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Elution of zinc in dust discharged from electric arc furnace in carbonic acid solution

S Yokoyama¹, T Sasaki², J Sasano¹ and M Izaki¹

¹Department of Mechanical Engineering, Toyohashi University of Technology, Hibarigaoka, Tempaku-cho, Toyohashi, Aichi, 441-8105, Japan

² Graduate Program of Mechanical Engineering for Master's Degree, Toyohashi University of Technology, Hibarigaoka, Tempaku-cho, Toyohashi, Aichi, 441-8105, Japan

E-mail: yokoyama@me.tut.ac.jp

Abstract. The dust discharged from an electric arc furnace (EAF) is a valuable resource of zinc. As a fundamental study of extraction of zinc, iron and chlorine in the EAF dust, the elution behavior of them in carbonic acid solution was studied. The influence of the weight of the EAF dust on the elution behavior was examined in this study. Experiment was carried out putting the EAF dust from 1 g to 200 g in weight into 1 L of water that was introduced by CO₂. Generally, the pH in the aqueous solution increased with an increase in weight of the additive EAF dust. Maximums of the eluted concentrations of zinc and chloride ion increased with an increase in the weight of the additive EAF dust whereas the extraction ratios of both of them decreased with an increase in the weight of the additive EAF dust. Iron in the EAF dust remained in the dust without elution. The limit of extraction of zinc from the EAF dust to water was given by the solubilities of ZnFe₂O₄ and ZnO expressed by eq. (6) and eq. (9) respectively.

1. Introduction

Zinc coated steel is a superior materials economically to prevent rusting and galvanic corrosion. It has been widely used as a material for buildings, vehicles, electronics and so force. Therefore, iron and steel scrap contains zinc in itself. Steelmaking in electric arc furnace (EAF) uses steel scrap as the primary raw material. Because zinc is a volatile metal, zinc was collected as dust from off-gas in an EAF steelmaking process. Ninety two percent of zinc that is used in Japan is imported. Therefore, EAF dust is a valuable resource of zinc. Because it is very important to recover zinc from industrial wastes, numerous researches have been performed until now [1-11]. Zinc is one of the environmentally regulated substances. Then, EAF dust cannot be disposed easily. Whereas zinc has been already recovered from EAF dust, it is desired to develop of the recovery process of zinc with higher efficiency. In this work, as a fundamental study of the recovery of zinc from EAF dust, we studied the elution behaviour of EAF dust, especially zinc, iron and chlorine, in carbonic acid solution.

2. Experiment

2.1. Dust

The dust that was discharged from an electronic arc furnace (EAF) was used in this experiment. Table 1 shows the chemical compositions of the dust. The compositions were measured with an EPMA, an

X-ray fluorescence and a combustion analysis. Figure 1 shows the XRD pattern of the dust. The Fe_3O_4 , ZnFe_2O_4 , CaCO_3 , SiO_2 , KCl and C were identified in the used EAF dust. However, the metallic particle of stainless steel was observed in the EAF dust. In addition, as described later, ZnO that was not detected by an XRD existed in the EAF dust. Here, the XRD patterns of Fe_3O_4 and ZnFe_2O_4 were almost the same each other. However, there are slight differences in the peak at around 60 degree (2θ) between them. It was judged from the detection of these peaks around 60 degrees and the chemical composition that both Fe_3O_4 and ZnFe_2O_4 existed. The EAF dust of 1 g to 200 g in weight was used in the experiment.

Table 1. Chemical compositions of used EAF dust.

