



## Evaluation of environmental compatibility of EAFD using different leaching standards

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### ABSTRACT

A study on laboratory scale to evaluate the environmental compatibility of electric arc furnace dust (EAFD) is reported in this article. EAFD, a waste by-product of the steel-making process, was generated on a steel plant located in Brazil. Different leaching tests, NBR10005 (Brazilian), AFNORX31-210 (French), JST-13 (Japanese), DIN38414-S4 (German), TCLP (American), and NEN 7343 (Netherlands) were conducted. These leaching procedures are batch tests and are columns conducted in a way that an equilibrium condition should be achieved. The pH of the medium showed a crucial parameter governing the release of metals from the solid phase into solution. As the pH of the medium varies with the leachant used, this determines the dissolution of the elements. Zn, Pb, Mn, Cd, and Cu presented high leachability at NBR10005 procedures (acid pH). Except Pb and Cr, the leachability of all other metals in leaching tests with alkaline pH decreases with the increase of the pH. NBR10005 classifies the EAFD as a hazardous waste due to high concentration of Pb and Cd in leachate. The column tests are presented in the following order of leaching: Pb > Cr > Zn > Mn > Cu > Cd.

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### 1. Introduction

The electric arc steel-making process generates electric arc furnace dust (EAFD). During melting, there is generation of gaseous and particulate emissions due to the high temperatures used. Certain elements volatilize and, after oxidation and cooling, are collected in filters. The collected material has small particle size in media less than 3 μm and is called EAFD. Approximately 1–2% of the charge is converted into EAFD. About 650–700 kt/year of EAFD are generated in USA [1].

According to the Brazilian Steel Institute [2], the Brazilian steel industry has produced 30 million t crude steel in 2002. The production of steel by electric arc furnace was 4.5 million t considering a dust generation of 2%, approximately 90 kt of EAFD were generated from carbon and stainless steel production in 2002.

The disposal of this waste has become a serious problem in recent years. The greater availability of steel scrap, which represents a large part of the charge, has increased the production of steel from electric arc furnaces [1]. Nowadays, resources are being applied in Brazil for the implementation of modern dust-removal systems at the mills, using technologies that prevent the emission of residues into the atmosphere. Socked filters are pri-

marily used to prevent contamination of these particles in the atmosphere. In the previous year, these pollution control systems have generated important quantities of dust to be disposed. The study of the mobility of heavy metals from the solid waste into water became necessary due to the widespread generation of these wastes.

The mobility of metals into environmental media has different controlling parameters. Factors such as pH, redox potential, and temperature by itself, but mainly in combination and often abetted by bacterial processes, affect the solubility, mobilization, and precipitation/deposition of potentially toxic metals. These factors along with several other factors in complex reactions determine the chemical forms (metal species) that are introduced to an environment. They also influence changes of metal species that may take place once equilibrium is established during interaction with an environment [3].

Leaching tests are done in waste materials to provide information about the release of specific contaminants under reference conditions or under conditions that may approximate more closely or may simulate the actual field situation under consideration. The number of available leaching/extraction tests is very large throughout the world, but all existing tests can be grouped according to their main characteristics such as: (i) single-batch leaching tests (equilibrium based), (ii) multiple-batch and percolation tests (mostly based on Local Equilibrium), (iii) static methods, and (iv) dynamic leaching tests [4].

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**Table 1**  
Leaching conditions, advantages (a) and disadvantages (d) for different leaching procedures.

Test Method	NBR10005 (Brazilian) EP-EPA (USA)	X 31-210 (French)	JST-13 (Japan)	DIN 38414 (German)	NEN 7343 (Netherland)
Leaching Medium	Acetic acid (d)	Deionized water (a)	Deionized water	Deionized water	Nitric acid
Number of Extractions	1 (d)	3 (a)	1 (d)	1 (d)	7 (a)
Time of Extractions	24 h (d)	Successive Leaching (24 h + 16h + 16h) (a)	16 h (d)	24 h (d)	400 h (a)
Liquid: Solid Ratio	16–20 L kg <sup>-1</sup>	10 L kg <sup>-1</sup>	10 L kg <sup>-1</sup>	10 L kg <sup>-1</sup>	10 L kg <sup>-1</sup>
pH	5.0 ± 0.2 (d)	Determined by sample (a)	Determined by sample (a)	Determined by sample (a)	Determined by sample (a)
Mechanism	Batch (d)	Batch (d)	Batch (d)	Batch (d)	Column (a)
Particle Size (φ)	<9.5 mm	<4 mm	0.5–5 mm	<10 mm	<10 mm

