

Hydrometallurgical processing of carbon steel EAF dust

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Abstract

In this study, the hydrometallurgical processing of electric arc furnace (EAF) steelmaking dust is investigated on a laboratory scale under normal temperature and pressure conditions. The behaviour of zinc and iron under the influence of sulphuric acid as the leaching agent is discussed. The dependence between the temperature and acid concentration is investigated. The main aim is the transfer of zinc into the solution while iron ought to remain as a solid residue.

The hydrometallurgical recovery of zinc from EAF dust is feasible with relatively high recovery yield, while iron mostly remains in the solid phase. It results from the use of sulphuric acid in low concentration. This way, it is possible to set up the conditions for the EAF dust leaching, adjusting sulphuric acid concentration in order to achieve an optimum zinc yield to the solution without iron dissolution. However, the problem is that the chemical and mineralogical composition of each steelmaking dust is individual.

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1. Introduction

During the production of steel from scrap in electric arc furnaces, a considerable degree of dust is recovered in gas cleaning units. Iron oxides are the main component of this dust. By smelting of scrap, also considerable amounts of volatile components are transferred into the dust. These are zinc, lead, cadmium and other metallic compounds. Zinc and iron contents in EAF dust is in wide range according to various authors as shown in Table 1 [1–19].

The content of other components in EAF steelmaking dust is shown in Table 2.

Such amounts of admixtures are unacceptable for simple recycling to iron or steel production and at the same time these amounts of heavy metals define the EAF dust as hazardous waste. That is why it is necessary to submit this material to further treatment.

Zinc is present in EAF dust as franklinite, $ZnFe_2O_4$, or franklinite with isomorphously substituted metals, $(Zn_x, Me_y)Fe_2O_4$, where $Me = Mn, Co, Ni, Cr, Ca$, etc., and zincite, ZnO . Iron is present, except of franklinite, mostly as magnetite, Fe_3O_4 .

There are three possible processes to manage EAF dust: hydrometallurgical, pyrometallurgical and hybrid in nature. They consist in extraction of nonferrous metals, such as zinc or lead, in order to recycle dust in steelmaking industry.

Pyrometallurgical methods require some reducing agents and relatively high temperatures to produce raw zinc oxide of low commercial value. Most of these installations are based on rotary Waelz kiln technology [20,21], which works in a large-scale and therefore the dust must be collected from numerous sources and transported to the relatively large processing plant. Plasma-based [22] treatment processes are currently developed, and they are custom-designed for the capacity of specific steelmaking location. Several pyrometallurgical processes are currently at commercial level, like Enviroplas (South Africa), Allmet (USA), Metwool (USA), and Ausmelt (Australia) [17].

The treating of EAF dust, directly in the primary hydrometallurgical zinc production, has the advantage of low energy consumption, which has led to various process developments [23]. In

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Table 1
Zinc and iron contents in EAF dust according to various authors

Zn amount (wt.%)	Fe amount (wt.%)	Reference
18.54	45	[1]
19.4	24.6	[2]
23	30	[3]
13–22	11–45	[4]
2–46	10–45	[5]
34.6	42.5	[6]
20	30	[7]
22.3–24.35	30.37–35.3	[8]
39	22.1	[9]
15.9–26.7	32.3–37.7	[10]
21.1	31.25	[11]
20	30	[12]
15–25	31–40	[13]
13.6	29.8	[14]
21.8–32	19.3–25.5	[15]
18.5	29.1	[16]
15–25	30–45	[17]
21.3	21.3	[18]
20.9	27.8	[19]
22.14	30.8	Average value

alkaline leaching processes as Amax or Cebedau [24,25] heavy metals like zinc, lead and other are leached in alkaline media whereas iron is not. In acidic processes, the EAF dust is leached by acetic acid [18,26], sulphuric acid [27,28] or hydrochloric acid [29,30]. Several pilot plants were built to test hydrometallurgical technologies such as the Ezinex process [31].

