

Ultrasound effects on zinc recovery from EAF dust by sulfuric acid leaching

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(Received: 2 July 2014; revised: 17 September 2014; accepted: 22 September 2014)

Abstract: In this work, an ultrasound-assisted leaching process was studied for the recovery of zinc from electric arc furnace (EAF) dust, in which zinc was mainly present in the form of franklinite (60%). Hydrometallurgy is emerging as a preferred process for the recovery of a variety of metals, and the use of ultrasound could offer advantages over the conventional leaching process, especially for the dissolution of franklinite. Franklinite is a refractory phase that is difficult to leach and represents the main obstacle in conventional hydrometallurgy processing. Atmospheric leaching with different sulfuric acid concentrations (0.2–2.0 M) at two temperatures (323 and 353 K) was performed. The tests were conducted using both conventional and ultrasound-assisted leaching. After the leaching tests, the solid residues were characterized by scanning electron microscopy (SEM) and X-ray diffraction (XRD) techniques, whereas the leach liquor was analyzed by inductively coupled plasma spectroscopy (ICP). The use of ultrasound facilitated the dissolution of franklinite at low acid concentrations and resulted in a greater zinc recovery under all of the investigated operating conditions.

Keywords: zinc recovery; ultrasound; electric arc furnaces; dust; leaching

1. Introduction

Over the last few years, ultrasound has been investigated to assist the extraction of hydrometallurgical metals from a large variety of materials; however, the industrial application of this process has been limited, despite the recognized advantages derived from its application. For example, ultrasound-assisted leaching allows a greater release of metals in a shorter time and enables the use of lower-concentration acids at lower temperatures. Ultrasonication generates alternating low-pressure and high-pressure waves in liquids, which leads to the formation and violent collapse of small vacuum bubbles. The values of local temperature and pressure generated by the collapse of the bubbles are approximately 5000 K and 100 MPa, respectively. This phenomenon is termed “cavitation” and causes high-speed impinging liquid jets and strong hydrodynamic shear forces. These effects are used for the deagglomeration and milling of micrometric- and nanometric-sized materials, as well as for the disintegration of cells or the mixing of reactants. Because of the ultrasound effect, particles are individually scrubbed into the micron domain and mass transfer rates and molecular

diffusion are increased. Cavitation also increases instantaneous particle velocity, improving wetting and liquid–solid interface dynamics. The hydrometallurgical process is rapidly emerging as a preferred process for the recovery of a variety of metals because its energy consumption and smelter emissions are lower than those of conventional pyrometallurgical processes. Ultrasound could provide a further advantage because its use could increase the yield and reduce the reagent consumption, the energy consumption, and the thermal supply required by conventional processes [1–4].

The production of steel from scrap in electric arc furnaces (EAFs) produces a considerable amount of dust containing primarily Fe and Zn and considerable amounts of harmful elements such as Cd, As, and Cr. Because of its chemical and physical properties, the EAF dust is classified as a hazardous waste and its disposal is not trivial. Therefore, the use of this waste as a secondary raw material for iron or steel production is complicated. Mineralogical analysis has shown that the chemical and mineralogical compositions of each EAF dust are unique. Iron is present mostly as hematite (Fe_2O_3), magnetite (Fe_3O_4), and franklinite (ZnFe_2O_4), and zinc is present principally as zincite (ZnO) and franklinite

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(ZnFe_2O_4) [5–7]. The mineralogical form of the zinc in the EAF dust is considered the principal indicator for the effectiveness of the EAF dust treatment: zincite is an easily workable mineral for hydrometallurgical methods, whereas the franklinite phase is considerably complex and difficult to leach. The hydrometallurgical processes can be conducted using either acid or alkaline leaching. Alkaline leaching can be advantageous for leaving iron insoluble; however, it is not suitable for EAF dust containing franklinite because of its inability to dissolve this phase [8]. In the case of acid leaching, the use of sulfuric acid might be suitable because of its low cost. Other researchers have reported that franklinite dissolved in 3 M sulfuric acid at elevated temperatures, resulting in Zn and Fe extractions of approximately 100% and 90%, respectively; however, they also reported that diluted sulfuric acid at atmospheric pressure had no effect [9–10].

Although several research groups have investigated the leaching of EAF dust [11–15], only two studies were conducted using ultrasound. Barrera-Godinez *et al.* [16] investigated the ultrasonic-assisted leaching of pellets produced by double-kiln treated EAF dust, using acidified calcium chloride. They demonstrated that ultrasound was beneficial in the recovery of zinc from the pellets. However, the pellets contained zinc essentially as zincite (without franklinite) and the effects of ultrasound on the pellets, whose size distribution was between 100 μm and 1 mm, are not comparable with the effects on EAF dust, whose particles are substantially smaller. Dutra *et al.* [8], who used ultrasound during alkaline leaching of EAF dust, did not report any benefit to the zinc recovery.

In this work, an ultrasonic acid leaching process of EAF dust, in which zinc was mainly present as franklinite, was investigated on a laboratory-scale apparatus and under atmospheric pressure. The effects of ultrasound on the zinc recovery and franklinite dissolution, in comparison with those achieved by conventional leaching, were studied for both the leach liquor and the solid residues.

2. Materials and methods

The EAF dust was used in as-received condition from the dust collection system of a carbon steelmaking factory in Italy. The chemical composition of the EAF dust was analyzed by X-ray fluorescence spectroscopy (XRF) and by scanning electron microscopy (SEM). The SEM studies were performed on a Cambridge Stereoscan 440 scanning electron microscope equipped with a Philips PV9800 energy-dispersive X-ray spectrometer (EDS). The images were

collected in backscattered mode (BSE) with the microscope operating at 25 kV.

Mineralogical analysis was performed by X-ray diffraction (XRD) using a D500 Siemens diffractometer with a nickel-filtered Cu K_α radiation source ($\lambda = 0.15405 \text{ nm}$) operated at 40 kV and 30 mA. The size distribution of the particles was analyzed by laser light scattering measurements performed using a Malvern Mastersizer.