In this study, the applicability of different batch leaching procedures and one column test, considered a dynamic one, was evaluated. In all batch tests evaluated, the waste was exposed to a leachant with agitation for a determined period of time. The leachate is then analyzed for the concerned contaminants. These batch tests are a simple and inexpensive method for assessing the leachability of a waste, but they very often aim to achieve equilibrium at the end of an extraction period. According to a stipulated liquid/solid ratio (L/S), an equilibrium state is reached when waste solubilization in closed vessels is at the maximum value in a given leaching time. Particle size in the waste samples is often reduced to accelerate the attainment of equilibrium [5]. The release of heavy metals in water is a function of pH, L/S ratio, particle size or superficial area, and experiment duration. These parameters are different for each leaching test, which makes the study of the influence of the different conditions important, because none of the tests have actually been field validated to verify the prediction of contaminant release [6].

The column tests are singled out as being the simulating what really happens in the environment, by providing a mechanism for the dynamic process of leaching. However at the international level does not know parameters that classify the solid waste that can be used for comparison with results from column tests [4].

The aim of this study was to evaluate the environmental compatibility of the EAFD using different leaching conditions. The metals selected for this study, for their concentration and hazardousness, were Cr, Pb, Zn, Cu, Cd, and Mn. The assays was in accordance with Brazilian (NBR 10005) [7], French (AFNOR X31-210) [8], Japanese (JST-13) [9] and German (DIN 38414-S4) [10] batch tests. Besides a column test (NEN 4373) was also applied the French test is a multiple batch test, while the others all single batch tests.

Table 1 shows the leaching test conditions and the advantages and disadvantages of the parameters based on the rules of leaching studied. The different ways in which the tests of leaching occur determine the different pH values with different solubilizations from the elements. Use acetic acid as leachant, according to Brazilian and American standards (pH 5 ± 0.2), elevates the values of concentrations of metals by increasing the solubilization of these metals. Thus, these standards, also for being a batch and not a dynamic system of leaching, do not give a good simulation for the happenings in the environment. In contrast, batch tests use deionized water as leachant and the residue as pH determinant of the resources in a real simulate the behavior of leaching of metals, according to what happens in the environment. Therefore, the French, Japanese, and German standards show values of concentrations of metals well below the standard shown by the Brazilian. In addition the exchange mechanism for batch column, with the standard Dutch by the time of increased leaching of tests, shows a more actual extraction of elements as those that occur in nature.

Information regarding physical and chemical properties of EAFD is prerequisite for the study the leaching characteristics and the consequent environmental aspect of waste disposal; therefore, characterization of EAFD is included in the aim of this article.

## 2. Experimental procedure

The EAFD studied in this article was generated in a steel mill of Brazil and was collected directly from the sleeve filter. They generate approximately 10 kg of EAFD per ton of steel produced.

Dust chemical analysis was carried out by an X-Ray Fluorescence Spectrometer (XRF), model RIX – 2000 RIGAKU. The results were obtained by comparative analysis with materials patterns of similar chemical composition. The EAFD was also directly analyzed in the Scanning Electron Microscope (SEM) model JSM.5800 coupled with a microprobe Voyager (EDS) model EDS-3.4.2. The mineralogical composition of the dust was determined by an X-Ray Diffractometer (XRD), SIEMENS model D – 5000, at a 2θ angle with 2 to 70° degrees' scanning, which allowed a clear splitting of the interatomic pitches of crystals in the sample. Another XRD analysis was done after leaching according to Brazilian and French tests. In this case, the leachate was evaporated, and the obtained concentrate was analyzed for the determination of the crystalline phases. To prepare a sample to XRD analysis, an aliquot was placed on an electric plate at 250 °C and was then dried in an oven at 50 °C. Salts obtained from the leached extract were then analyzed by X-ray diffraction.

