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Recovery of metal-doped zinc ferrite from zinc-containing electric arc furnace dust: Process development and examination of elemental migration

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ABSTRACT

We herein report elemental migration during the hydrothermal treatment of zinc-containing electric arc furnace dust (Zn-containing EAFD). Initially, the Zn-containing EAFD was characterized and the crystalline phases of Zn, lead (Pb), and chromium (Cr) were determined to be mainly in the form of (Zn,Mn)Fe_2O_4, PbO ($PbSO_4$), and Cr_2O_3 (CrO_3/CrO_4^{2-}), respectively. Subsequently, the elemental migration and transformation behavior during the Zn hydrothermal extraction process using ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$) was studied in detail. Results indicated that under optimum leaching conditions, Zn, Pb, calcium (Ca), and manganese (Mn) were extracted with high leaching efficiencies of 97.4%, 98.3%, 93.4%, and 97.5%, respectively. Furthermore, suitable adjustment of the leaching solution pH resulted in the simultaneous precipitation of Fe, Zn, Mn, and Pb ions in the form of the stable spinel ferrite MFe_2O_4 (M: Zn, Mn, and Pb), which was recovered following calcination of the precipitate at 1000 °C for 2 h. Meanwhile, the majority of Fe, total Cr, and silicon (Si) existed as Fe_2O_3, Cr_2O_3, and SiO_2, respectively, in the leaching residue, which can be recycled in the steel mill.

1. Introduction

Zinc-containing electric arc furnace dust (Zn-containing EAFD) is classified as hazardous waste according to the US Environmental Protection Agency (Hagni et al., 1991), as it contains various heavy and hazardous metals such as lead (Pb) and chromium (Cr), among others. An efficient treatment method to reduce the environmental impact of Zn-containing EAFD is therefore necessary. In recent years, the extraction of valuable metals (e.g., Zn and iron (Fe)) from EAFD has received increasing attention from metallurgists (Dutra et al., 2006; Havlik et al., 2005; Shawabkeh, 2010), and as such, the crystalline phases of elements present in EAFD must be understood in detail prior to investigating their behavior during the extraction process. Although a number of studies have investigated the chemical composition and mineralogical phases of Zn-containing EAFD (Machado et al., 2006; Martins et al., 2008), the majority discussed the most abundant phases (i.e., content >3%), while hazardous elements such as Pb and Cr, which were present in smaller quantities, received little attention.

Interestingly, as the Zn content in Zn-containing EAFD samples obtained from China is relatively low (i.e., 3–17 wt.%) (Wang et al., 1998; She et al., 2009) compared to that in the EAFD from other countries, hydrometallurgical processes such as acid leaching (Leclerc et al., 2003; Oustadakis et al., 2010; Wang et al., 2016) are more suitable for the treatment of Zn-containing EAFD. Although Leclerc et al. reported Zn extraction from synthesized Zn ferrites and EAFD using $FeCl_3 \cdot 6H_2O$ via a hydrothermal process, no studies have discussed the subsequent separation of Zn from the leaching solution. Indeed, we also previously reported the extraction of Zn from pure Zn ferrite and Zn-containing EAFD using $FeCl_3 \cdot 6H_2O$, where Zn and Fe separation was achieved by adjusting the pH of the leaching solution according to the precipitation pH values of the corresponding metal hydroxides. This complex separation process was conducted in sequence, and as such, the loss of valuable elements was inevitable. As mentioned above, the majority of studies have focused on the Zn extraction efficiency from EAFD, with little information being available regarding the migration behavior of hazardous elements during the Zn extraction process (Laforest and Duchesne, 2006; Leclerc et al., 2002). Furthermore, following extraction of such toxic elements with Zn, the leaching solution must be treated appropriately to decrease its toxicity and meet the discharge criterion (Sebag et al., 2009). Moreover, if Zn metal is to be obtained directly from the leaching solution, the hazardous elements must be removed in the form of precipitates or others during purification of the Zn-containing solutions (Herrero et al., 2010; Tsakiridis et al., 2010). It is therefore necessary to determine the behavior of metals such as Zn, Pb, and Cr in the Zn hydrometallurgical extraction process to provide guidance regarding the purification and detoxification of leaching solutions and residues.

