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EAFD – A THERMODYNAMIC ANALYSIS AND CLASSIFICATION OF DUST TYPES*

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Abstract

Steel production process generates waste, among which is the electric arc furnace dust (EAFD). This waste has varying amounts of zinc, which is principally contained in the minerals zincite (ZnO) and franklinite (Fe_2ZnO_4). The presence of Zn is capable of economically motivating the recycling of this residue and facilitating the co-processing of the remaining material. The zinc content in EAFD and the presence of zincite, however, have significant influence on the application of hydrometallurgical recovery processes (economic alternatives to the Waelz process). In this study, based on the computational thermodynamic tool FactSage, theoretical considerations are made on the conditions for the formation of zinc-containing minerals found in EAFD. Through thermodynamic analysis, the best conditions for obtaining a high fraction of ZnO were analyzed, which favor the hydrometallurgical processes for EAFD treatment. A classification of dust depending on the phases present in the EAFD is suggested as a guide for the feasibility analysis for the use of a hydrometallurgical process.

Keywords: EAFD; EAFD-phases and mineralogy; Computational thermodynamics.

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1 INTRODUCTION

During the production of steel in semi-integrated plants, the generation of dust in electric arc furnace, EAFD, is unavoidable. This waste is constituted essentially by the solid oxides of iron; however, when using galvanized scrap in steelmaking, the main EAFD constituents become solid oxides of iron and *zinc*.

EAFD generation takes place by *vaporization* of ferrous and nonferrous metals and *ejection* and *dragging* of steel, slag and other particles. According to Guézennec *et al.* [1], the prevailing mechanisms of dust precursor emission appear to be the volatilization (27 wt.%) and the bursting of CO bubbles (60 wt.%); the direct dragging of solid particles also takes place, but is very limited – if sufficient operating cautions are taken.

The residue is carried from the oven along with gases from scrap smelting and steel refining operations and air admitted to the post-combustion and cooling step. To avoid dispersion of particulate matter in the atmosphere, restraint systems, typically consisting of bag filters, are installed; the particulate matter retained on filters of the emissions treatment system is the EAFD.

The EAFD composition is strongly dependent on the composition of scrap inputs, of its formation mechanism and some specific parameters of each plant. Thus, EAFD is the end result of a series of phenomena that occur since its birth in the oven, through the successive physical and chemical equilibria by which it passes during its trajectory until it is captured by the dedusting system. These facts define its mineral and chemical composition. According to Nyirenda [2], the dust has an extremely variable composition, and may have not only changes from one day to another, but from one heat to another, within the same plant.

Also in this context, it can be stated that at a given point in time, inside the oven, conditions which can produce different dust types may coexist due to dissimilarities in chemical composition of the charge inside the furnace in one and another point.

Due to the existence of galvanized scrap, temperatures and pressures used, compounds like zincite (ZnO) and franklinite ($ZnFe_2O_4$) are generated; iron (apart from franklinite) is present in EAFD mostly as magnetite [3]; yet several other elements are also present. Accordingly, EAFD is considered a hazardous waste in most industrialized countries since it fails the toxicity test for lead, cadmium and chromium [4].

1.2 Recycling or Disposal?

Depending on the scale of production, significant amounts of EAFD can be generated by the plant that should be either ‘recycled’ (with recovery of valuable metals and co-processing) or conducted to the waste heaps. In the last case, costs for disposal are not negligible (especially when considering the transportation costs to the place of end disposal). In addition to the rising costs of disposal, in Brazil there is a responsibility on the waste by the generator for any environmental impact – a fact that has driven mills to seek other ways of managing the EAFD.

With regard to material recycling, over time many processes (mainly focused on the recovery of zinc) have been developed. These processes can have a hydrometallurgical or pyrometallurgical basis, or both. However, only the so called ‘HTMR’ (High Temperature Metals Recovery), based on pyrometallurgical techniques, have achieved commercial success, most of them being variations of the Waelz process. This process type is currently processing more than 80% of the

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volume of all the recycled EAFD worldwide [5]. The Waelz process is operated in Brazil by Votorantim Metals, whose plant is located in Juiz de Fora – MG.

In pyrometallurgical processing, EAFD is put in the presence of carbonaceous materials to react. Thus, zinc oxides (zincite and franklinite) are reduced to metallic zinc, and are simultaneously vaporized at process temperatures – usually around 1200°C. This happens because the reduction temperature is higher than that of the boiling point of zinc.

Among the main reasons for the greater success of HTMR processes are, first, their ability to treat equally well both zincite and franklinite; secondly, they are more tolerant to usual (low level) EAFD contaminants, such as chlorides. These compounds are volatilized at temperatures of the reduction process and concentrate in the crude zinc oxide produced, from where they can be removed by leaching before the zinc oxide recovery. One of the disadvantages of the pyrometallurgical EAFD recycling processes, as the Waelz process, is the high investment required for their installation and operation. Because of this, the scale of production should be high enough to maintain profitability. This brings the focus of operations in certain places, which is not a favorable factor for plants located in more distant points. Another issue with the Waelz process is that it leaves a residue called ‘Waelz *slag* (or clinker)’ containing, among other, the oxides of iron and calcium.

