



Developing a sustainable solution for recycling electric arc furnace dust via organic acid leaching



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ABSTRACT

The current study demonstrates and proposes a new method for sustainable treatment of electric arc furnace (EAF) dust. The raw material is one of the largest and fast growing hazardous waste fractions produced. Globally, annual EAF production of steel generates more than 12 M tonnes of dust today. The dust is rich in Fe but also contains toxic and volatile metals. The main components in the dust include Fe, Mn, Zn, Pb, and Cr. However, Zn and Pb in the dust inhibits its recycling back to the steel process. The novel process approach employs alkaline roasting with NaOH at 450 °C, followed by organic acid leaching using 0.8 M citric acid at 40 °C under oxygen purging, for two hours. Zinc ferrite ($ZnFe_2O_4$), which is the main Zn carrier in the EAF dust is highly refractory making selective Zn vs. Fe leaching impracticable. Roasting was observed to transform EAF dust $ZnFe_2O_4$ mineralogy into favourable form of sodium zincate (Na_2ZnO_2) and sodium ferrate ($NaFeO_2$), the former being an easily leachable Zn compound and the latter keeping Fe in solids. Under the optimized conditions, selective leaching of 100% of Zn vs. Fe (< 10%) could be conducted, in addition $\geq 80\%$ of Pb could be leached. Consequently, Fe and Zn rich recyclable secondary fractions were produced from hazardous EAF dust, both fractions being suitable raw materials for primary metal production.

1. Introduction

Recycling of steel from end-of-life products saves the world from its fast depleting natural resources. Currently, more than 600 M tonnes of annual global steel production is from secondary raw materials (Holappa, 2017). The dominating technology for production of steel from scrap is the Electric Arc Furnace (EAF) (Oustadakis et al., 2010; Havlik et al., 2005; Jha et al., 2001; Herrero et al., 2010). However, production of steel using the EAF generates up to 20 kg of dust per tonne of steel (Dutra et al., 2006). This large amount of dust is classified a pollutant to the environment if disposed for landfilling (Pelino et al., 2002; Council, 2003). Classification of EAF dust as a pollutant is based on the content and toxicity of metals such as chromium (Cr) and lead (Pb) present (Council, 2003; Ruiz et al., 2007). On the other hand, this dust cannot be recycled directly into the steel process (electric arc furnace) because it contains Zn, which negatively affects the furnace refractory and hence compromises the economics of the furnace operation (Dutra et al., 2006). Although Cr (Petrilli and De Flora, 1977; Vutukuru, 2005) and Pb (Needleman, 2004) are not problematic for the steel process itself and are often used as additives in the steel industry, they are toxicants and are thus strongly regulated by the European Commission (EC) (Council, 2003; European Commission, 2002) as EU

legislation regulates both feed and product of EAF (Council, 2003). Other metals present in large amounts such as iron (Fe) and manganese (Mn) can freely be recycled without causing any challenges into current operations (Havlik et al., 2005). The other species (alkalis) K, Mg, Na, and Ca behave as slag formers in the EAF process and as such are fully recyclable into the process. Equally, aluminium and silicon are suitable fluxing agents, hence tolerable in EAF feed (Aromaa et al., 2013). Furthermore, carbon (originating from the corrosion of EAF electrodes) is also present in the EAF dust in large proportion (> 1%), but it can be easily removed by the process of decarburization when fed into the steel process (Aromaa et al., 2013).

The EC restrictions limiting the use of EAF dust as a secondary metal source necessitate the development of a process for treating furnace dust to facilitate recycling. Whilst landfilling is still an option at present in many parts of the world, developed countries have strict regulation that makes it a non-lucrative option for the future (Council, 2003; European Commission, 2002). Therefore, an EAF dust treatment process must be able to remove Zn from the dust while leaving the valuable Fe and steel compatible metals into the residue. In addition, the Pb and Cr concentrations in the Fe-rich solids need to be kept within the limits of the EU regulations, 0.01% (European Commission, 2002), if the product is shipped elsewhere. Moreover, the usage of such material in

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the steel plant itself is not so strictly regulated by EU, which will encourage the recycling of valuable secondary raw material.

Selective leaching of metals is seen as one of the most lucrative solutions to solving the challenges related to EAF dust recycling (Havlik et al., 2005; Halli et al., 2017; Hamuyuni et al., 2018). The twofold benefit of this method is that dissolved metals Zn and Pb can separately be recovered from the solution as products while at the same time the Fe rich residue can be used as a secondary raw material for EAF process.

Inorganic acids such as sulphuric, nitric, and hydrochloric acids have previously been intensively investigated for selective dissolution of metals from EAF flue dust (Montenegro et al., 2016, 2013; Cruells et al., 1992; Caravaca et al., 1994; Langová et al., 2009). Of the three inorganic acids, sulphuric acid is the most extensively studied and promising option (Oustadakis et al., 2010; Havlik et al., 2005; Montenegro et al., 2016, 2013; Cruells et al., 1992). While positive results have been observed, most inorganic (mineral) acids would come at a large cost. Nitric and hydrochloric acids for example, would require water-intensive washing of nitrates and chlorides, respectively, to avoid these species ending up to furnace (Halli et al., 2017). On the other hand, organic acids, such as citric acid, are highly selective for Zn (Halli et al., 2017), whilst containing elements fully compatible with EAF. However, there are no previous observations (Halli et al., 2017) in which satisfactory Zn extractions (> 80%) from EAF dust by organic acid leaching. In order to make the dust suitable as feed material for steel plant, Zn content need to be decreased to $\leq 10\%$. The main cause of the insufficient Zn extractions is the existence of zinc ferrite, $ZnFe_2O_4$, (or with isomorphously substituted metals i.e. $(Zn_x, Me_y)Fe_2O_4$), from which Zn is not fully leachable in organic acid (Havlik et al., 2005).