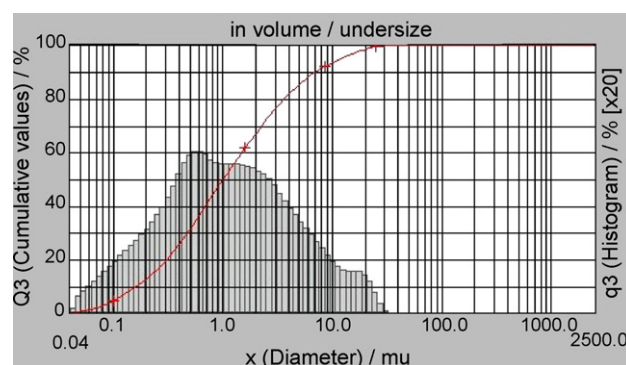
Leaching tests to determine the release of Pb, Cu, Mn, Cr, Cd, and Zn from the solid phase to the water followed the methodologies of international leaching tests. The advantages and disadvantages of leaching conditions of the applied leaching tests are summarized on Table 1.

After the dust was leached (three samples each time), the metal concentrations in the extract were determined by atomic absorption spectrometer. The equipment, model Analyst – 100 PERKIN ELMER, used flame air acetylene for all determinations (FAAS).

## 3. Results and discussions

Fig. 1 shows a granulometric distribution of EAFD and Fig. 2 represents a photograph of the EAFD evaluated.

Leaching conditions, advantages (a) and disadvantages (d) for different Leaching Procedures Test Method.



**Fig. 1.** Granulometric distribution of EAFD.



Fig. 2. EAFD image.

The results of the particle size distribution of EAFD are shown in Table 2. Table 3 represents the chemical composition of EAFD obtained by XRF and EDS and mineral composition by DRX. The element Cd was analyzed also by FAAS because its concentration was lesser than the detection limits from the other techniques. Three different analyses were done to obtain the mineralogical composition. Fig. 3 shows the presence of the following crystalline phases on EAFD: Pirolusite, Magnetite syn, Graphite, Hematite syn, Aluminum Chromite syn. Fig. 4 shows the presence of magnetite and hematite. Fig. 5 shows the presence of hydrated calcium sulfite.

**Table 2**  
Distribution for particle size.

Distribution for particle size	Diameter
10%	<0.16 $\mu\text{m}$
50%	<1.00 $\mu\text{m}$
90%	<6.82 $\mu\text{m}$
Medium	<2.58 $\mu\text{m}$

**Table 3**  
Chemical composition of EAFD.

Species	mg kg <sup>-1</sup>
Cr	13,800
Mn	22,500
Cu	1,839
Zn	197,400
Cd	300
Pb	10,000

The elements present on all these phases were already detected during the analysis of the chemical composition done by XRF and EDS. Figs. 6 and 7 present the case where the leachate was evaporated and the obtained concentrate was analyzed to determine the crystalline phases. This was done with the leachate obtained after Brazilian and French tests. The results showed one crystalline phase: gypsum,  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . The presence of this compound could be explained by the oxidation of the hydrated calcium sulfite ( $\text{CaSO}_3 \cdot \frac{3}{2}\text{H}_2\text{O}$ ) present on the EAFD (Fig. 5).

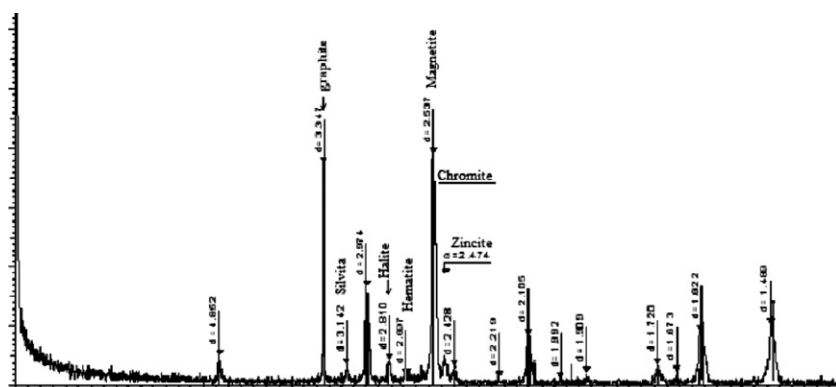


Fig. 3. X-Ray Diffractogram of EAFD—First analysis.

