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Environmental impact and economic assessment of secondary lead production: Comparison of main spent lead-acid battery recycling processes in China

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ABSTRACT

China is the largest lead-acid battery (LAB) consumer and recycler, but suffering from lead contamination due to the spent-lead recycling problems. This paper describes a comparative study of five typical LAB recycling processes in China by compiling data about the input materials, energy consumptions, pollution emissions, and final products. We compared the environmental impacts of these processes in six categories using the Chinese Life Cycle Database (CLCD) and analyzed their economic efficiencies using technology cost modeling (TCM) based on the local market prices of materials and energy. According to the results, we found that not all of the innovative hydrometallurgical processes are healthy alternatives. We should pay attention on indirect emissions in the environmental inspection, take account pollution treatment costs into green profit analysis, and recommend the best process with the change of the society resource supply structure.

1. Introduction

In recent decades, lead acid batteries (LAB) have been used worldwide mainly in motor vehicle start-light-ignition (SLI), traction (T Liu et al., 2015; Wu et al., 2015) and energy storage applications (Díaz-González et al., 2012). At the end of their lifecycles, spent-leads are collected and delivered to lead recycling plants where they are often repurposed into the refined lead and used as LAB materials (Gomes et al., 2011). A total of 11.3 million tons of lead metal was produced globally in 2014, and 56% of that amount was obtained from spent LAB (ILZSG, 2015; Li et al., 2016). Lead is a hazardous metal with negative impacts on human health (Tian et al., 2015a,b,c). Unfortunately, because of the outdated recycling processes in some developing countries, large amounts of lead dust, fumes, and hazardous waste are discharged, seriously affecting public health (Chen et al., 2009; Gottesfeld and Pokhrel, 2011; Uzu et al., 2011).

In the last decade, China has been the largest spent lead recycler in the world (Liu et al., 2015; Lopez et al., 2015; Tian et al., 2015a,b,c). More than 1.6 million tons of lead scrap was recycled in 2014 in China, accounting for 25% of the world total (CNMA, 2015). China is a typical representative of most developing countries, with some lead blood poisoning accidents occurring in recent years (Wu et al., 2002). More than 70% of lead recycling plants in China were closed in 2012 due to process and pollution emissions problems (Tian et al., 2014).

The standard spent-lead recycling production line includes the following procedures: 1) crash and separate the battery at ambient temperature and 2) recycle the lead paste, lead grids, plastic case, and waste acid (Blanpain et al., 2014). Traditional pyrometallurgical processes are used to recycle refined lead from lead paste; this is the primary pollution emitting unit of the entire production line (Gomes et al., 2011). Many types of recycling processes are used in more than 300 Chinese lead recycling plants (Lassin et al., 2007; Smaniotto et al., 2009; Tian et al., 2014), and some additional innovative processes have been developed at Chinese institutes (Duan et al., 2016; Ellis and Mirza, 2010; Gao et al., 2014; Li et al., 2012; Maruthamuthu et al., 2011; Sun et al., 2014).

Previous environment impact assessments (EIA) and economic

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analyses of LAB recycling have been limited (Brogaard et al., 2014; Zhang et al., 2016). For example, the lifecycle assessments (LCAs) of different end of life (EOL) scenarios for battery recycling were compared, and the environmental benefits of one type of recycling process were analyzed (Daniel and Pappis, 2008; Sun et al., 2016). However, a single process and the LCA database in Europe are not representative of the global recycling industry. While the recycling benefits of four different technologies used in the USA were also compared and the technologies that have proper pollution prevention and waste minimization were identified. These processes in the USA are not popularly adopted in China, so the research results are not direct valuable references to the China authorities (Genaidy et al., 2009). Furthermore, the consumption and contaminant species of most recycling processes in China are completely different from those in Europe and the USA, making it more difficult to identify the most environmentally friendly process. It is, therefore, necessary to perform a special environmental impact and economic assessment of the typical processes used in China (McKenna et al., 2013; Troy et al., 2016). The assessment was adopted to compare different technologies, such as lignocellulosic biomass thermochemical conversion technologies (Patel et al., 2016) and funnel glass waste management options for cathode ray tubes (CRT) in the USA (Xu et al., 2013). We also can use it for the assessment of the LAB recycling process in China.

In this study, we first compile the consumption and emission data from five lead paste recycling processes. Then, we perform the EIA using the Chinese Life Cycle Database (CLCD) and economic analysis with technical cost modeling (TCM). Finally, we discuss the results of the environmental and economic performance analyses.