Atmospheric acid leaching was performed using sulfuric acid with concentrations ranging between 0.2 and 2.0 M at a temperature of 323 or 353 K for 30 min. Data from similar experiments showed that long periods of treatment are not necessary and that 30 min is sufficient to achieve the maximum dissolution of Zn. Heating was provided by a thermostated water bath, and the temperature of the liquid was periodically controlled. The experiments were conducted in a 400-mL Pyrex vessel containing 200 mL of leaching agent and 20 g of dust (liquid/solid ratio of 10 mL/g). The low frequency (20 kHz) ultrasound waves were generated by a probe made of titanium alloy (Ti–6Al–4V), which was immersed into the solution and linked to a Bandelin model Sonopuls HD 3200 sonicator capable of supplying an ultrasonic power in the range of 0–75 W/cm^2 (60 W/cm^2 was used for the experiments). In the conventional leaching process, i.e., leaching performed without ultrasonic irradiation, a constant stirring speed of 400 r/min was used to ensure the suspension of the particles.

At the end of each leaching test, the solid residue was separated from the leach liquor by vacuum filtration. The solid residue was analyzed by SEM–EDS and XRD, whereas the leach liquor was analyzed by inductively coupled plasma spectroscopy (ICP) using a Spectro Genesis spectrometer.

3. Results and discussion

3.1. EAF dust

The particle size distribution of the EAF dust is shown in Fig. 1. The particle size was not coarser than 15 μm , and approximately 70% of the particles were smaller than 2 μm . Moreover, a bimodal distribution with two major fractions was observed: a very fine-grained fraction at approximately 0.2 μm and a fine fraction at 1.8 μm . The fine size distribution indicated that physical concentration methods, such as magnetic or gravity separation, would not be appropriate for treating this material. However, leaching may be an attractive route because the reaction kinetics involving this dust should be fast as a consequence of the preponderance of fine particles sizes.

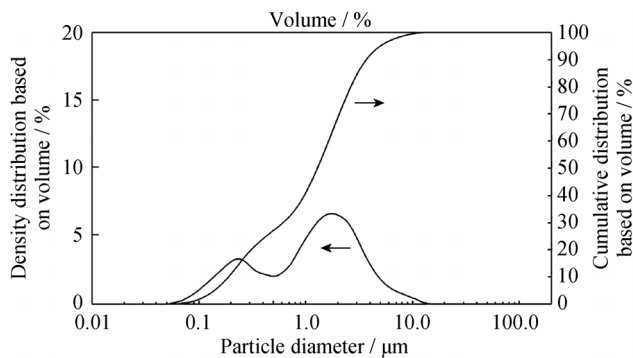


Fig. 1. Particle size distribution of EAF dust.

The XRF chemical analysis results are presented in Table 1. Iron (25.13wt%) and zinc (25.18wt%) are the main components of the dust, with small amounts of very toxic heavy metals (cadmium, chromium, and lead). The other components are calcium, silicon, and magnesium, which originate from the slag used for the production of the steel in the EAF, and manganese that derives from scrap oxidation.

The SEM–BSE images and the EDS spectra of a small but representative amount of EAF dust are shown in Fig. 2. These analyses confirmed the results obtained by the previous examinations for both the composition and the dimen-

sions of the particles. Detailed examinations of EAF dust have been reported in the literature, whereas, in this work, the analysis was mainly focused on identification of the phases in which zinc was present, in particular, franklinite. The analysis corresponding to the agglomerates composed of fine spherical particles (smaller than 1 μm) revealed that zinc and oxygen were the main constituents; consequently, they were attributed to zincite. In addition to these agglomerates, several smooth and spherical particles with a diameter between 1 μm and 5 μm and some coarser particles with a diameter of approximately 15 μm were observed. The majority of the smaller spherical particles were rich in iron, zinc, and manganese, whereas the larger ones contained chromium and calcium in addition to iron, zinc, and manganese (Fig. 2). All these spherical particles are attributed to the presence of the complex phase of franklinite. Indeed, in EAF dust, both franklinite in the form of ZnFe_2O_4 and franklinite with isomorphously substituted metals $(\text{Zn}_x\text{Me}_y)\text{Fe}_2\text{O}_4$ (Me = Mn, Cr, Ni, Co, Ca) can be present [14]. The presence of calcium, the third principal element in the dust, was observed to correspond to dust particles with irregular shapes and also to spherical particles rich in iron.

Table 1. Chemical composition of EAF dust

Fe_2O_3	ZnO	CaO	MnO	SiO_2	MgO	Cl	PbO	SO_3	Cr_2O_3	CuO	CdO	NiO
35.92	31.34	13.32	3.92	3.89	2.52	2.06	1.72	1.07	0.85	0.26	0.09	0.44
Fe	Zn	Ca	Mn	Si	Mg	Cl	Pb	S	Cr	Cu	Cd	Ni
25.13	25.18	9.53	3.06	1.82	1.52	2.06	1.59	0.42	0.58	0.21	0.08	0.03

