



## Recovery of iron and lead from a secondary lead smelter matte by magnetic separation



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### ABSTRACT

A secondary Pb smelter matte with a total Fe grade of ca. 50% (as FeS, Fe<sub>3</sub>O<sub>4</sub>, FeO and metallic Fe), shows good potential for being used as a secondary Fe source. However, requirements for Fe ore are: a total Fe grade > 60% and a sulphur content as low as possible. Therefore, further treatment steps are necessary to increase the Fe content in the matte. Dry low intensity, wet low intensity and wet high intensity magnetic separation experiments were performed on different particle size fractions of the matte. Mineral liberation analysis (MLA) was performed to explain different behaviours of Pb in the different size fractions during magnetic separation. Moreover, oxidizing roasting at 600 °C was carried out to transform FeS to Fe<sub>2</sub>O<sub>3</sub>, which is the more preferred form of Fe for pig iron production. Results show that by combining low and high intensity magnetic separation and oxidizing roasting, material with a total Fe grade of 61% (as oxide) can be recovered in the magnetic fraction representing circa 50% of the initial weight of the sample. Moreover, the sulphur content was reduced from initial 20 to 4% in the final magnetic fraction. Further reduction in sulphur content would require an additional desulphurization step.

### 1. Introduction

In 2013, the global secondary Pb production was 6.1 million tonnes, representing 54% of the total Pb production (ILA, 2017). Secondary Pb production significantly contributes to a more sustainable development as it requires only 35–40% of the energy consumed during the production of Pb from primary ores (Smaniotto et al., 2009). Pb recycling also significantly eases the pressure on mining of primary ores (ILA, 2017).

Used lead-acid batteries (ULABs) are the most significant resource for secondary Pb production, accounting for more than 85% of the total amount of secondary Pb (Zhang et al., 2016). The traditional recycling process is based on high temperature reduction of Pb compounds (sulphates, oxides) present in ULABs to metallic Pb (Zhang et al., 2016). This recycling process, however, also generates around 200,000 tpa of solid residues in Europe (Lassin et al., 2007). Examples of the chemical composition for some of these solid residues are shown in Table 1.

As can be seen from Table 1, the chemical composition of the residues varies significantly, depending on the process and raw materials being used (Gomes et al., 2011; Smaniotto et al., 2009). The most common phases present in such solid residues are: pyrrhotite (Fe<sub>1-x</sub>S),

wüstite (FeO) (Angelis et al., 2002; Gomes et al., 2011; Lassin et al., 2007) magnetite (Fe<sub>3</sub>O<sub>4</sub>) (Gomes et al., 2011; Lassin et al., 2007), galena (PbS) (Angelis et al., 2002; Lassin et al., 2007) metallic Pb and anglesite (PbSO<sub>4</sub>) (Gomes et al., 2011). Sodium is mostly present as Na<sub>2</sub>S (Gomes et al., 2011) or NaFeS<sub>2</sub> (Lassin et al., 2007).

As solid residues from secondary Pb production are often considered as hazardous wastes (Angelis et al., 2002; Smaniotto et al., 2009), they represent significant costs for companies as well as an environmental hazard for society.

So far, processing of these residues was mainly focused on reducing their quantity and toxicity (Gomes et al., 2011; Lewis and Beauteant, 2002), as well as on their recycling as a construction material (Angelis et al., 2002; Penpolcharoen, 2005).

Recently, a lot of attention has been paid to the processing of solid residues from ULABs recycling industry. Previous studies were focused on selective leaching of valuable metals (Pb, Cu, Ni, Zn) by using nitric acid (Kim et al., 2017c) and citrate solution (Kim et al., 2017a) as leaching reagents. Moreover, the effect of milling, as a pre-treatment step, on the leaching efficiencies of Fe, Pb, Cu, Ni and Zn (Kukurugya et al., 2017) and recovery of these metals through ion exchange (Kim et al., 2017b) were studied. These studies clearly showed that more

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**Table 1**  
Content of main elements in solid residues from secondary Pb production.

Element [wt%]	Source					
	Lewis and Beautement (2002)	Gomes et al. (2011)*			Angelis et al. (2002)	Penpolcharoen (2005)
		I	II	III		
Fe	22	58	51	42	41	36
Pb	9.2	1.3	1.7	9.3	17	1.3
S	7.6	7.1	6.9	7.6	11	2.6
Ca	1.3	n.a.	n.a.	n.a.	1.3	7.5
Si	n.a.	n.a.	n.a.	n.a.	3.0	11
Na	16	8.6	5.0	10	0.4	3.1

\* Three different charges into the furnace.

than 90% of Pb (as  $\text{PbSO}_4$ ) can be leached without dissolving the Fe-rich matrix material.

To the best of our knowledge processing of these solid residues to recover Fe or/and Pb by physical separation has not yet been investigated. Recovering Fe and Pb from these residues would help to save primary resources and would make a significant contribution to (near) zero-waste recycling of the investigated material (Kim et al., 2017a). According to the literature, the total Fe grade of secondary resources should be 60 to 70% with less than 10% of contaminants (e.g. sulphur, other metals, etc.) (Ezhov and Shvaljov, 2015). Pb grade in primary ores is usually 3–10% (Havlik, 2013). The Fe and Pb contents, shown in Table 1, indicate a potential of these solid residues to be used as a secondary raw materials in pig iron and Pb production. However, in order to use these residues, a concentration step must be carried out.