Due to various systems of operation and different scrap grades obtained from steel mills, the chemical compositions and mineral phases

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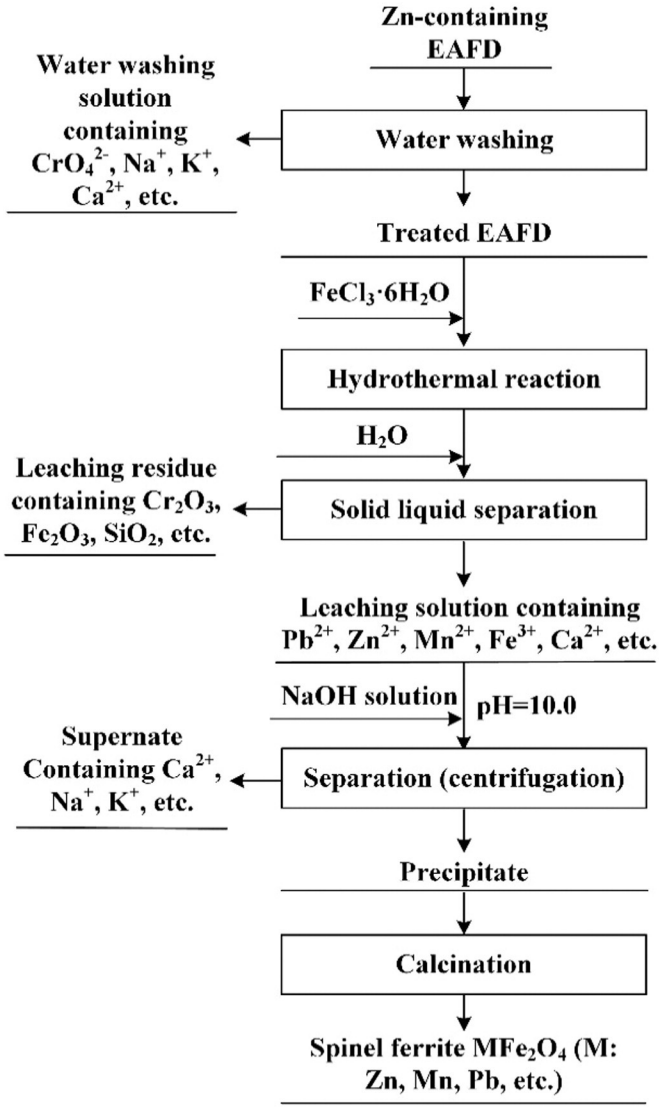

Fig. 1. Flow chart representation of Zn extraction from Zn-containing EAFD.

of the Zn-containing EAFD can vary immensely (Langová and Matýsek, 2010; Sofilic et al., 2004). We therefore present a detailed characterization of Zn-containing EAFD provided by the Tianjin Pipe (Group) Corporation, giving particular attention to the crystalline phases of Zn, Pb, and Cr. We also report the development of a simple hydrothermal process to extract Zn from the treated (i.e., water-washed) EAFD using hexahydrate ferric chloride ($FeCl_3 \cdot 6H_2O$) as a decomposition agent. The effects of the mass ratio of solid $FeCl_3 \cdot 6H_2O$ to treated EAFD ($R_{F/TE}$, g/g), hydrothermal temperature and time on the process of Zn extraction and the migration behavior of Zn, Pb, Cr, Mn, Fe, etc., were studied in detail. In addition, the leaching solution was employed to synthesize metal-doped Zn ferrite MFe_2O_4 (M: Zn, Mn, and Pb) from the extracted valuable metals. We expect that the methods described herein could contribute to the facile treatment and highly efficient utilization of Zn-containing EAFD on an industrial scale.