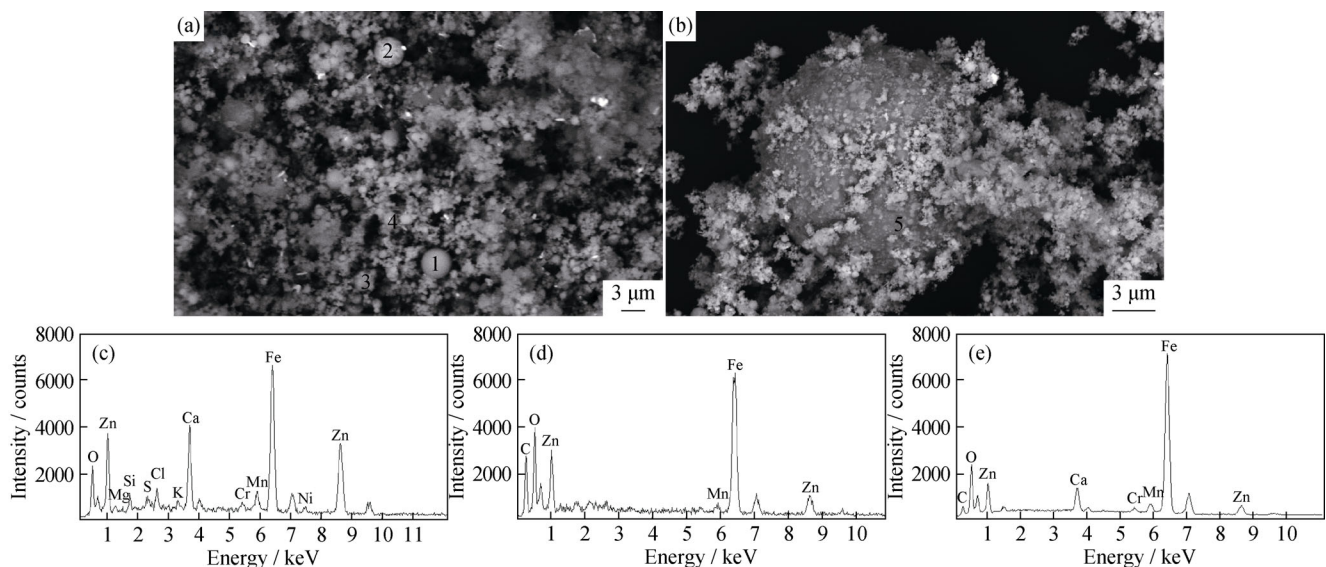


Fig. 2. (a, b) SEM-BSE images of EAF dust; (c) EDS spectrum of an extended area of EAF dust; (d) EDS spectrum in correspondence of spherical particles, marked 1, 2, 3, and 4; (e) EDS spectrum in correspondence of spherical particle, marked 5.

The XRD pattern of the as-received EAF dust is shown in Fig. 3. The patterns were evaluated using the Match!1.9d software. The three main phases were identified as franklinite ($\text{Zn}_{0.8}\text{Mn}_{0.2}\text{Fe}_2\text{O}_4$), calcium-magnetite ($\text{Ca}_{0.15}\text{Fe}_{2.85}\text{O}_4$), and zincite (ZnO). Other phases, such as CaO , MnO_2 , and SiO_2 , were likely present as minor constituents, but at concentrations below the detection limit of the instrument.

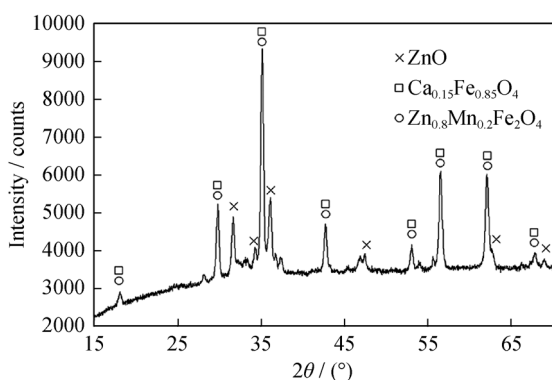


Fig. 3. XRD pattern of EAF dust.

The major obstacle in hydrometallurgical extraction is the presence of highly stable oxides containing both zinc and iron. Knowledge of the amount of franklinite in the dust is therefore important; however, quantitative analysis of the franklinite is difficult because of the significant overlap of the peaks of the franklinite phase with those of calcium-magnetite. The data were elaborated using the Maud 2.14 software to quantitatively analyze the three main phases via the Rietveld method [17]. The results of this

analysis were compared with the atomic percentage of Zn and Fe obtained from the chemical analyses, and a composition of 30wt% zincite, 45wt% franklinite, and 25wt% magnetite was deduced. In particular, (i) we assumed that all of the zinc originated from zincite and franklinite; (ii) the content of zincite was calculated via the Rietveld method with magnetite and franklinite considered as unique phases; (iii) the percentage of franklinite was deduced from the amount of remaining Zn, which was calculated on the basis of chemical analysis results; and (iv) the remaining iron was attributed to magnetite.

3.2. Leach liquors from leaching tests

The leach liquors from the leaching processes performed for 30 min were analyzed by ICP to determine Zn and Fe recovery percentages. In Figs. 4(a) and 4(b), the Zn and Fe recoveries at 323 K and 353 K, respectively, are reported as functions of the acid concentration. The Zn extraction achieved by conventional and ultrasound-assisted leaching increased with increasing acid concentration and was almost independent of temperature, except at the highest acid concentration (2.0 M), where higher yields were achieved at 353 K. The ultrasound-assisted leaching allowed a greater Zn recovery than conventional leaching at all operative conditions, and the improvement in the extraction reached a maximum value for 0.5 M acid, where the yield was increased by approximately 41% at 323 K and 55% at 353 K. Indeed, at 353 K, the Zn recovery was 38% for conventional leaching vs. 59% for ultrasonic leaching.

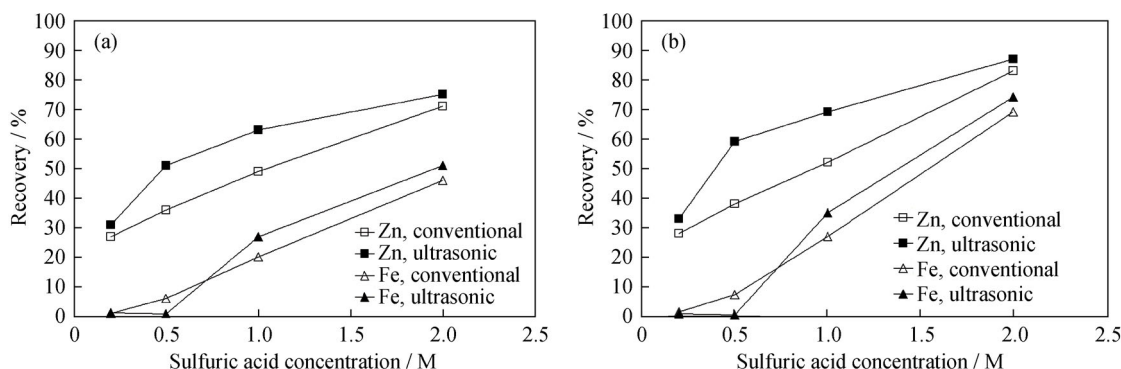


Fig. 4. Extraction of Zn and Fe by sulfuric acid with conventional and ultrasonic leaching at 323 K (a) and 353 K (b).