An excellent overview of present status of EAF dust treatment is reported by Zunkel [17]. Hydrometallurgical methods of EAF dust treatment are discussed in [27].

The form how zinc is found is the biggest problem of its release. The ZnO does not cause any problems to neither alkaline nor acid leaching. However, the majority of zinc is present as zinc ferrite (franklinite) and it is considerably refractory against leaching. Alkaline leaching seems to be advantageous because heavy metals are leached whereas iron is inert. These methods, however, require relatively concentrated leaching medium, regardless of the fact that zinc present in franklinite is hardly accessible to leaching and that is why an intermediate thermal treatment is requested.

Acid leaching needs not so concentrated solution as the alkaline one, but iron is partially transferred into solution in this

Table 2
Other components amount in EAF dust

Element	Amount (wt.%)
Pb	0.5–2
Cd	0.1–0.3
Ca	1–7
Cu	0.01–0.2
Al	0.1–1
Mg	0.1–3
Na	0.1–1
Si	0.1–2
Cl	0.5–3
F	0.05–0.1

Table 3
Chemical analysis of EAF dust from Gerdau S.A.

Element	Amount (mass %)
Zn	33
Fe	26.5
Pb	2.17
Mn	2.3
Ca	0.9
Cu	0.2
Cr	0.2
Ni	0.1
C	3.99
S	2.69
O ₂	16
Insoluble rest	8.0

case. The franklinite is considerably refractory in this case, too.

Except of operational cost, used leaching medium also decides what method of acid leaching will be suitable – relatively cheap acetic acid [18], cheap and available sulphuric acid [27,28], or aggressive chloride medium [30,31].

Dissolved metals from leaching medium are obtained from what is a relatively cheap process, when the sulphuric acid is used for the leaching. The chloride solutions treatment is fairly complicated. The additional cost from iron removal is on record in both cases. Obtained solid product can be dumped as harmless waste or it can be recycled into primary iron or steel production.

The most suitable is, however, the process in which the heavy non-ferrous metals would be transferred into solution by a cheap method, whereas the iron remains in solid rest. These metals should be obtained from the leaching solution by simple method. The purged solution would be recycled in a leaching step. Such idea is also the aim of this work.

2. Experimental

2.1. Material

The EAF dust sample that was used was given by Gerdau SA (Siderúrgica Riograndense), Brazil, and is originated from smelting of carbon steel scrap. The following values of the chemical analysis for some elements (Table 3) were obtained.

The amount of 3.7% of soluble components was recorded in the sample. Particle size distribution made by CILAS 1180 Particle Size Analyzer was as given in Table 4.

The sample of EAF dust is also examined by an X-ray diffraction qualitative phase analysis (Fig. 1). The results are shown in Fig. 1, from which the presence of compounds such as franklinite Zn_2FeO_4 , zincite ZnO , and magnetite Fe_3O_4 can be inferred.

Table 4
Particle size distribution

Diameter	(μm)
Diameter at 10%	0.21
Diameter at 50%	0.85
Diameter at 90%	8.07
Mean diameter	2.83

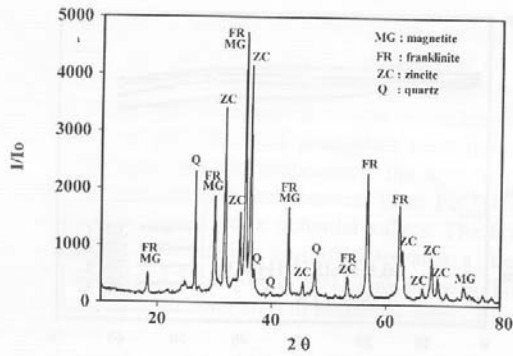


Fig. 1. XRD pattern of the Gerdau S.A. sample.

Other phases, which, on the basis of the chemical analysis, could be present, (Table 1), are probably below the detection limit.

3. Experimental set-up and procedure

Leaching experiments were performed in the apparatus and the project is shown in Fig. 2.