Element	Fe	Cr	Ca	Zn	Si	Mn	Na	Mg	Ni	K	Cu	Pb	Al	O	C	Cl
mass%	32.1	9.2	7.8	7.2	6.4	3.6	3.6	1.7	2.8	1.5	0.7	0.4	0.3	15.3	2.6	4.8

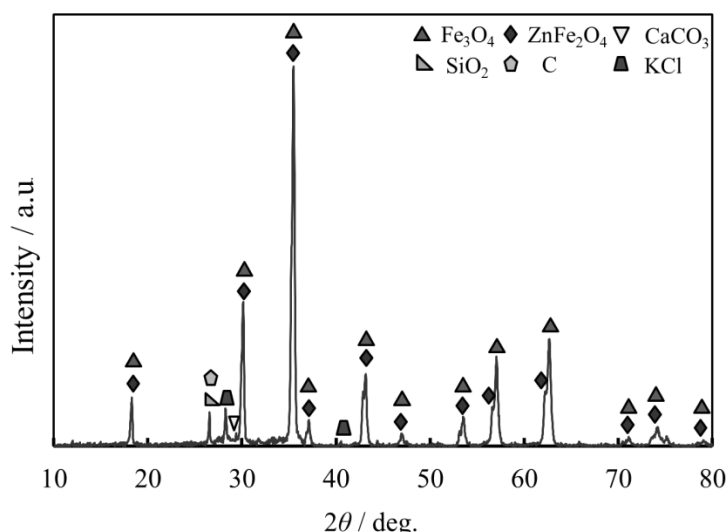


Figure 1. XRD pattern of used EAF dust.

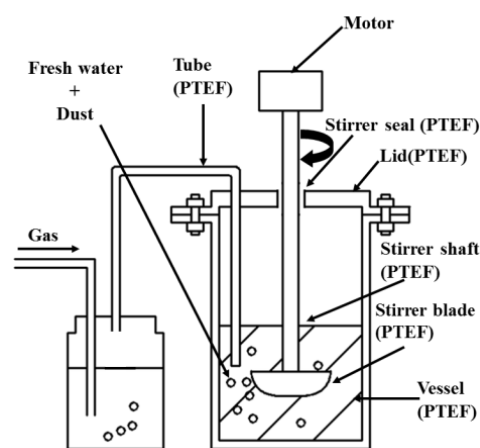


Figure 2. Schematic of used experimental apparatus.

2.2. Procedure

Figure 2 shows the schematic of the experimental apparatus. The experimental apparatus and procedure were as similar as those described in the previous paper [12, 13]. However, in order to prevent the aqueous solution with EAF dust from vaporizing, CO_2 gas was passed through pure water prior to introduction of gas into the aqueous solution. This treatment diminish made amount of water due to vaporization a one twentieth. The parts that contact with the aqueous solution are made of PTFE. Commercial grade CO_2 was used in the experiment. The apparatus was set in a simple globe box that was made of vinyl sheet to prevent interfusion of suspended dust in the air into the vessel. First, the pure water of 1 L in volume was poured into the vessel. Subsequently, CO_2 gas was introduced into the water at the flow rate of 100 mL/min (NTP) with mixing by the blade at the rotation speed of 200 rpm. After the hydrogen ion potential (pH) became stable, the EAF dust of weight of 0.1-200 g was put in the vessel. During experiment, most EAF dust was suspended in the aqueous solution, whereas a few of the dust was laid at the bottom of the vessel. The pH of the aqueous solution in the container was measured by a pH meter with glass electrode. The aqueous solution that was sucked by a whole pipette was passed through the filter of which pore size was 0.45 μm . The concentrations of iron and zinc in the filtrate were measured with an inductively-coupled

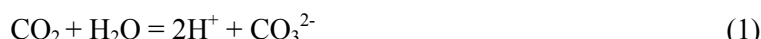
plasma atomic emission spectroscopy (ICP-AES), and the chloride ion in the filtrate was measured with a chloride ion meter using a solid membrane.

3. Results and discussions

3.1. Change in pH with time

Figure 3 shows the change of pH in the aqueous solution with time. Generally, the pH increased steeply immediately after the addition of the EAF dust. Subsequently, it gradually increased. In addition, the pH increased with an increase in weight of the additive FAF dust.