As expected, Fe was not recovered at the lowest acid concentration, and its recovery was highly dependent on the temperature and the acid concentration. The ultrasonic irradiation treatment promoted the extraction of Fe, except in the cases of experiments performed with 0.5 M acid. At an acid concentration of 0.5 M, the Fe recoveries of 6% and 7.2% were registered for the conventional test at 323 K and

353 K, respectively, whereas the Fe recoveries were negligible (0.9% at 323 K and 0.1% at 353 K) when ultrasonic leaching was used. The acidity measurements of the solutions gave pH 4.5 in the solutions from the ultrasonic leaching vs. pH 2 in the solution originating from the conventional leaching, suggesting that different reactions occurred during the use of ultrasound. Moreover, precipitated Fe was

observed in the bottom of the Pyrex vessel containing the leach liquor from ultrasonic leaching. The ultrasonic irradiation likely promoted greater dissolution of the phases containing Ca. This greater dissolution would have resulted in a larger consumption of acid during the reaction, causing an increase in the pH to 3, where Fe precipitation as iron hydroxide, $\text{Fe}(\text{OH})_3$, and/or iron oxide hydroxide, also known as goethite, FeOOH , started to occur.

The extractions of the harmful elements (Cr, Cd, and Pb) after the leaching tests with 0.5 M and 2.0 M sulfuric acid at 353 K are shown in Table 2. In the case of As, the extraction was not calculated because the initial content was too low to be determined by XRF analysis. The concentration of this element in the leach liquors was less than 0.01 mg/L.

Table 2. Extraction of some harmful elements present in the EAF dust, after leaching at 353 K at different sulfuric acid concentrations, with conventional and ultrasonic leaching

Element	Recovery / %			
	0.5 M, conventional	2.0 M, conventional	0.5 M, ultrasonic	2.0 M, ultrasonic
Cr	0.1	72	0.5	83
Cd	39	89	47	93
Pb	0.3	0.3	0.3	0.3

According to the results of the ICP analysis, the best zinc recovery was achieved with the highest concentration of acid (2.0 M) at 353 K; however, the elevated concentration of Fe created problems in the subsequent purification of the solution. Notably, in the case of 0.5 M acid and ultrasonic irradiation, a Zn recovery of approximately 59% was achieved, with a negligible concentration of Fe. Given that only 40% of zinc was in the form of zincite in the EAF dust, this high value of extraction suggests that franklinite was also partially dissolved using ultrasound. Other researches who conducted conventional leaching processing using both

the same sulfuric acid concentration and temperature used in the present work observed that only the zinc in the form of zincite was recovered [10–11].

3.3. Solid residues from leaching tests

The following analysis of the solid residues better explains the role of ultrasound during the leaching tests. The SEM and XRD analyses of the solid residues from the leaching tests at 323 and 353 K did not indicate appreciable variations in the amounts of Zn and Fe or in the relative amounts of the phases containing these two elements. This lack of variation was probably due to the low sensitivity of these two techniques. Therefore, in the following discussion, we report only the results for the leaching processes performed at 353 K.

The comparison between the EDS spectrum of the initial EAF dust and that of the solid residue from the leaching tests with 0.2 M sulfuric acid showed that Zn was partially removed during leaching (Fig. 5(a)). Moreover, no significant difference was observed in the signals of Fe and Zn between the conventional experiment and the ultrasonic leaching (Fig. 5(b)). The XRD patterns of the solid residues from the leaching treatments with 0.2 M sulfuric acid are shown in Fig. 6. In comparison with the XRD pattern of the EAF dust, the XRD pattern of the leaching product showed a partial reduction in the intensity of the zincite peaks, whereas the peaks of the franklinite and the calcium–magnetite remained almost unaffected, irrespective of whether conventional and ultrasonic leaching was used. The new peaks in the XRD patterns of the solid residues are attributed to calcium sulfate dihydrate (gypsum), which is a poorly soluble product of the reaction of calcium oxide with sulfuric acid. Thus, the leaching performed with 0.2 M sulfuric acid led to partial dissolution of the zincite phase, but did not attack the franklinite or the magnetite. The use of ultrasound did not appear to influence the recovery of Zn.

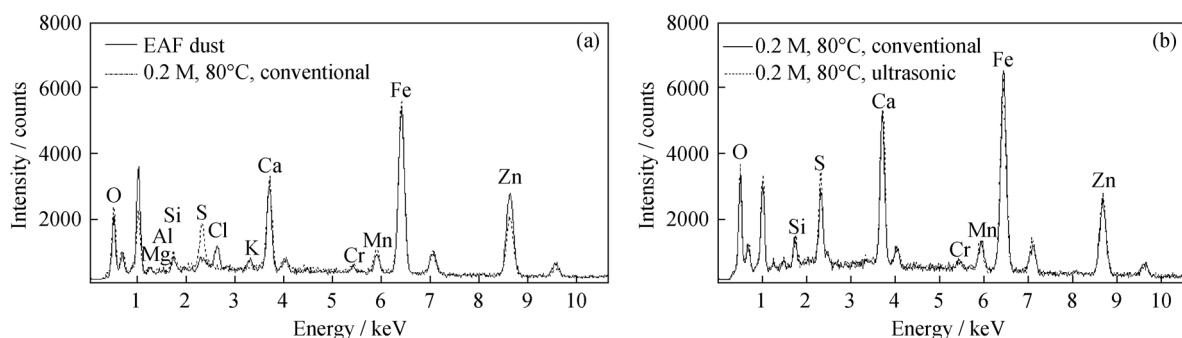


Fig. 5. (a) Comparison of EDS spectra of EAF dust and solid residue from conventional leaching with 0.2 M sulfuric acid at 353 K; (b) comparison of EDS spectra of solid residue from conventional and ultrasonic leaching with 0.2 M sulfuric acid at 353 K.