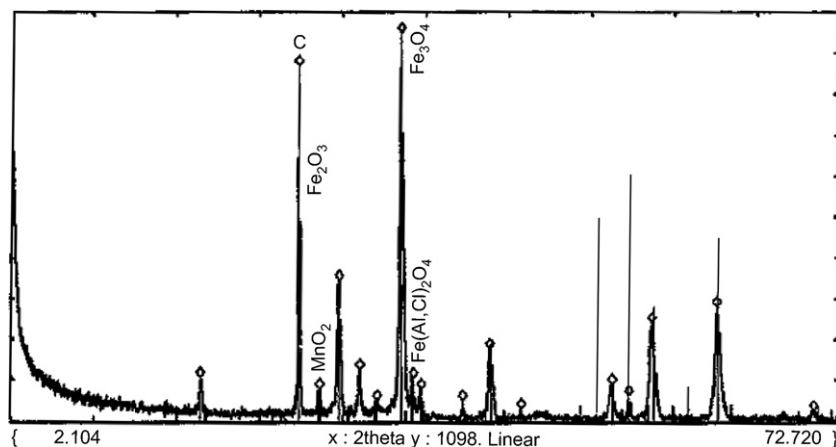


Fig. 4. X-Ray Diffractogram of EAFD—Second analysis.

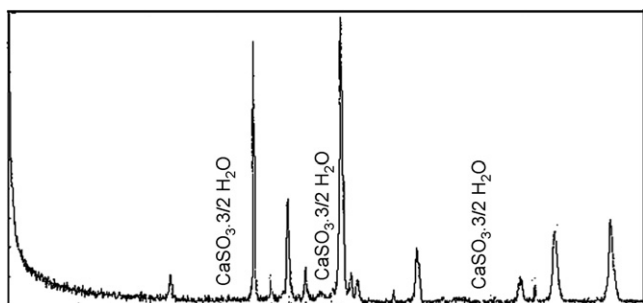


Fig. 5. X-Ray Diffractogram of EAFD—Third analysis.

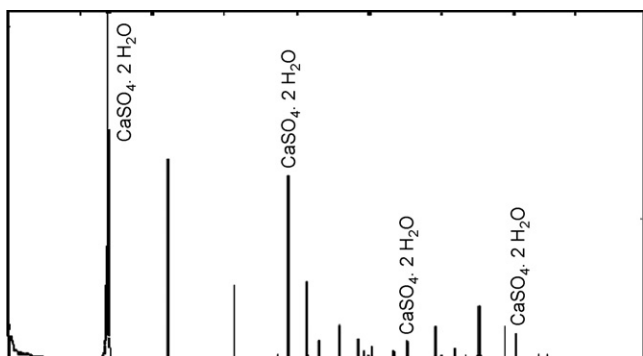


Fig. 6. X-Ray Diffractogram of EAFD after leaching according to NBR1005 (Brazilian Test).

Table 4 presents characteristics of the leachate obtained after leaching according to different leaching tests.

The chemical analysis of leachate obtained after NBR 10005 show higher concentrations for the following elements, in decreasing order: Zn, Pb, Mn, Cd, Cu, and Cr. The EAFD is extremely alkaline. To reach the pH required by NBR 10005, the maximal permitted acetic acid concentration was used. The pH remained in a range of  $5 \pm 0.2$ .

It is evident from Table 4 that the final pH of leachate from NBR10005 test is different from the others, which can bring differences on solubilization of metals. Some heavy metals will be mobilized under acidic conditions and others under basic conditions.