2. Methods

2.1. Material and energy balances

We first selected three traditional and two innovative lead paste recycling processes for LABs, which are widely accepted in China. Most small illegal secondary lead plants in developing countries use the process A (Stevenson, 2009); The process B is commonly used in large-scale (Annual capacity $\geq$100,000 tons batteries) plants worldwide (Rabah and Barakat, 2001; Stevenson, 2009); The process C is widely adopted by primary lead smelters to produce lead from mixtures of lead paste and lead ore concentrate (Du, 2013). Two companies in Hubei and Zhejiang provinces are testing two innovative lead recycling processes are in their pilot plants. The total recycling capacity of these two companies are more than 300 k tons/year in China. The process flow diagrams are shown in Fig. 1. We then compile and compare consumption and emission data for these processes.

A) The first of the three traditional processes is the direct reverberatory furnace melting of the battery. After draining the acid and manually dismantling the batteries, the lead paste from the LABs mixing with some grids and plastic, is thrown into the reverberatory furnace to produce crude lead (Blanpain et al., 2014).
B) The second traditional process is the rotary furnace melting of the desulfurized lead paste. After automatic crashing and separation, the lead paste is desulfurized with sodium carbonate. The desulfurized paste is then reduced by iron and carbon to lead in a rotary furnace, with the fuel of natural gas and pure oxygen.
C) The third traditional process is the QSL furnace (Sohn and Olivas-Martinez, 2014). The mixtures can be melted in a furnace without desulfurization because the sulfur is collected to produce sulfuric acid. The following sections include the reduction and electrolytic.

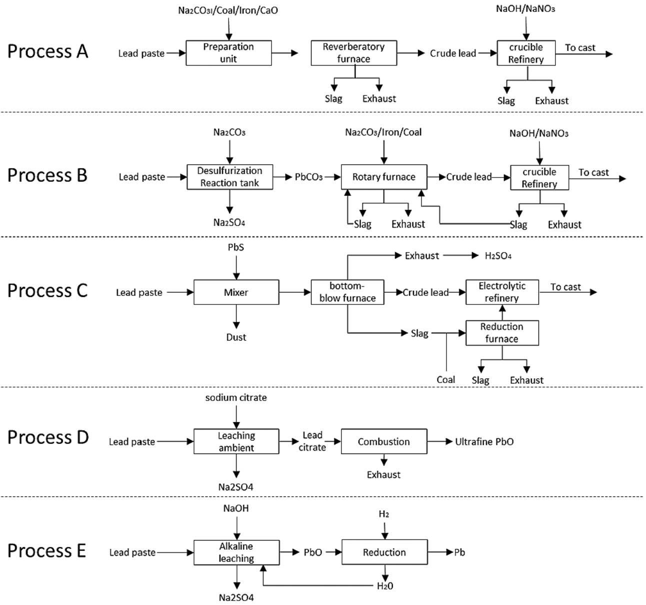

Fig. 1. Flow diagrams of the LAB lead paste recycling processes.

D) The first of the two innovative processes is the citrate leaching process (Sonmez and Kumar, 2009; Zhu et al., 2013). In this process, the lead paste is reacted with an aqueous sodium citrate and acetic acid solution, and pure lead citrate $Pb_3(C_6H_5O_7)_2 \cdot 3H_2O$ is crystallized in the leaching stage. Finally, after combustion at 350 °C, ultrafine lead oxide is produced, which does not need further refining.
E) The second innovative process is the alkaline leaching process (Pan et al., 2012, 2013). Here, PbO is prerefined via leaching in a sodium hydroxide solution. PbO is then reduced into Pb at a cathode in the presence of hydrogen gas.

For the material and energy balances analysis of each traditional process, the data are collected from the published literature for existing LAB recycling plants (Bernardes et al., 2004; Stevenson, 2009; Du, 2013). The data for the innovative hydrometallurgy processes are derived from the operational records of the pilot plants (Pan et al., 2012, 2013; Zhu et al., 2013). Some data are derived from material and energy balance calculations and private interviews.

2.2. Environmental impact assessment

2.2.1. Description of the system

This paper focuses on the direct and indirect environment impacts of spent-lead recycling processes. All consumption of materials and energy and the emissions of waste liquid, air and solids are calculated in the EIA. The direct environment impacts depend on the pollution emissions on site, but the indirect environment impacts rely on the consumption of materials and energy.

2.2.2. Functional unit

The functional unit in this study was 1000 kg of lead paste. It was recycled by the five typical processes in China. We calculate all the data for input materials, energy consumptions, pollution emissions, and final products based on this functional unit.