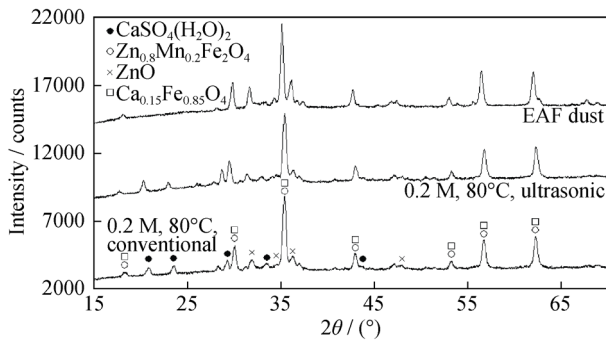


Fig. 6. XRD patterns of EAF dust and solid residue from conventional leaching and ultrasonic leaching with 0.2 M sulfuric acid at 353 K.

A comparison of the EDS spectra of the solid residues from the leaching tests conducted using 0.5 M sulfuric acid reveals that the increase in acid concentration resulted in a greater extraction of Zn while leaving the amount of Fe unchanged (Fig. 7(a)). This comparison also reveals that the use of ultrasound enhanced the recovery of Zn (Fig. 7(b)). The SEM images show that the particles of franklinite with a diameter greater than approximately 2 μm were still well observable in the solid residue derived from conventional leaching, whereas their presence was drastically reduced in the solid residue originating from the ultrasonic leaching (Figs. 8(a) and 8(b)). The elongated plates present in the solid residue are attributed to calcium sulfate dihydrate (gypsum). SEM images in Fig. 9 show two coarser, partially

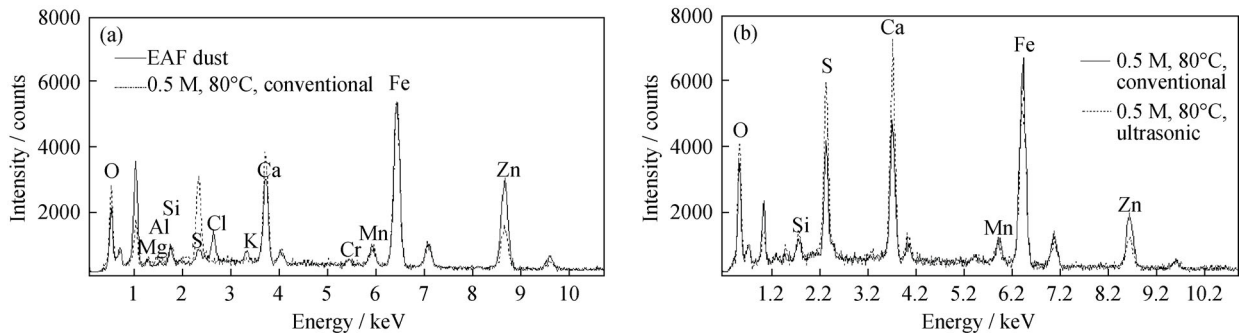


Fig. 7. (a) Comparison of EDS spectra of EAF dust and solid residue from conventional leaching with 0.5 M sulfuric acid at 353 K; (b) comparison of EDS spectra of solid residues from conventional and ultrasonic leaching with 0.5 M sulfuric acid at 353 K.

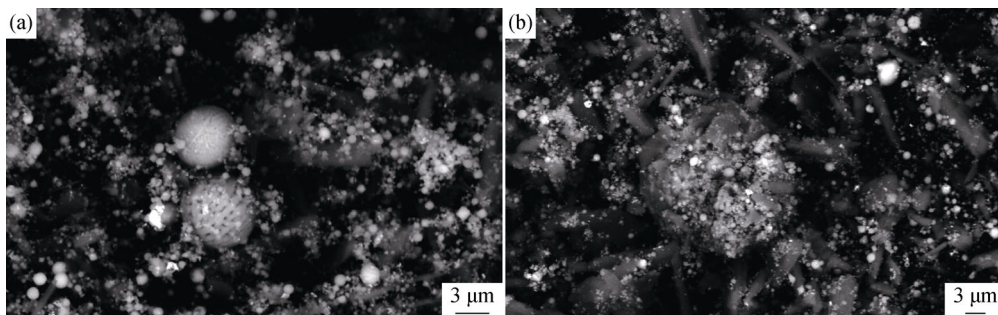


Fig. 8. SEM-BSE images of solid residues from conventional leaching (a) and ultrasonic leaching (b) with 0.5 M sulfuric acid at 353 K.

dissolved particles of franklinite. EDS analysis revealed that the darker zone of the particle in Fig. 9(a) was composed of Fe, Zn, and Mn, whereas the lighter zone was enriched with Cr, thereby explaining the failure of the particle to dissolve. The XRD patterns of the solid residues derived from leaching with 0.5 M sulfuric acid are shown in Fig. 10. The peaks corresponding to the ZnO phase were drastically reduced in intensity in the XRD pattern of the sample leached without ultrasound and completely disappeared in the pattern of the sample leached using ultrasound. Moreover, a decrease in the intensity of the peaks corresponding to the calcium–magnetite and franklinite and an increase in the intensity of peaks associated with calcium sulfate dihydrate (gypsum) were observed in the pattern of the sample leached using ultrasound. Quantitative analysis was performed by the Rietveld method using the Maud 2.14 software to quantify the calcium sulfate dihydrate phase in the conventional and ultrasonic leaching solid residues [17]. The results indicated that the calcium sulfate dihydrate concentrations in the conventional and ultrasonic leaching residues were 40wt% and 54wt%, respectively. These results suggest greater dissolution of calcium during ultrasonic leaching and, consequently, a greater amount of precipitated calcium sulfate dihydrate in the solid residue. This result can explain both the pH value of 4.5 measured in the leach liquor from the ultrasonic leaching and the aforementioned precipitation of iron.