The leaching experiments were performed in a glass reactor of 800 ml provided with a cap in the central hole in which the axis of a glass stirrer for adjustable revolutions is placed (300 rev/min in each experiment). Openings for a mercury thermometer, a

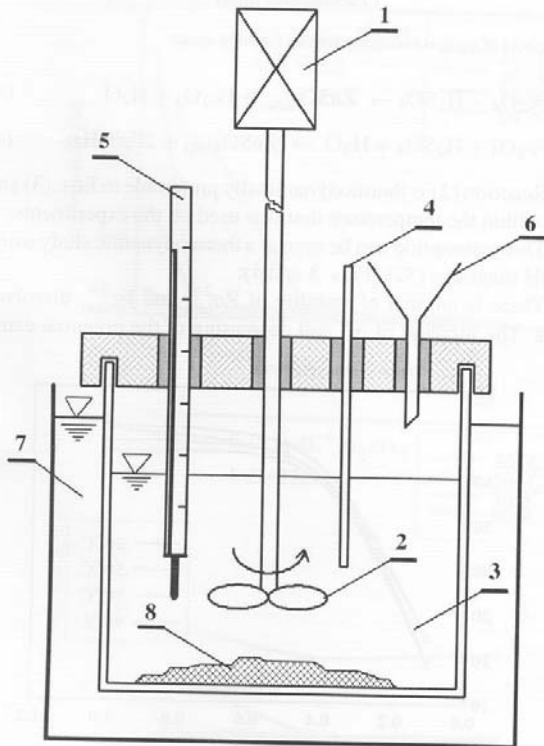


Fig. 2. Schematic view of the leaching apparatus. 1 – stirrer engine; 2 – propeller; 3 – pulp; 4 – sampler; 5 – thermometer; 6 – feeder; 7 – water thermostat; 8 – EAF sample.

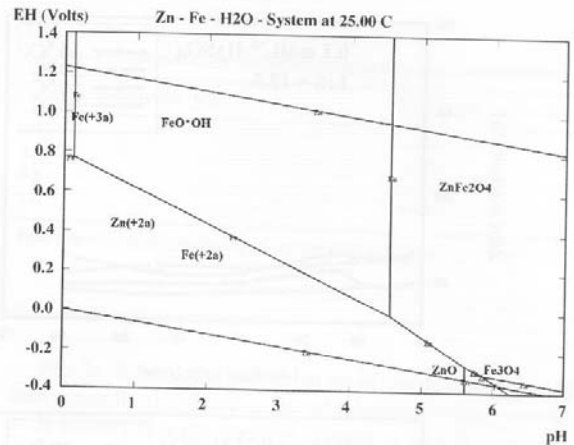


Fig. 3. E-pH diagram of the system Zn–Fe–S–H₂O at 25 °C.

sampling liquid specimen, and an input of the powdered sample were situated in cap. The reactor was placed in a water bath controlled by a thermostat. This allowed the conducting of the leaching at the desired temperature. Leaching tests were performed with a solution of 500 ml of 0.1–1.0 mol L⁻¹ H₂SO₄. The sample weight was 20 and 40 g, respectively. A solution with an acid-to-EAF dust ratio of 12.5 and 25 was used. The temperatures being used were 30, 50, 70, and 90 °C, under normal atmospheric pressure.

The samples for the chemical analysis were taken according to a fixed-schedule after 2, 10, 15, 30, 45 and 60 min. The samples were then filtered and inserted into test tubes. A clear solution was obtained. The solution was colourless, weakly yellow or weakly emerald depending on the leaching conditions. In some samples, a white precipitate of calcium sulphate had appeared after some time.

The chemical analysis, carried out in order to determine Zn and Fe, was made by means of AAS method. No other compounds in the rest solutions were traced as well as no chemical analysis of solid leaching rests were made. All results were recalculated because of the change of the pulp volume due to the sampling and evaporation.