Iron was analyzed to serve as a reference for the other elements, mainly as a reference to chromium because these two elements dissolve in different pH ranges (compare with the results presented on Table 4). The EAFD contains Fe and Cr. At a pH around 5.0,  $\text{Fe}^{2+}$  is solubilized and remains partially soluble at the leachate. Chromium

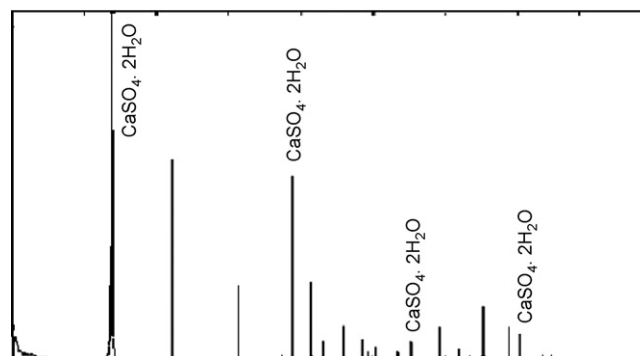


Fig. 7. X-Ray Diffractogram of EAFD after leaching according to AFNOR-X31210.

remains as  $\text{Cr}^{3+}$  and is not solubilized at these conditions. At an alkaline pH, the  $\text{Cr}^{3+}$  can be oxidized to  $\text{Cr}^{6+}$ , which is soluble; it was already indicated by studies of Aldrich [11]. Different oxidants could also act on this medium. At this pH range, Fe will precipitate and will not remain on the leachate. This indicates that according to the pH, we will find Fe or Cr predominantly on the leachate. On the analysis of the leachate obtained after NBR 10005 test, a strong presence of iron and a small quantity of chromium on the leachate can be seen. This was confirmed by qualitative analysis done according to the Standard Methods for examination of water and wastewater [12], which indicate a negative test for  $\text{Cr}^{6+}$  on the leachate obtained after the NBR 10005 leaching experiment. On the other way, the results obtained after French, Japanese, and German Standards have shown higher chromium concentration associated with a smaller iron concentration. This was confirmed by the test of the Standard Methods, where  $\text{Cr}^{6+}$  were detected.

The presence of Cr on EAFD cannot be shown by the chromite,  $\text{FeCr}_2\text{O}_4$  (Fig. 3), which is explained by the coincidence of the main line from chromite with the main line of magnetite on the XRD analysis. The secondary lines of chromite remain as an evidence of its presence on the dust. The compound Aluminum Chromite syn,  $\text{Fe}(\text{AlCr})_2\text{O}_4$ , (Fig. 4), is an indicative of chromium on EAFD.

Oxyanionic species, like chromium, feature their highest leachability at mild alkaline to neutral pH [13,14]. Therefore, the behavior of chromium is markedly different from that of other metals. The oxyanions often show a maximum leachability around a neutral pH, where metallic cation leachability is at a minimum. In the experiments carried out in this article, Cr showed the lowest mobility, compared with other metals, in acid extracts (NBR 10005). Experiments done by Sanchez [15] showed greater Cr mobility in a basic extract of coal ash.

Zn presented higher solubility than other metals in a moderately acidic pH medium, among values that range from 5.0 to 6.0.

Table 4  
Comparison among results of leaching tests in different countries.

	Brazil	French	Japan	Germany	Netherland
Leaching Test	NBR 10005	X 31-210	JST - 13	DIN 38414-S4	NEN 7343
Total Test Duration (h)	24 h	56 h (24 h + 16h + 16h)	16 h	24 h	400 h
Final Leachate pH	$5 \pm 0.8$	>12.5	>12.5	>12.5	12
Final Leachate Conductivity ( $\lambda$ ) ( $\text{mScm}^{-1}$ )	7.9	8.4/1.0/0.7	9.0	9.0	–
Species ( $\text{mg L}^{-1}$ )					
Cu	3.6	0.03/ND/ND	0.02	0.02	0.015
Zn	2800	0.51/0.07/0.06	0.8	0.21	0.45
Pb	41	8.6/0.27/ND	29	14	14.9
Cr	0.12	4.9/2.2/1.4	5.1	5.1	2.3
Cd	9.6	0.01/ND/ND	ND	ND	<0.01
Mn	34	0.03/0.03/ND	0.01	0.01	0.025
Fe	5.0	0.06/NA/NA	0.1	0.08	ND

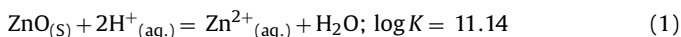
ND = Not Detectable.

NA = Not Analyzed.