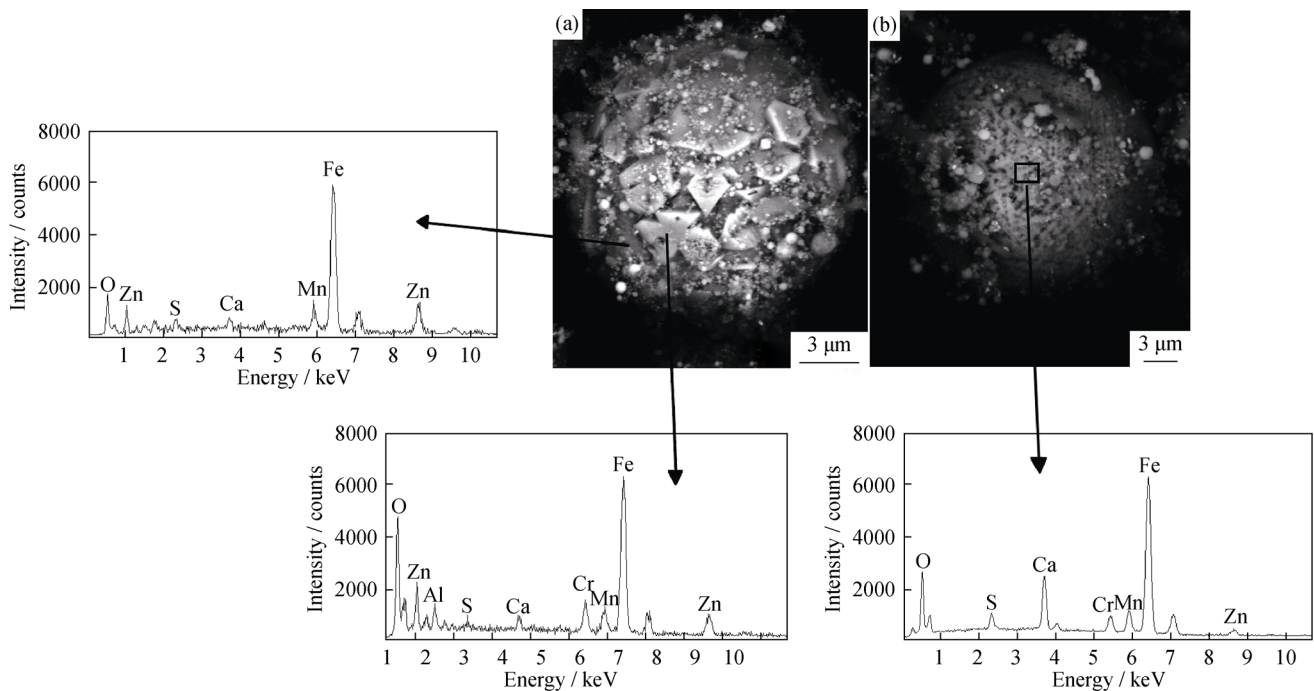


Fig. 9. SEM-BSE image of particles of franklinite in solid residue from ultrasonic leaching with 0.5 M sulfuric acid at 353 K and EDS spectra.

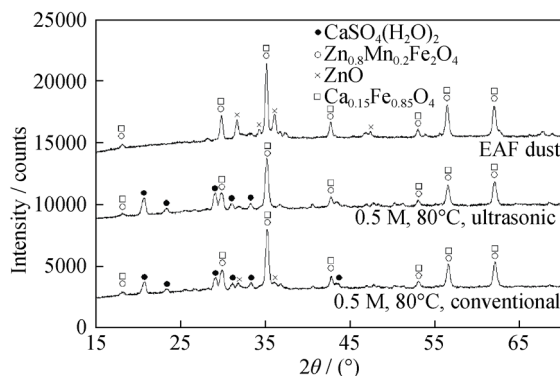


Fig. 10. XRD patterns of EAF dust and solid residues from conventional and ultrasonic leaching with 0.5 M sulfuric acid at 353 K.

By comparing the results of the ICP, SEM, EDS, and XRD analyses of the leaching with 0.5 M sulfuric acid, the use of ultrasound appeared to increase the Zn recovery because of the enhanced attack of the franklinite phase under ultrasound irradiation. In fact, as previously reported, leaching conducted under the same operative conditions in the absence of ultrasound was ineffective in dissolving franklinite [10–11].

Although ultrasound certainly improves the extraction yields of several metals from their ores, the exact mechanism is not well understood. Swamy and Narayana developed a model to describe the mechanism of ultrasound-enhanced extraction, and we adopt their model in this study to

explain the different results obtained by conventional and ultrasound-assisted leaching [1,4]. During ultrasound-assisted leaching, the explosion of the cavitation bubble generated a high effective temperature and pressure. The high temperature increased the solubility of the analytes in the leachant, and the high pressure promoted the penetration of leachant. The collapse of the cavitation bubble generated in proximity of the particles' surface produced high speed microjets, causing surface erosion and the formation of microcracks, through which the leaching solution diffused by capillary action into the interior of the particles, promoting both the breaking of the particles and the leaching process. Moreover, the particles–liquid interface was partially destroyed by acoustic streaming, promoting the dissolution of the surface. All these factors were responsible for the faster kinetics of leaching and greater yield in comparison with those of conventional leaching. According to the proposed mechanism, the coarser spherical particles of franklinite were partially dissolved and the smaller particles eroded and/or broken. The SEM images show that this mechanism mainly occurred with the particles with a diameter coarser than approximately 2 μm. In fact, the concept of “critical grain size” was elaborated by Swamy and Narayana, who demonstrated that greater benefits of using ultrasound were achieved with larger ore particles [4].

The EDS analysis of the leaching treatments conducted in 1.0 M sulfuric acid showed that both Zn and Fe were re-

moved from the EAF dust and that the amount of these two elements was lower in the solid residue from ultrasonic leaching (Figs. 11(a) and 11(b)). The XRD analysis showed that the ZnO phase was no longer present, and a significant reduction in the intensity of the peaks associated with calcium–magnetite and franklinite was observed in the XRD patterns of the solid residue from ultrasonic leaching (Fig. 12). In addition to the peaks of calcium sulfate dihydrate, peaks of lead sulfate were also detected. Similar to calcium

oxide, the lead species reacted with sulfuric acid to form very poorly soluble lead sulfate (anglesite).