In this study, CO₂ was introduced into the aqueous solution. Accordingly, following dissolution reaction of CO₂ occurred:



These dissolution reactions of CO₂ into the aqueous solution generate H⁺, and decrease the pH. In addition, adsorption of H⁺ to the surface hydroxyl, -OH, of the EAF dust is thought to occur [14]. This adsorption reaction is given by



This adsorption reaction increases the pH. Furthermore, as mentioned later, the dissolutions of substances of the EAF dust into water decrease the pH. The elution of CO₂ into the aqueous solution, the adsorption of H⁺ and the dissolutions of substances of the EAF determined the behavior of pH. The increase in weight of the additive EAF dust implies the increase in the interfacial area of EAF dust and solution. Therefore, pH is influenced by the interfacial area. The pH is mainly determined by adsorption of H⁺ to the surface hydroxyl of the FAF dust.

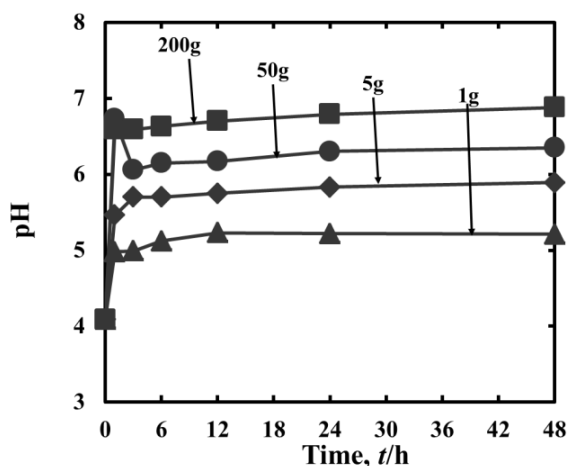


Figure 3. Change in pH in aqueous solution with time.

3.2. Elution behavior

3.2.1. Chlorine

Figure 4 shows the elution behaviour of chlorine from the EAF dust into the aqueous solution. Generally, the eluted concentrations of chloride ion increased steeply at an early stage of the elution. Subsequently it gradually increased with time. Figure 5 shows the relation between the eluted concentration of chloride ion at the end of experiment and the weight of the additive EAF dust. Whereas the eluted concentration of zinc increased with an increase in the weight of the additive EAF dust, it did not increase proportionally with an increase in the weight of the additive EAF dust.

3.2.2. Iron

Figure 6 shows the elution behavior of iron. The eluted concentrations of iron were below approximately 0.27 mg/L. The influence of the weight of the additive EAF dust on the elution behavior of iron was not observed. Maximums of the eluted concentrations of iron for the additive EAF dust of 5 and 50 g in weight were almost the same each other, and were higher than those for the additive EAF dust of 1 and 200 g in weight.

Generally, the solubilities of FeO and Fe(OH)_2 are larger than those of Fe_2O_3 and Fe(OH)_3 [12]. As mentioned earlier, the used EAF dust contains Fe_3O_4 and metallic stainless steel. Iron ion may be eluted as a Fe^{2+} . Following two things are considered as the reason that the eluted concentration iron is small. One was that the hexavalent chromium ion, which was observed in the aqueous solution, oxidized Fe^{2+} . Another was the formation of FeCO_3 due to the reaction between Fe^{2+} and CO_3^{2-} . The relation between maximum of the eluted concentration of iron and the pH was as similar as the solubility of iron carbonate. Further study was needed for clarify the reason.

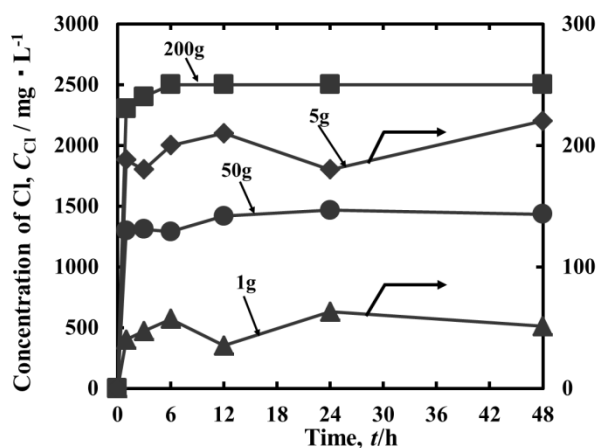


Figure 4. Change in eluted concentration of chloride ion with time.