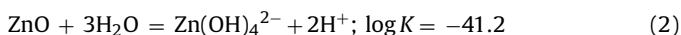
**Table 5**  
Batch Leaching Tests with high L/S ratio total.

Total Test Duration (h)	Liquid: Solid Ratio (L kg <sup>-1</sup> )	Particle Size (mm)	Final Leachate pH	Final Leachate Conductivity (mS cm <sup>-1</sup> )	Leachate Concentration (mg L <sup>-1</sup> )						
					Cu	Zn	Pb	Cr	Cd	Mn	Fe
24 h	20	<9.5	12.0	5.2	0.02	0.71	2.4	1.4	ND	0.01	0.03

According to Heck and Weber [16,18], the reaction of the dissolution of zincite in the acid medium is:



The leachability of Zn is similar to that of lead because it also has amphoteric properties. However, leachability at a high pH is not as pronounced as that of lead (Table 5). According to Heck and Weber, the reaction that can illustrate this behavior is:



From reactions (1) and (2), it can be seen that there could be dissolution of zinc in acidic and alkaline medium. However, the formation constants show that reaction (2) would not be spontaneous. The mineralogical analysis of EAFD showed zincite on the composition. This is a basic oxide. These compounds are usually not easily dissolved in water.

Lopez studying lead slag has also observed a decrease in zinc concentration on leachate with an increase of pH [17]. According to Siegel, elements such as Cu and Zn are essentially immobile under basic pH conditions [3].

In all leaching results, the presence of Cu can be observed. However, the mineralogical analyses of dust, done by XRD, did not indicate the presence of copper in any mineralogical compound (Figs. 3–5). This occurs probably because the copper concentration on the dust was smaller than the detection limit of the technique. However, copper was verified by XRF (Table 2). According to Heck [16], Pb and Cu can be present on EAFD as  $\text{PbFe}_{0.67}\text{W}_{0.33}\text{O}_3$  and  $\text{Ni}_{0.41}\text{Zn}_{0.60}\text{Cu}_{0.022}\text{Sn}_{0.01}\text{Fe}_2$ , which have lines that coincide with those of franklinite ( $\text{ZnFe}_2\text{O}_4$ ) and zincite ( $\text{ZnO}$ ), or both, which bring some difficulties for identification by X-ray diffraction.

The element Cu, also forming basic oxides, is more soluble in a favorably acid medium. This can be seen in Table 4, where the Cu solubilization is greater for leaching according to NBR 10005 than for leaching with higher pH. The leachability of Cu decreases as pH increases. Sloot, Heasman, and Quevauviller [4] have also cited this result. Same results were observed for Cd mobility.

Mn, from pyrolusite ( $\text{MnO}_2$ ) (Fig. 4), solubilize in a greater extend when leaching is done according to NBR 10005. As pH values increased, according to the French, Japanese, and German standards, there was a sudden decrease on Mn concentrations, indicating a low mobility of this element.

**Table 6**  
Comparison of contaminant concentration in leachate and maximal concentration of elements according to Brazilian and German regulations.

	Leaching according to NBR 10005	Maximal Concentration according to NBR10004	Leaching according to DIN 38414-S4	Maximal Concentration according to TA-Abfall
Final Leachate pH	5 ± 0.8	NR	>12.5	4–13
Final Leachate Conductivity (λ) (mS cm <sup>-1</sup> )	7.9	NR	9.0	100
Species (mg L <sup>-1</sup> )				
Cu	3.6	NR	0.02	10
Zn	2800	NR	0.21	10
Pb	41	5.0	14	2
Cr	0.12	5.0	5.1	0.5
Cd	9.6	0.5	ND	0.5
Mn	34	NR	0.01	NR
Fe	5.0	NR	0.08	NR

ND = Not Detectable.

NR = Not specified by the regulation.

The conductivity values indicated the presence of dissolved ions in the Brazilian, French, Japanese, and German tests. For successive leachings according to French procedure, conductivity values decrease each time. This result is associated with metal concentrations (Table 5). For the French test, it was possible to observe that although some elements including Cu, Zn, Pb, Cd, Mn, and Fe were released rapidly in the initial stage of leaching, measurable amounts of metals were still detectable in EAFD treated three times with deionized water. Such leaching behavior can indicate slow and long-term leaching of elements. Choi [19] has also detected a similar behavior in the leaching of fly ash. For chromium, this behavior was much more pronounced. Because redox conditions can affect the Cr leaching significantly [20], other studies will be done to evaluate this parameter.