Magnetic separation is a possible way of recovering an Fe-rich product out of solid residues from secondary lead production, given the ferromagnetic ( $\text{FeS}$  and  $\text{Fe}_3\text{O}_4$ ) or strong paramagnetic ( $\text{FeO}$ ) properties of the iron phases in contrast to the diamagnetic properties of the lead phases ( $\text{Pb}$ ,  $\text{PbS}$  and  $\text{PbSO}_4$ ) present in these residues (Svoboda, 2004).

In general, magnetic separation can be divided into dry and wet processes. In both processes the magnetic intensity can be adjusted. The preferred magnetic separation technique depends in most cases on the particle size. Generally, above  $75\ \mu\text{m}$  both dry or wet magnetic separation can be used, while below  $75\ \mu\text{m}$  wet magnetic separation is more appropriate (Svoboda, 2004).

A combination of low and high intensity magnetic separation on a steelmaking slag has been shown to increase the Fe grade from 21 to 35% (Menad et al., 2014).

(Ma and Houser, 2014) demonstrated that high quality products with an Fe grade  $> 80\%$  can be obtained from a steelmaking slag with an initial Fe grade of 32% by using low intensity magnetic separation followed by selective screening.

In some studies, magnetic separation was used in combination with thermal treatment. For example, magnetic separation in combination with thermal reduction has been applied with success for processing of Cu and Cr slags (Kim et al., 2013; Long et al., 2015). Cu slag containing 40% Fe was processed by thermal reduction and dry low intensity magnetic separation. The final magnetic fraction had an Fe grade of 65% (Kim et al., 2013). Furthermore, a Cr slag with initial Fe and Cr grades of respectively 31.5% and 5.9% was processed by reduction roasting and magnetic separation to obtain a Cr-Fe rich powder of 73% Fe and 14% Cr (Long et al., 2015).

The current study aims to concentrate Fe and Pb present in a secondary Pb smelter matte by magnetic separation and oxidizing roasting to levels that Pb can be easily leached and the final Fe-rich residue fulfils the acceptance criteria for use as a secondary source in pig iron production. Furthermore, a rigorous study of the mineral liberation was carried out to interpret the results of the magnetic separation.

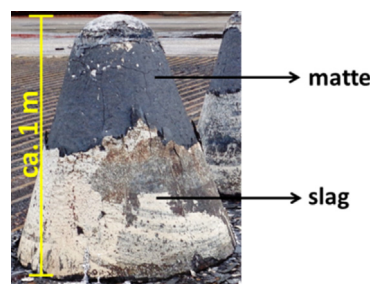


Fig. 1. Layers of the matte and the slag as recovered from the slag pot.

## 2. Experimental

### 2.1. Material

The material used in this study was provided by a company recycling ULABs. The recycling process is based on the reduction of Pb compounds present in ULABs to metallic Pb at temperatures higher than  $1000\ ^\circ\text{C}$ . During the smelting process, three layers with different densities are formed in the furnace. The bottom layer consists of metallic Pb that is separated and further refined. The other two layers, a matte (middle layer) and a slag (top layer), are tapped into a “slag pot”. After cooling, the slag pot is turned over and the matte and slag (Fig. 1) are separated physically with a jaw pulverizer.

Part of the slag (fraction  $> 15\ \text{cm}$ ) is recycled in the smelting process as a partial replacement of fluxes. The remaining fraction of the slag ( $< 15\ \text{cm}$ ) and matte are landfilled or processed by a third party, which incurs high costs.

The Fe and Pb grades of the slag ( $\sim 37\%$  Fe, 3% Pb) and the matte ( $\sim 50\%$  Fe, 6% Pb) clearly show that the matte has a higher potential to be used as a secondary source in Fe and Pb production. Moreover, the amount of the annual matte production (e.g. ca. 120 kt/y in Europe) during ULABs recycling is higher than that of the slag (e.g. ca. 80 kt/y in Europe), with a weight ratio matte:slag of ca. 1.5. Thus, the matte was selected as the input material for this investigation.

### 2.2. Methods

#### 2.2.1. Sampling and sample pre-treatment

A representative sample of the matte was collected on site by taking a total of 16 individual scoops (4 L each) at different locations across the material heap (estimated volume  $40\ \text{m}^3$ ). The samples were transported to the laboratory in 20 L buckets (each containing 4 scoops) and subsequently mixed.

This mixed sample containing big pieces ( $> 150\ \text{mm}$ ), was first crushed using a jaw crusher (Model BB300, Retsch) to a particle size below 40 mm. In a second step, the sample was crushed to a particle size below 2 mm by a laboratory jaw crusher (Model BB200, Retsch). A representative sample of 5 kg was taken for further experiments. The sample was sieved using a vibratory sieve shaker (Model AS200, Retsch). After sieving six size fractions were obtained: 1–2 mm, 0.5–1 mm, 250–500  $\mu\text{m}$ , 125–250  $\mu\text{m}$ , 63–125  $\mu\text{m}$  and  $< 63\ \mu\text{m}$ . The chemical composition of each size fraction was analyzed by handheld XRF (on powders of  $< 125\ \mu\text{m}$ ).

Additional grain size reductions of the samples for analytical or experimental purposes was performed using a planetary ball mill (Model PM400, Retsch), with a 250 ml grinding jar at 400 rpm for 5 min. Five stainless steel milling balls, with diameter of 28.3 mm, were used as a grinding medium.

#### 2.2.2. Characterization of the sample

A handheld XRF analyzer Niton XL3t GOLDD+, placed in a mobile test stand, was used for elemental analysis. The XRF analyzer is equipped with a Ag anode (50 kV and 0.2 mA). Measuring time was 120

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