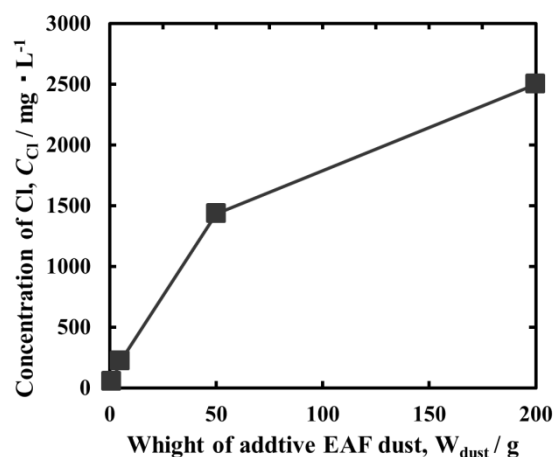


Figure 5. Relation between eluted concentration of chloride ion and weight of additive EAF dust. The concentration of chloride ion is the concentration at the elution time of 48 h.

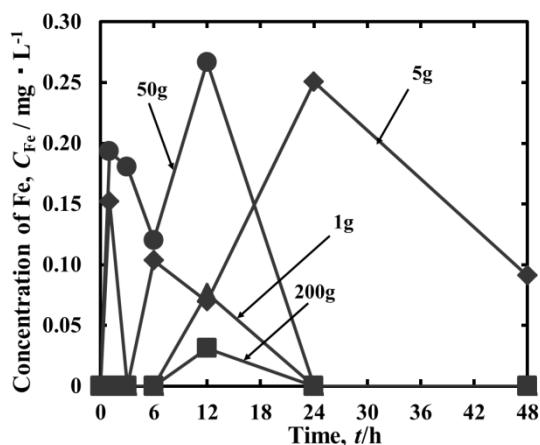


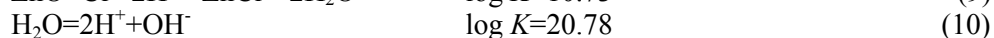
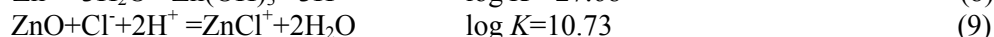
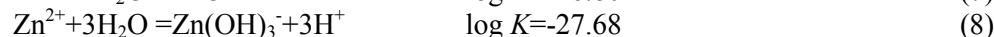
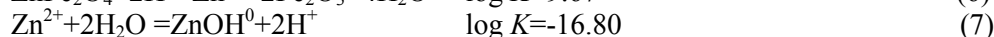
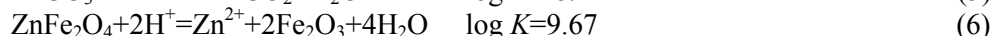
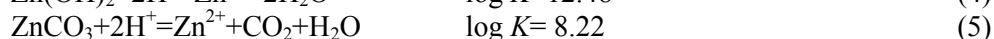
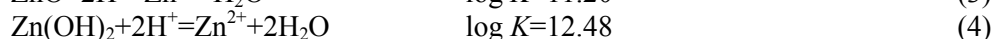
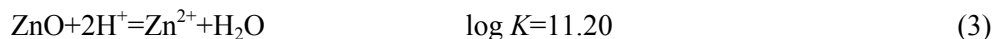
Figure 6. Change in eluted concentration of iron with time.

