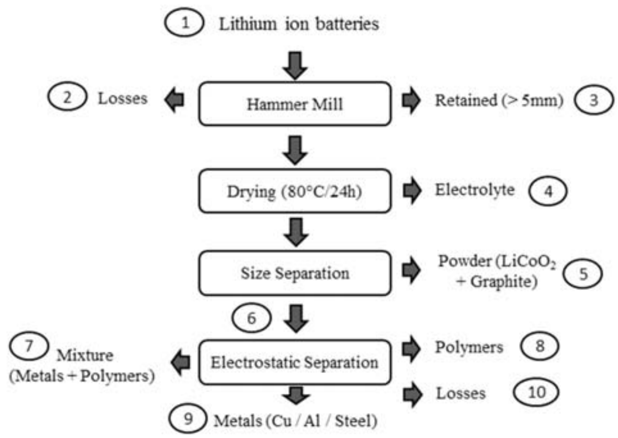

Fig. 1. Flow chart of the LIBs recycling process.

sustainable society (Costa et al., 2015). Electrostatic separation, which is based on the differences in electrical properties of materials, is a very promising process for their separation. The separation is achieved using forces acting on charged or polarized particles in an electric field. The charging of particles can be accomplished by many methods, such as conductive induction, contact electrification, and ion bombardment (Ortega-Rivas, 2012).

In the ion bombardment method, electrical charges are produced by the ionization of air resulting from the discharge generated by a corona electrode connected to a high voltage direct current source. In this equipment, the mixture to be separated is fed onto a rotating cylinder by a vibratory feeder and passes through the electric field generated between the roll electrode and the active electrodes (Ortega-Rivas, 2012; Tilmatine et al., 2009; Younes et al., 2013).

After the ion bombardment, the nonconductive fraction (NCF) remains attached to the surface of the rotating roll electrode, due to the electric image force. These materials fall into the NCF collector when the gravitational force becomes greater than the electric image force. Due to electrostatic induction, the conductive fraction (CF) acquires a charge of opposite polarity to that of the rotating roll electrode and is attracted towards the electrostatic electrode, falling into the CF collector (Ortega-Rivas, 2012; Tilmatine et al., 2009; Younes et al., 2013). According with Dascalescu et al. (2016), several factors can influence the electrostatic separation processes: shape and size of the particles, radius and speed of the roll electrode, superficial moisture and type of corona electrode (Dascalescu et al., 2016).

Electrostatic separation has been studied for the recovery of metals from WEEE (Ruan and Xu, 2016), the recycling of chopped electric wire (Medles et al., 2007), of printed circuit boards (Jiang et al., 2008; Huang et al., 2009), and the separation of different polymeric residues (Tilmatine et al., 2009; Wu et al., 2013). However, to the best of our knowledge, there have been no previous studies concerning the application of electrostatic separation for the recycling of LIBs.

The present work concerns the application of electrostatic separation for the recovery of valuable materials from spent LIBs. Firstly, different models of LIBs were dismantled and characterized. Subsequently, the main parameters of the electrostatic separation were evaluated. Finally, the best processing conditions were used to effectively recover valuable materials from three different types of LIBs.

2. Materials and methods

2.1. LIBs characterization

Three different battery brands were used to quantify the materials present in the LIBs. The batteries were first manually dismantled and classified into individual parts in order to determine the amounts of the different materials (metal, polymer, and powder fractions) present in each battery type. The components were separated using pliers and cutting saws, and were quantified in terms of mass percentages. After the manual dismantling, the materials were weighed and dried for 24 h at 80 °C to eliminate the organic electrolyte.

Characterization of the active materials adhered to the anode and cathode was performed by X-ray diffraction (Miniflex 300, Rigaku). The metals present in the batteries (external casing and electrodes) were identified by scanning electron microscopy (SEM) (VEGA-3 SBU, TESCAN) coupled with energy dispersive spectroscopy (EDS) (INCAx-act, Oxford Instruments). Characterization of the polymeric fraction was achieved using differential scanning calorimetry (DSC–60 Plus, Shimadzu).

2.2. Mechanical processing

Fig. 1 shows the sequence of operations used. In all experiments, the LIBs were first submitted to mechanical processes, followed by electrostatic separation, in order to obtain metal concentrates.

2.2.1. Comminution, sieving and drying

The spent LIBs were comminuted in a hammer mill (Tiger A4) fitted with a 5 mm output sieve. The comminuted materials (powder, polymers, and metals) were dried for 24 h at 80 °C to eliminate the organic electrolytes and were then submitted to a granulometric separation by sieving (Fig. 1). The powder fraction (which was composed of the active material of both electrodes) was separated from the metallic and polymeric fractions using a 65 Tyler sieve (0.212 mm). The remaining material with a particle size between 0.212 mm to 5.0 mm was subjected to granulometric analysis on vibrating screens (Tyler 20, 10, 7 and 5).

2.2.2. Electrostatic separation

Fig. 2 shows a schematic illustration of the roll-type electrostatic separator employed in the separation of the different materials obtained after the comminution, sieving, and drying operations. Evaluation of the electrostatic separation process was divided into two stages. In the first stage, the best parameters of the electrostatic separator were determined using a synthetic sample that simulated real samples in terms of both chemical composition and mass percentages. The purpose of using a synthetic sample was to be able to define the ideal separation conditions using materials free from contaminants such as graphite, adhesives, and $LiCoO_2$, which could affect the results. After defining the ideal conditions, new tests were performed using real samples obtained

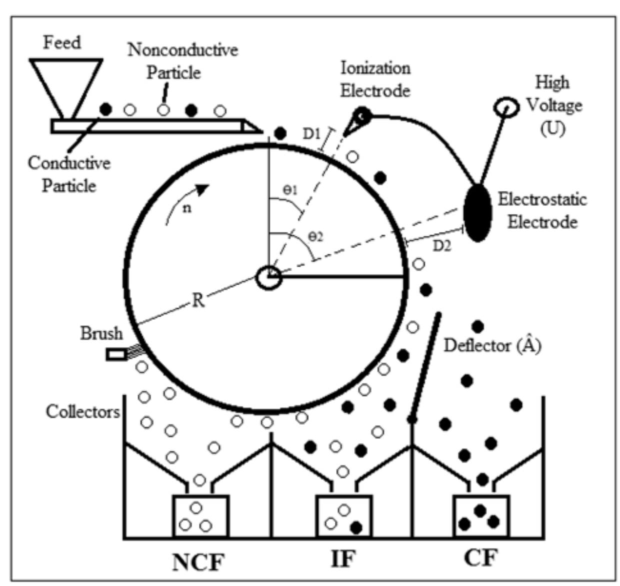

Fig. 2. Schematic illustration of the operation of the roll-type electrostatic separator.