The Japanese test is done for 16 h, and the German and Brazilian tests for 24 h. On the concentration results, it was not possible to determine changes of Cu and Mn on leachate on these two different times of experiments. To the other metals (Pb, Zn, Cr, and Fe), there were concentrations changes, but it was not possible to determine a behavior tendency in the mobility of metals.

The predominant extractor medium for German, Japan, and French leaching tests was only deionized water; thus, pH values of the medium above 12 predominated for these standards, showing a low leaching mobility for most of the metals except Pb and Cr. However, lacks of uniformity in the results of the metal concentrations were observed (Table 6).

Table 6 shows a comparison of contaminant concentration in leachate and maximal concentration of elements according to Brazilian and German regulations.

To evaluate differences on L/S ratio, a new experiment, similar to the German test, with deionized water, pH 12 and a higher L/S ratio (20 L/kg) was determined. This new L/S ratio is similar to the Brazilian one. The results are shown in Table 5. From these results, it can be seen that there are no clear differences between this and the other alkaline tests (French, Japanese, and German).

The tests studied in this research followed the same batch leaching mechanism. When the leachant reaches the dust, an alkaline residue, the pH of the solution greatly increases. Because the solution is confined in the leaching flask, the equilibrium is attained, rendering the mobility of metals non representative under these conditions. According to Jackson [21], extraction methods using

deionized water as extraction fluid are more representative from natural phenomena than procedures using acetic acid or acetate buffer. The introduction of an acid source, as in the test from USA (TCLP) or NBR10005, greatly overestimates the leaching potential of a waste. However, the experiments presented in this article show that for alkaline wastes, as EAFD, pH is the most crucial parameter because in water, they achieve a pH higher than 12. Sloot [13] and Chang [22] have already determined the high significance of pH on leaching wastes. According to the literature, the pH of the resulting leachate is the factor governing the concentration of metals in solution and is more important than the concentration of the element in the waste [23].

By the results of leaching according to NBR10004, the Brazilian Regulation of solid wastes [24] classifies the EAFD as a hazardous waste due to high concentrations of Pb and Cd at the leachate. According to German Regulations, the waste would also be classified as hazardous, but this time due to Pb and Cr concentrations. Table 6 presents these results and the comparison of these regulations.

#### 4. Conclusions

The different concentrations of obtained metals in the batch tests by the international leaching standard and column tests, which are study in this work, show the leaching conditions importance in the behavior of extraction of these elements in the EAFD.

The pH values for the different procedures have a significant influence on the behavior of mobility of metals by leaching batch tests. The results can be divided into two groups based on the different pH values: Group I, NBR10005 and Group II, French, Japan, German, and Netherland tests. Zn, Pb, Mn, Cd, and Cu were the main elements leached from these wastes according to NBR 10005. For French, Japanese, German, and Netherland standards, the same elements, except Pb, showed an abrupt reduction on mobility. Pb, both in acidic and alkaline medium, showed high release from solid phase, although the results presented disparities. Cr presented higher mobility when the waste was leached according to French, Japanese, and German standards. All these results suggest that the conditions of leaching test should be carefully specified, for example the leaching medium. Due to the different obtained results of leaching procedures, it is necessary to optimize a leaching methodology to this waste to obtain a uniformity of dates.

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#### Appendix A

##### Symbols

AFNOR X31-210	Association Française de Normalization
DIN 38414S4	Deutsches Institut für Normung
EAF	Electric arc furnace
EPA	Environmental Protection Agency
FAAS	Flame atomic absorption spectrometry
JST-13	Japanese standard
mScm-1	Millisiemens per centimeter

NBR10005	Leaching Brazilian standard by solid waste
NEN 7343	Leaching Netherland standard
Ratio L/S	liquid/solid ratio
SEM-EDS	Scanning electron microscopy-electron diffuse spectrometer
XRD	X-ray powder diffraction
XRF	X-ray fluorescence

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