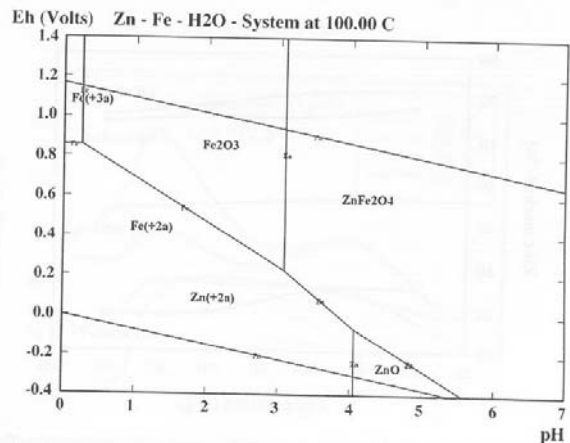


Fig. 4. E-pH diagram of the system Zn–Fe–S–H₂O at 100 °C.

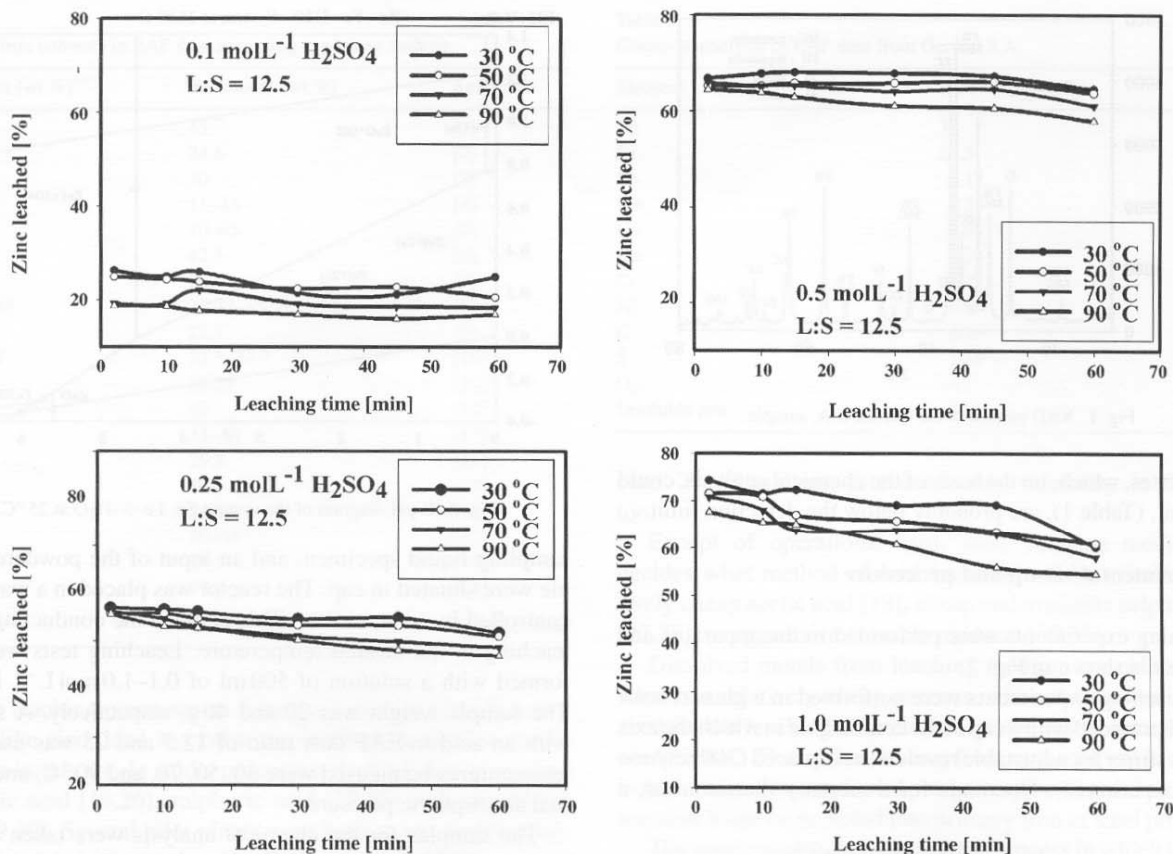
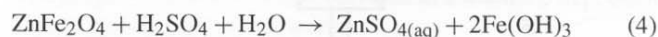
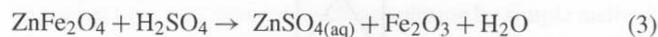
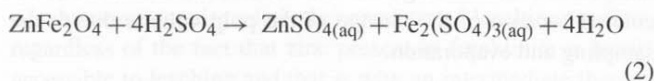
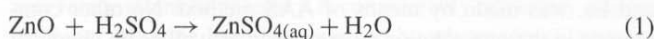