3.2.3. Zinc

Figure 7 shows the elution behavior of zinc. Generally, the eluted concentration of zinc increased steeply at an early stage of the elution. Subsequently, in the case of the additive EAF dust of 50 g and

200 g, the eluted concentrations of zinc decreased with time. In the case of the additive EAF dust of 1 g and 5 g, the eluted concentrations of zinc were unchanged with time.

Figure 8 shows the solubility of ZnO, Zn(OH)₂, ZnCO₃ and ZnFe₂O₄. Here, the partial pressure of CO₂ was 1 atm. The each solubility was calculated with both Debye-Hückel theory and the combination of the following thermodynamic data [15].



where K is the equilibrium constant. In addition, the relation between the eluted concentration of Zn and pH, which were obtained experimentally, was also shown in figure 8.

In the case of the weight of the additive EAF dust of 200 g, the eluted concentrations of zinc ranged between the solubility of ZnO given by eq. (3) and ZnFe₂O₄ given by eq. (6), and approached the solubility of ZnFe₂O₄. This result means that the used EAF dust contains ZnO which is not detected by an XRD. In the case of the weight of the additive EAF dust of 50 g, the eluted concentrations of zinc were below the solubility of ZnFe₂O₄ and approached the solubility of ZnFe₂O₄. These two results means that ZnFe₂O₄ forms by the reaction between Zn²⁺ eluted from ZnO and Fe₃O₄. In the case of the weight of the additive EAF dust of 1 g and 5 g, the eluted concentrations of zinc approached the solubility of ZnO which is given by eq. (9) according as the concentration of chloride ion. Accordingly, eluted concentration of zinc is influenced by concentration of chloride ion. However, the solubility of ZnO given by eq. (9) was as similar as that of ZnCO₃ given by eq. (5). Further study is needed for the elution reaction of dust into aqueous solution.

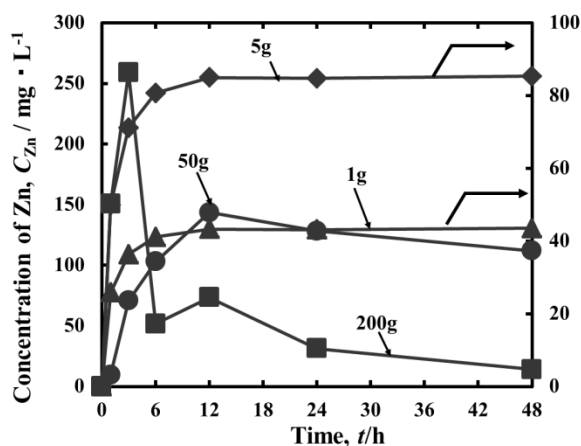


Figure 7. Change in eluted concentration of zinc with time.

3.3. Extraction ratio

Table 2 shows the extraction ratio in zinc, iron and chlorine from EAF dust to aqueous solution. Here, the extraction ratio of the substance is the ratio of extracted weight of the substance in aqueous solution and weight of the substance in the additive dust before the extraction. In addition, maximums of the eluted concentrations were used for calculation of the extraction ratio. The extraction ratios of chlorine and zinc decreased with an increase in the weight of the additive EAF dust. The extraction ratio of iron was nearly equal to zero, and was not influenced by the additive weight of the EAF dust.

Accordingly, if a small amount of the EAF dust is put in carbonic acid solution, zinc and chlorine in the EAF dust are extracted from the dust into carbonic acid solution. In addition, it was found that iron in the dust remained in the dust.