In this study, a possibility to use direct citric acid leaching is investigated in order to turn the hazardous waste into recyclable residue for steel production.

2. Experimental

2.1. Materials

The raw material (EAF dust) employed in this study was obtained from Ovako Imatra Oy steel plant in Finland. Chemical composition of this industrial material is depicted in Table 1. The table shows that Fe, Zn, Pb, Mn, and Cr, which form the basis of this study, are among the species present in larger proportions. Preliminary characterization

Table 1

Chemical analysis of the main metallic elements within the investigated EAF dust sample (total leaching, AAS, ICP-OES, S/C Analyzer) and monthly average EAF dust composition of the plant (provided by Ovako Imatra Oy).

Element	The raw EAF dust sample [wt.%]	The roasted EAF dust sample [wt.%]	Monthly average of EAF dust at plant [wt.%]
Cr	0.23	0.19	0.38
Mn	2.52	1.63	2.01
Fe	17.89	11.4	21.27
Zn	33.16	20.9	33.43
Pb	1.64	1.35	1.50
Al	0.36	0.27	0.40
Na	1.75	14.6	2.50
Mg	0.64	0.548	0.73
Si	0.83	1.22	1.77
K	2.39	2.15	1.93
Ca	3.59	3.1	2.78
V	0.02	0.02	0.01
Ni	0.03	0.02	0.04
Cu	0.2	0.13	0.17
C	1.17	0.97	1.37
P	0.02	0.05	0.06
S	1.05	0.86	1.02
Others	32.51	40.59	28.63

showed that Fe was present mainly as Fe_3O_4 and $ZnFe_2O_4$, Zn also as ZnO, Pb as PbO, Mn as MnO and Cr as Cr_2O_3 . The other species include alkalis; K, Mg, Na, and Ca. Aluminium, silicon and carbon were also present in significant amounts. The raw material particle size was < 80 μm , mean size was 1.3 μm and $p_{80} = 4.03 \mu m$. Anhydrous citric acid, $\leq 99.5\%$ assay (VWR Chemicals, Belgium), was employed for all the leaching experiments. Sodium hydroxide powder, NaOH, 99.9% (VWR Chemicals, Belgium), was used for alkaline roasting experiments. The choice of citric acid and sodium hydroxide as leaching media and decomposing agent, respectively, is already validated in recent studies (Aromaa et al., 2013; Halli et al., 2017).

2.2. Apparatuses

A portable X-ray Fluorescence (XRF) (Geological Survey of Finland, GTK), was used to monitor subsampling homogeneity of the EAF dust sample. Particle size distribution was measured using a Mastersizer 3000 (Malvern Instruments Ltd.) A Scandia oven, (Type K4/PDI 40) was employed for thermal roasting of samples. For chemical analysis of solutions, AAS (atomic absorption spectroscopy, Varian AA240, USA), and ICP-OES (inductively coupled plasma, optical emission spectroscopy, Perkin Elmer Optima 7100 DV, USA), were employed in the study. Complete carbon combustion was monitored using an Eltra CS-580 analyser (Germany). Dried residues were analysed using XRD (X-ray Diffraction), X'Pert PRO Powder, NL. A 1000 mL lab scale reactor and a LAUDA Aqualine AL 25 (for maintaining constant temperature) were used to conduct the leaching experiment. Mixing was done by using a four blade lab agitator stirrer. For pH and Redox potential measurement, a Digital Glass Body pH Electrode HI 11,310 (Hanna Instruments) and an InLab Ag/AgCl 3 M KCl (Mettler Toledo) respectively, were employed.

2.3. Procedures

Prior to conducting experiments, the EAF dust was homogenized so as to obtain a uniform and representative sample for analysis. The homogenization procedure was done by Geological Survey of Finland (GTK). During the procedure, a sample of ca. 60 L in volume was spread, mixed and dispersed into a uniformly thick layer from which, 5 L subsamples were collected by systematically scooping equal volumes of material from all areas. Particle size distribution analysis was performed on dry samples under 0.35 MPa pressure.

A summary of roasting and leaching parameters applied is presented in Table 2. During the roasting, a magnesia crucible containing a sample was placed in an AISI 316 stainless steel reactor and hermetically closed before placing it in the furnace. Temperature accuracy of the furnace's hot zone, where the sample was placed, is $\pm 2^\circ C$. Roasting was done at $450^\circ C$ in air for an hour. For leaching experiments, solid to liquid (S:L) ratio was kept constant at 100 g/L. Experiments were conducted for a maximum time of 120 min.

During the leaching process, the solution was agitated at 500 RPM with continuous oxygen purging at 2000 mL per minute. Samples were

Table 2

Parameters utilized in the EAF dust roasting (°) and leaching (**) experiments.

Varied parameters	Constant parameters
	Roasting temperature; $450^\circ C^\circ$
	Roasting time 60 min [*]
	EAF dust: NaOH ratio (2:1 g) [*]
Temperature; 30, 40, $50^\circ C^{**}$	Stirrer rotation speed; 500 RPM ^{**}
Citric acid concentration; 0.05, 0.1, 0.2, 0.4, 0.8 M ^{**}	Oxygen flow; 2 000 mL/min ^{**}
	Solid to liquid ratio; 100 g/L ^{**}
	Sampling Interval; 5, 15, 30, 60, 120 min ^{**}

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