Fig. 5. (a)–(d). Extraction of zinc from EAF dust using sulphuric acid of various concentrations as leaching agent.

4. Theoretical

The reactions of the main species occurring in the sample and their stoichiometry can be stated as follows:



Reaction (2) is thermodynamically preferable to Eqs. (3) and (4) within the temperature that was used on the experiments.

This assumption can be seen at a thermodynamic study using E-pH diagrams [32] (Figs. 3 and 4):

There is an area of stability of Zn_(aq)²⁺ and Fe_(aq)²⁺ dissolved ions. The increase of pH and decreasing of the potential cause

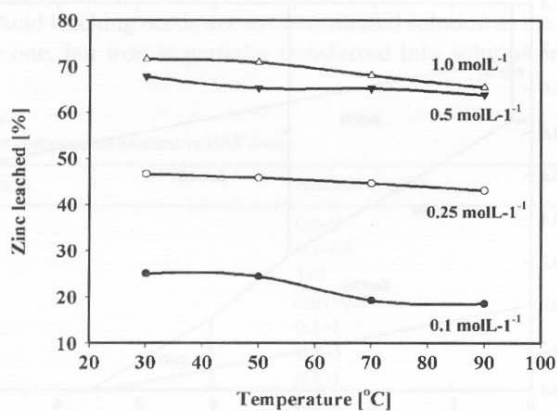


Fig. 6. Extraction of zinc depending on temperature and acid concentration after 10 min of leaching.

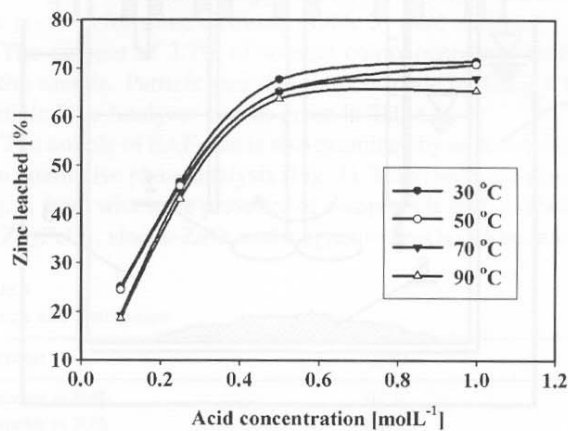


Fig. 7. Extraction of zinc depending on acid concentration after 10 min of leaching.

the precipitation of ferrous ion whereas zinc remains in the solution. In very acidic area ($\text{pH} \sim 0$) exists also an area of ferric stability ions. These ions participate on leaching as an oxidative agent. After consumption of acid by leaching reaction and increase of pH, they will precipitate from the solution as FeOOH . Increasing the temperature, the area of stability of $\text{Fe}_{(\text{aq})}^{3+}$ is enlarged and the potential limit $\text{Fe}_{(\text{aq})}^{2+}/\text{Fe}_{(\text{aq})}^{3+}$ is shifted towards higher redox potential values. The boundary limit of $\text{Zn}_{(\text{aq})}^{2+}$ precipitation is shifted towards a lower pH value, from $\text{pH}_{25} 4.55$ to $\text{pH}_{100} 3.91$. This value is, however, still sufficiently high compared to boundary limit of iron ions precipitation.