The smaller amount of Fe measured in the solid residues indicated that both magnetite and franklinite were likely leached during the treatment. Thus, leaching with 1.0 M acid led to the complete dissolution of the zincite and to the partial dissolution of both magnetite and franklinite and the use of ultrasonic irradiation improved the recovery of both the Zn and Fe.

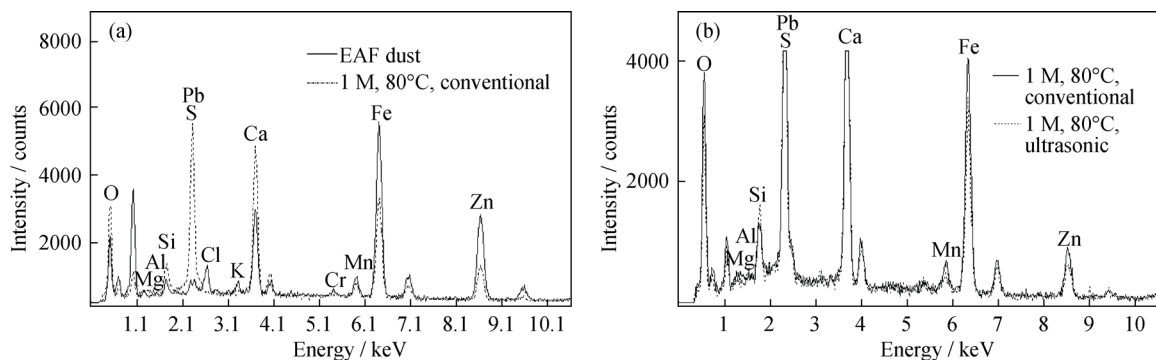


Fig. 11. (a) Comparison of EDS spectra of EAF dust and solid residue from conventional leaching with 1.0 M sulfuric acid at 353 K; (b) comparison of EDS spectra of solid residues from conventional and ultrasonic leaching with 1.0 M sulfuric acid at 353 K.

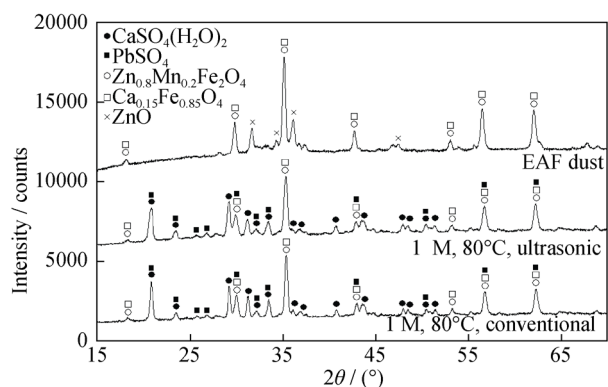


Fig. 12. XRD patterns of EAF dust and solid residues from conventional and ultrasonic leaching with 1.0 M sulfuric acid at 353 K.

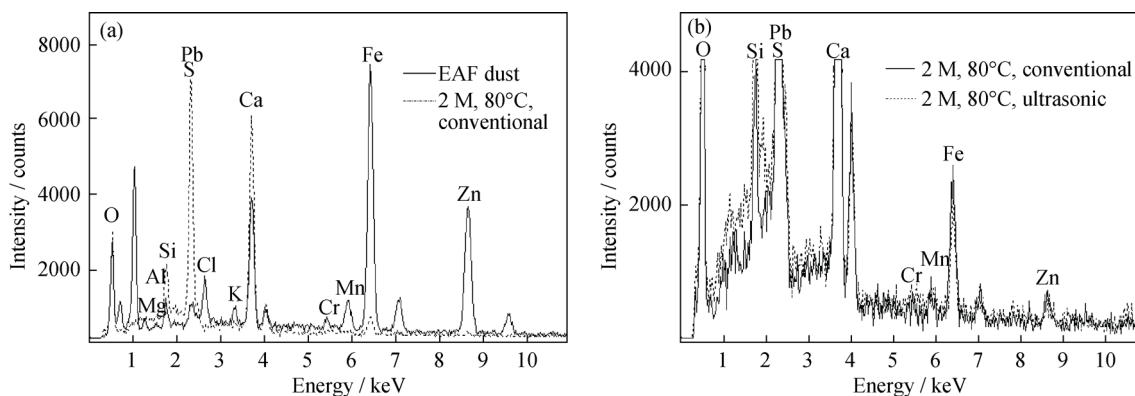


Fig. 13. (a) Comparison of EDS spectra of EAF dust and solid residue from conventional leaching with 2.0 M sulfuric acid at 353 K; (b) comparison of EDS spectra of solid residues from conventional and ultrasonic leaching with 2.0 M sulfuric acid at 353 K.

The EDS analysis of the solid residues from the leaching with 2.0 M acid indicated that the majority of Zn and Fe were removed from the EAF dust and that the ultrasound did not appear to influence the Zn recovery (Figs. 13(a) and 13(b)). The XRD analysis revealed that the main phases were calcium sulfate dihydrate and lead sulfate. The peaks of calcium–magnetite and franklinite were still present, and their intensity was reduced to a greater extent in the case of the solid residue from ultrasonic leaching (Fig. 14). During leaching with 2.0 M acid, almost all of the Zn and Fe were recovered, as evidenced by the EDS and XRD results, and the effects of ultrasound were less pronounced under these conditions.

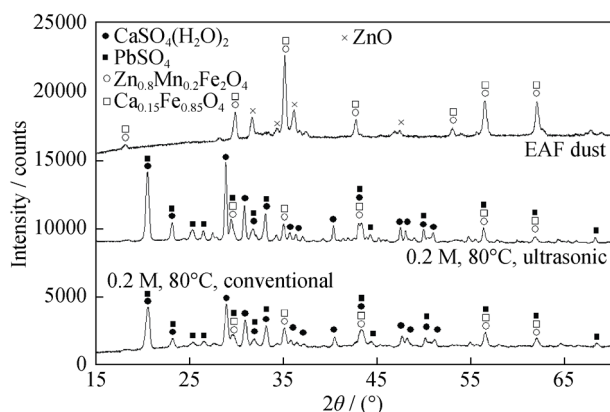


Fig. 14. XRD patterns of EAF dust and solid residues from conventional and ultrasonic leaching with 2.0 M sulfuric acid at 353 K.