2. Experimental

2.1. Materials

Analytical reagent grade $FeCl_3 \cdot 6H_2O$, hydrochloric acid (HCl) (36–38%), solid sodium hydroxide (NaOH) and deionized water were purchased from the Beijing Reagent Factory, China. The Zn-containing EAFD used in this study was supplied by the Tianjin Pipe (Group) Corporation.

2.2. Experimental procedure

A flow diagram representing the extraction of Zn from Zn-containing EAFD is provided in Fig. 1.

2.2.1. Pretreatment of the Zn-containing EAFD

Pretreatment of the Zn-containing EAFD was performed by putting aqueous or HCl solutions of various initial concentrations (0.1, 0.3, 0.5, 0.7, 1.0 and 1.5 mol/L) and EAFD in a 250 mL beaker with the desired liquid-solid ratio (100/10, mL/g) of solution/EAFD, and stirring at 1000 rpm for 10 h at 25 °C. These mixtures were used in subsequent steps to provide the desired washing solutions and residues.

2.2.2. Zn extraction process

$FeCl_3 \cdot 6H_2O$ (7 g) was mixed with the desired amount of treated EAFD (5, 7, or 10 g), and the mixture was transferred into a 100 mL Teflon inner kettle, which was placed inside a stainless steel autoclave. The above mixture was then heated in a drying oven at 200, 210, or 220 °C for the desired time (6, 8, 10, 12, 16, or 20 h). After this time, deionized water (100 mL) was added to the obtained product and the resulting mixture electromagnetically stirred at 1000 rpm for 10 min at 25 °C. A solid–liquid separation was then conducted in an RJ-TDL-50A centrifuge (Ruijiang Analysis Instrument Co., LTD, Wuxi, China) at 4500 rpm for 5 min. Subsequently, the leaching solutions containing the various metals were transferred into volumetric flasks and diluted to 500 mL with deionized water. The leaching residues dried at 105 °C for 24 h. All experiments were conducted in triplicate to ensure repeatability.

2.2.3. Synthesis of spinel ferrite MFe_2O_4

2 g/L $FeCl_3$ solution (50 mL) was added to 100 mL leaching solution to give a Fe:M molar ratio of 2:1 (M: Zn, Pb, Mn, etc.). The pH value of the resulting solution was then adjusted to 10.0 using a solution of NaOH (1.0 mol/L). Following solid–liquid separation by centrifugation (4500 rpm, 5 min, 25 °C), the obtained precipitate was washed several times with deionized water, then dried and calcined at 1000 °C for 2 h to give the pure spinel ferrite, MFe_2O_4.

2.3. Analysis and characterization

Determination of the chemical compositions of the Zn-containing EAFD, treated EAFD, and leaching residue was carried out by X-ray fluorescence (XRF-1800, Japan). In addition, the phase structures of the Zn-containing EAFD, treated EAFD, leaching residue, and spinel ferrites were determined by X-ray diffraction (XRD, Japan, Rigaku) and scanning electron microscopy (SEM, Germany, Zeiss) combined with energy dispersive spectroscopy (EDS). The valences of Pb and Cr were determined by X-ray photoelectron spectroscopy (XPS) using an AXIS Ultra DLD spectrometer equipped with monochromated Al Kα radiation

Table 1
Elemental compositions of Zn-containing EAFD and treated EAFD.

Element	Fe	Zn	Ca	Si	Na	K	Cl	S	Mn	Pb	Cr
Zn-containing EAFD (wt.%)	37.29	7.79	5.33	2.21	2.78	3.11	2.63	0.70	1.18	1.16	0.15
Treated EAFD (wt.%)	42.46	8.61	5.55	2.73	0.918	0.779	0.0878	0.182	1.34	1.26	0.17