The thermodynamic study shows the possibility of transfer of zinc into solution, whereas iron remains in the solid rest by pH control. Of course, also other components of EAF dust will react, but for rough estimation the following consideration could be used.

Forty grams of EAF sample containing 13.2 g of zinc were leached for total reacting of this zinc amount, 11.12 mL of concentrated H_2SO_4 are theoretically necessary. 500 mol L^{-1} of acid solution was the volume used for each experiment. Different acid concentrations were used, as shown in Table 5.

It follows from the thermodynamic presumption, that the acid will react preferentially with zinc and that is why the iron will precipitate from the solution.

Table 5

Amount of sulphuric acid in the solution depending on its concentration

H_2SO_4 concentration (M)	H_2SO_4 amount (mL)
0.1	2.76
0.25	6.88
0.5	13.77
1.0	27.55

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5. Leaching experiments

Fig. 5a–d shows the kinetic curves of zinc leaching from EAF dust depending on temperature and sulphuric acid concentration.

It follows that the leaching process is very fast and practically immediately after starting all amount of zinc from EAF dust is leached at the given conditions. Even though the amount of extracted zinc slightly reduced with time, what is probably related to the effort of the system to achieve the dynamical equilibrium as follows from the thermodynamic study.

The concentration of acid influences the leaching of zinc preferentially, whereas the temperature only marginally affects the process. It can be seen, however, that the increase of temperature at the individual concentrations reduces the amount of leached zinc. The differences are rather small, although by