Thus, ultrasound played a fundamental role in the leaching of Zn at an acid concentration of 0.5 M. At the lowest concentration (0.2 M), although ultrasound served as a mechanical surface activator for the leaching reaction, its effect was probably limited because the leaching reaction rates were very slow. At the highest acid concentration (2.0 M), the cavitation and microgrinding actions of ultrasound were less important because, under this condition, the leaching reaction rate was very fast, independent of the use of ultrasound.

Another important aspect to consider is the effect of ultrasound on diffusion, which has previously been demonstrated to be the parameter controlling the process of dissolution of zinc [11]. The improvement in the rate of dissolution and the recovery of zinc, under all of the investigated conditions, is also attributed to the enhanced diffusion induced by ultrasound via the cavitation phenomenon [1].

4. Conclusions

In this work, the effect of ultrasound on the leaching of EAF dust for Zn recovery was investigated. Atmospheric acid leaching was performed using different concentrations of sulfuric acid at temperatures of 323 K and 353 K for 30 min. According to results obtained in this study, the chemical and mechanical effects of ultrasound resulted in a greater Zn recovery under all of the investigated sets of conditions. Ultrasound irradiation was particularly efficacious during leaching with 0.5 M sulfuric acid at 353 K because it improved the dissolution of franklinite, resulting in a 55% increase in the Zn yield compared to the yield achieved using conventional leaching and induced re-precipitation of all of the dissolved Fe. At higher acid concentrations the application of ultrasound was more effective in metal recovery than

the conventional method, but its benefit was less marked. Although each EAF dust is characterized by a peculiar chemical and mineralogical composition and the effective processing conditions for each dust must be studied individually, this preliminary study indicated that ultrasonic-assisted leaching is a suitable technique to improve the dissolution of the franklinite phase, which represents the main obstacle during conventional leaching.

Acknowledgements

This work was supported by a Senior Research Grant 2011, University of Padua (Protocol GRIC13VPE5).

References

- [1] M.D. Luque de Castro and F. Priego Capote, *Analytical Applications of Ultrasound*, Elsevier Science B.V., Amsterdam, 2007, p. 99.
- [2] J.L. Luque-García and M.D. Luque de Castro, Ultrasound: a powerful tool for leaching, *Trends Anal. Chem.*, 22(2003), No. 1, p. 41.
- [3] K.S. Suslick, The chemical effects of ultrasound, *Sci. Am.*, 260(1989), No. 2, p. 80.
- [4] K.L. Swamy and K.L. Narayana, Ultrasonically assisted leaching, [in] *Advances in Sonochemistry*, Elsevier Science B.V., Amsterdam, 2001, vol. 6, p. 141.
- [5] J.G.M.S. Machado, F.A. Brehm, C.A. Mendes Moraes, C.A. dos Santos, A.C. Faria Vilela, and J.B. Marimon da Cunha, Chemical, physical, structural and morphological characterization of the electric arc furnace dust, *J. Hazard. Mater.*, 136(2006), No. 3, p. 953.
- [6] F.M. Martins, J.M. dos Reis Neto, and C.J. da Cunha, Mineral phases of weathered and recent electric arc furnace dust, *J. Hazard. Mater.*, 154(2008), No. 1-3, p. 417.
- [7] T. Sofilić, A. Rastovčan-Mioč, Š. Cerjan-Stefanović, V. Novosel-Radović, and M. Jenko, Characterization of steel mill electric-arc furnace dust, *J. Hazard. Mater.*, 109(2004), No. 1-3, p. 59.
- [8] A.J.B. Dutra, P.R.P. Paiva, and L.M. Tavares, Alkaline leaching of zinc from electric arc furnace steel dust, *Miner. Eng.*, 19(2006), No. 5, p. 478.
- [9] Š. Langová, J. Ríplová, and S. Vallová, Atmospheric leaching of steel-making wastes and the precipitation of goethite from the ferric sulphate solution, *Hydrometallurgy*, 87(2007), No. 3-4, p. 157.
- [10] V. Montenegro, P. Oustadakis, P.E. Tsakiridis, and S. Agatzini-Leonardou, Hydrometallurgical treatment of steel-making electric arc furnace dusts (EAFD), *Metall. Mater. Trans. B*, 44(2013), p. 1058.
- [11] M. Cruells, A. Roca, and C. Núñez, Electric arc furnace flue dusts: characterization and leaching with sulphuric acid, *Hy-*

- drometallurgy*, 31(1992), No. 3, p. 213.
- [12] T. Havlik, B. Friedrich, and S. Stopić, Pressure leaching of EAF dust with sulphuric acid, *World Metall. ERZMETALL*, 57(2004), No. 2, p. 113.
- [13] T. Havlik, M. Turzakova, S. Stopic, and B. Friedrich, Atmospheric leaching of EAF dust with diluted sulphuric acid, *Hydrometallurgy*, 77(2005), No. 1-2, p. 41.
- [14] T. Havlik, B. Vidor e Souza, A.M. Bernardes, I.A.H. Schneider, and A. Miškufová, Hydrometallurgical processing of carbon steel EAF dust, *J. Hazard. Mater.*, 135(2006), No. 1-3, p. 311.
- [15] P. Oustadakis, P.E. Tsakiridiris, A. Katsiapi, and S. Agatzini-Leonardou, Hydrometallurgical process for zinc recovery from electric arc furnace dust (EAFD): Part I. Characterization and leaching by diluted sulphuric acid, *J. Hazard. Mater.*, 179(2010), No. 1-3, p. 1.
- [16] J.A. Barrera-Godinez, T.J. O'Keefe, and J.L. Watson, Effect of ultrasound on acidified brine leaching of double-kiln treated EAF dust, *Miner. Eng.*, 5(1992), No. 10-12, p. 1365.
- [17] L. Lutterotti, *Maud Version 2.0: Materials Analysis Using Diffraction*, 2005 [2014-04-20]. <http://www.ing.unitn.it/~maud>.