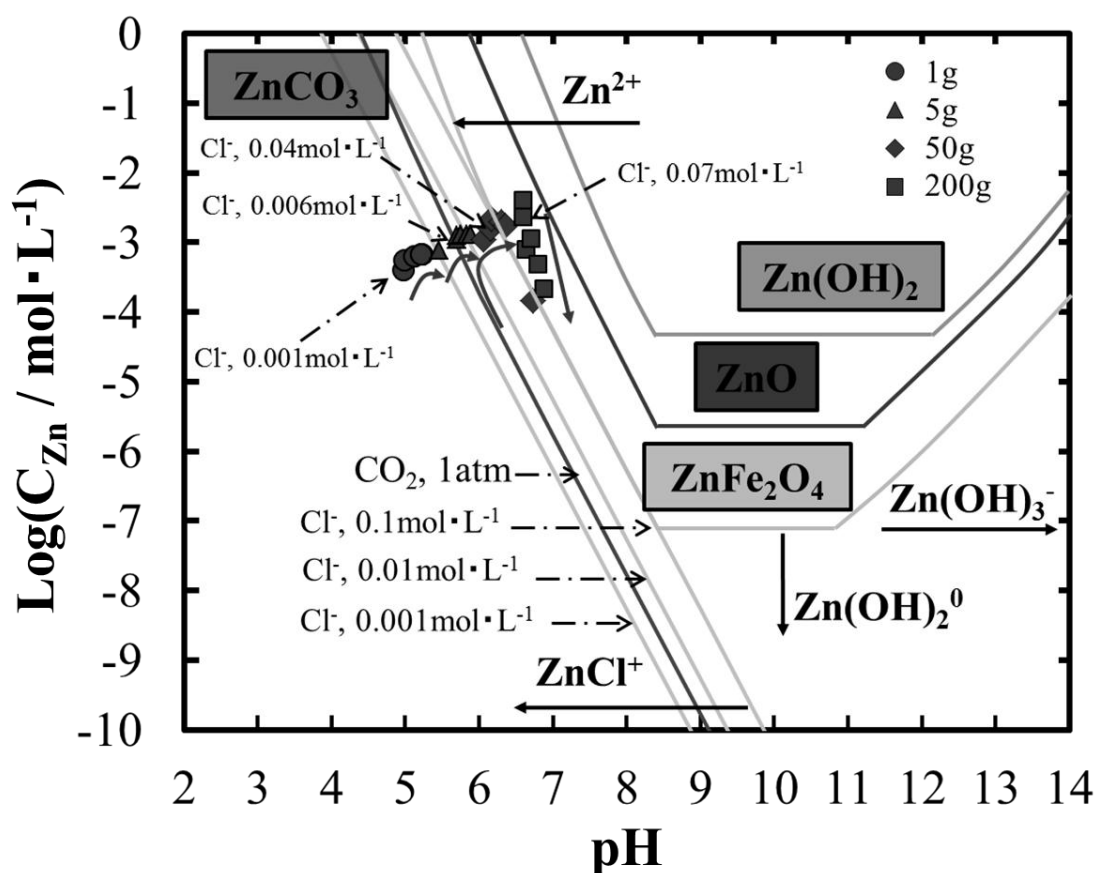


Figure 8. Solubility of ZnO, Zn(OH)₂ and ZnFe₂O₄.

Table 2. Extraction ratio in zinc, iron and chlorine from EAF dust to aqueous solution. [%]

	Weight of additive EAF dust [g]			
	1	5	50	200
Zn	60	24	4.0	1.8
Cl	100	94	62	26
Fe	0.02	0.02	0.002	0.00005

4. Conclusions

We studied the influence of weight of the additive EAF dust on elution behavior of the EAF dust into water. The obtained main results can be summarized as follows:

- (1) The pH in the aqueous solution increased with an increase in weight of the additive EAF dust.
- (2) Maximums of the eluted concentrations of zinc and chloride ion increased with an increase in the weight of the additive EAF dust. However, extraction ratios of both of them decreased with an increase in the weight of the additive EAF dust.

(3) Iron in the EAF dust remained in the dust without elution.

(4) The limit of extraction of zinc from the EAF dust to water was given by the solubility of eq. (6) and eq. (9) according as the experimental conditions. The eluted behavior of zinc was influenced by the pH and the concentration of zinc in the solution.

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