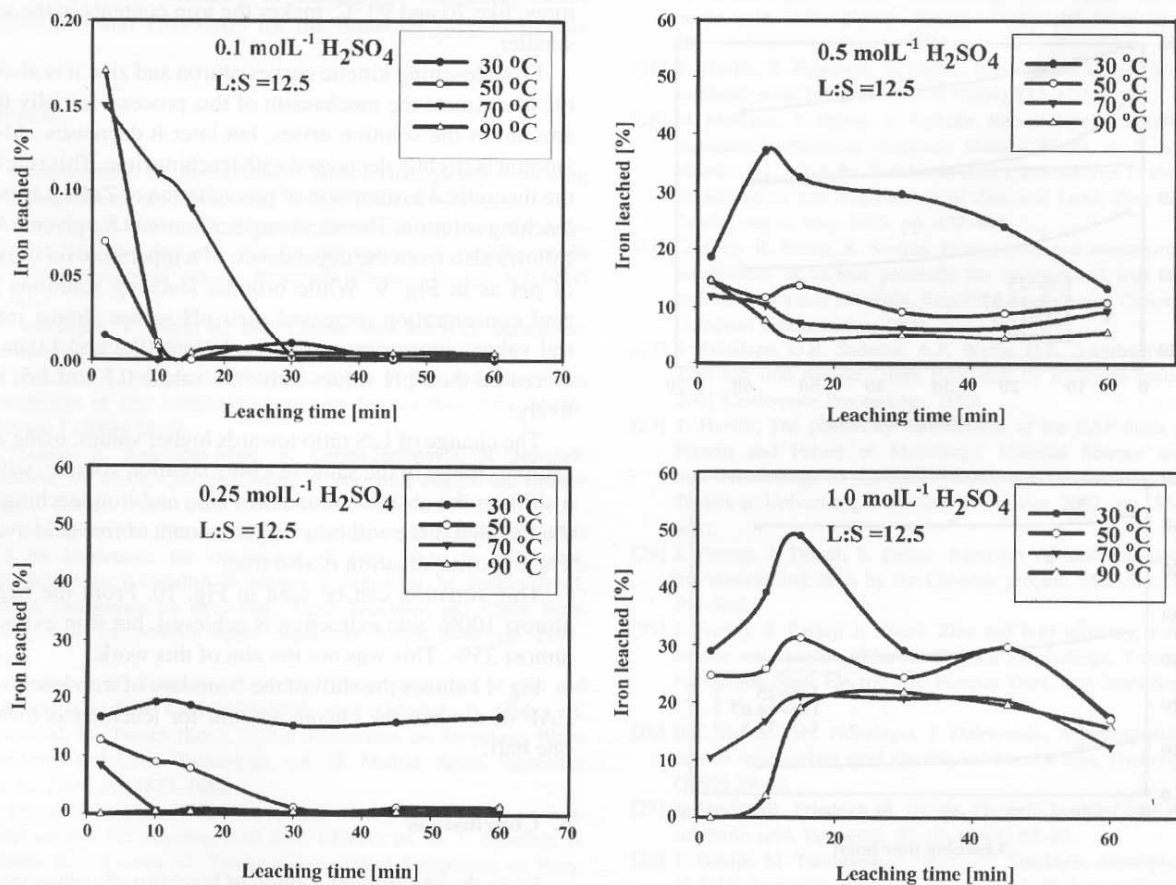


Fig. 8. Extraction of iron from EAF dust using sulphuric acid of various concentrations as leaching agent.

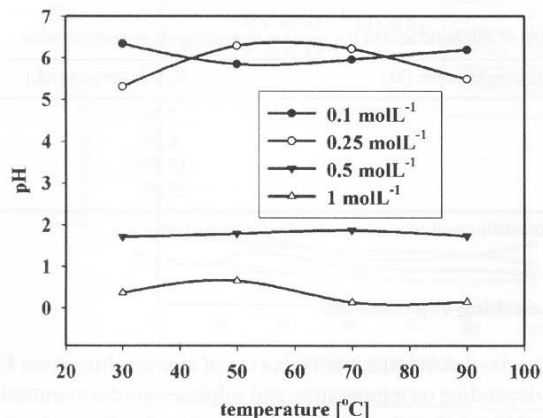


Fig. 9. Temperature dependence on the pH change of leaching solutions.

using 1 mol L^{-1} acid the slope is sharper and at the end of the experiment the leached zinc amounts are lower than by using of 0.5 mol L^{-1} acid.

Fig. 6 shows the dependence of zinc leached on temperature and acid concentration, from which it follows that by using relatively concentrated acid the leached zinc amount is almost the same, as shown in Fig. 7.

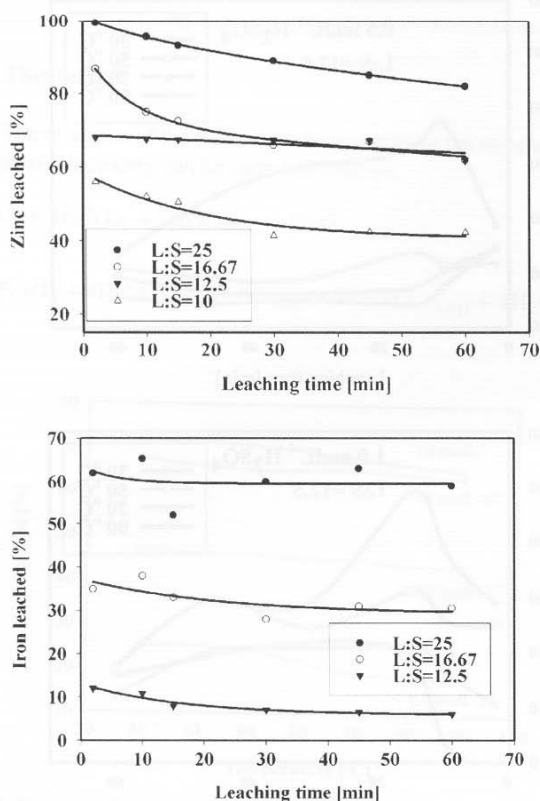


Fig. 10. (a)–(b). Leaching kinetic curves of zinc and iron by using various sample charges. Leaching temperature: 70°C ; H_2SO_4 concentration: 0.5 M .

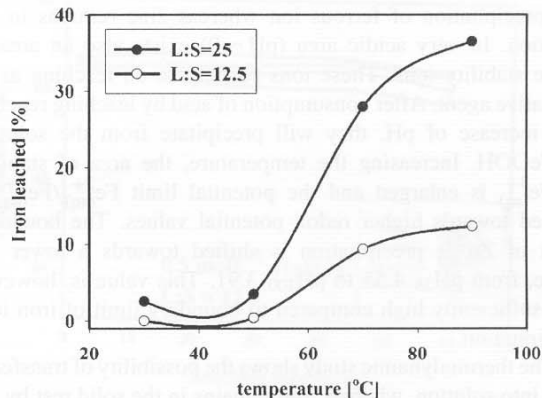


Fig. 11. Temperature dependence on iron leaching after 60 min. Leaching temperature: 70°C ; H_2SO_4 concentration: 0.5 M .

Fig. 8a–b shows the kinetic curves of iron depending on temperature and sulphuric acid concentration.

Practically, no iron is transferred into solution by leaching in 0.1 mol L^{-1} H_2SO_4 within the temperature range used. The increase of acid concentration causes the transfer of iron into solution, but this amount, in contrary to zinc, depends considerably on temperature. The amount of leached iron is relatively high at lower temperatures, but the use of higher temperatures, like 70 and 90°C , makes the iron contents in the solution smaller.

From leaching kinetic curves of iron and zinc it is also possible to estimate the mechanism of this process. Initially the iron amount in the solution arises, but later it decreases. Also zinc amount is slightly decreased with leaching time. This is related to the theoretical assumption of precipitation of ZnFe_2O_4 from the leaching solution. The necessary acid amount for given EAF dust follows also from the dependence of temperature on the change of pH as in Fig. 9. While original leaching solutions of low acid concentration increased their pH values almost into neutral values, more concentrated solutions (0.5 and 1.0 mol L^{-1}) increased their pH values only into values 0.5 and 1.5 , respectively.

The change of L:S ratio towards higher values, using a lower sample charge in the same leaching solution volume, will cause a shift on the absolute amount of zinc and iron leaching due to the fact that there will be a bigger amount of free acid available. The opposite situation is also true.

This situation can be seen in Fig. 10. From the beginning almost 100% zinc extraction is achieved, but iron extraction is almost 35%. This was not the aim of this work.

Fig. 11 shows the shift of the boundary of iron leaching from EAF dust when the charge amount for leaching is lowered to one half.

6. Conclusions

From the experimental study of leaching of carbon steel EAF dust (Gerdau S.A.) the following conclusions could be derived:

- (1) the leaching of zinc from steel EAF dust in water solution of sulphuric acid is a very fast process (minutes);
- (2) the leaching of zinc depends on sulphuric acid concentration. The maximum extraction (around 75%) was achieved by using concentrations higher than $0.5 \text{ mol L}^{-1} \text{ H}_2\text{SO}_4$;
- (3) the leaching of zinc is relatively independent on temperature, but at individual sulphuric acid concentrations, the maximum extractions of zinc were achieved at lower temperatures;
- (4) no iron from EAF dust is leached at low sulphuric acid concentrations as follows also from thermodynamic study;
- (5) the amount of extracted iron is increased with the increase of sulphuric acid concentration;
- (6) the amount of extracted iron at the individual sulphuric acid concentrations depends on the temperature used;
- (7) the optimal conditions for zinc removal from used EAF dust at the minimal iron extraction are: temperature $70\text{--}90^\circ\text{C}$ and $0.5 \text{ mol L}^{-1} \text{ H}_2\text{SO}_4$;
- (8) the decrease of L:S ratio causes also a decrease on the relative amount of leached zinc.

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