Although HTMR processes can cope with EAFD of low zinc content, the greater amount of inert material increases energy consumption. This usually brings a higher price charged by the recycling company for the processing of this type of dust. Therefore, in most cases, the recycling of the dust of less than 15 wt. % zinc content is impaired.

In developing countries like Brazil, this problem is usually found because the amount of galvanizing steel scrap is still low in some plants. In this case, Takano *et al.* [6] suggest an EAFD *recirculation* or ‘*recycling*’ step in the electric arc furnace before a zinc recovery process can be applied to the waste.

On the other hand, hydrometallurgical processes are likely to be implemented with lower investment (no need of complex reactors and materials) and can operate at lower production scales. However, their biggest disadvantage is the difficulty of processing the zinc contained in franklinite ($ZnFe_2O_4$) – which is quite stable and usually requires a prior pyrometallurgical step or implies in more severe processing conditions [4]. This fact brings several drawbacks and costs to hydrometallurgical processes, imposing a lower efficiency in the recovery of zinc. Conversely, for the reasons given above, it can be assumed that EAFD with greater amount of zinc in the form of zincite (ZnO), could be favored if processed by hydrometallurgical routes.

1.3 Recycling: What is Missing?

Among the objectives of the steel industry, within a safe operation, is the production of high quality steel products. This should be achieved through efficient equipment operation – fact which includes a high productivity as well as preservation of equipment integrity. When the topic shifts to environmental management, objectives are not as clear. An aspect to be sought, which still does not have the same importance as ‘avoid waste production’ or ‘reduce the amount of waste’, is to drive a process aiming to get a ‘waste with better quality’ – i.e., more suitable for further processing. How to merge this with the objectives of the steel industry is a challenge that remains to be demonstrated – producing high quality steel with a ‘reduced amount of waste’ is already in itself a very complex objective.

Within the frame of environmental management activities, one can say that a better degree of understanding on the mechanisms of EAFD generation and of its minerals composition can assist in making decisions about the development of proposals able to improve dust management. This, however, goes much further than the 'generated amount reduction' policy.

Considering EAFD that is collected on plants outside the reach of the Waelz process, the hydrometallurgical processing can be a good alternative only if a greater amount of Zn is contained in zincite. This is almost the same as to set a focus on the production of a particular mineral species. Obviously, when taking this option in account, the environmental manager must allow a reasonable degree of uncertainty with respect both to composition and mineralogy of the EAFD; among the difficulties are: successful scrap management and existence of 'local equilibria' inside the EAF.

Thus, aim of this study is to perform a theoretical analysis of the compounds containing iron, oxygen and zinc found in EAFD with respect to the conditions under which they can take part in the thermodynamic equilibrium states. Special emphasis is given to ZnO formation – the compound which can promote the utilization of the hydrometallurgical route for the recovery of zinc from EAFD.

Complementing the work, a general classification of dusts based on their chemical composition is suggested and, on that basis, different EAFD mentioned in the literature are analyzed.

2 METHODOLOGY

The thermodynamic equilibrium states for the Fe-O-Zn system under study in the present work were determined using the *Equilib* module of *FactSage* software (version 6.4) – the software is described elsewhere by Bale *et al.* [7].

The following databases were employed:

1. FToxid - FACT oxide compounds and solutions;
2. FactPS - FactSage general compound database.

All analyses were conducted at fixed O_2 partial pressures. This means that all equilibria are of the type generated by setting the oxygen *chemical potential* on the system (usually this is done by defining the chemical composition of the system).

3 THEORETICAL CONSIDERATIONS

Up to now, many investigations in this field were carried out regarding the understanding of the chemical nature of the EAFD. Many other tried to understand the *morphology* and *mineralogy* of the particles contained in the dust. This analysis, unlike the general trend, deals with the EAFD under the point of view of chemical thermodynamics – *i.e.* it deals with the phases present in the equilibrium state of the system. However, it must be bear in mind that a process cannot be represented by a single (or a multiplicity of) state(s) of equilibrium – especially a batch process like that of the EAF steel production.

Only the three most abundant elements on EAFD – namely: iron, oxygen and zinc – will be considered here for a fundamental thermodynamic, systematic, analysis. Iron and oxygen have obvious sources, the element zinc is coming with galvanized scrap (used for corrosion control of the steel parts).

Any analysis considering only few components, *i.e.* made for a system which is far simpler than the real one – like the intricate situation existing inside an EAF –, is subject to error or divergence from reality. Yet, simplicity of the model is frequently

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essential for clarity and understanding of complex problems. This is also true for thermodynamics, that is, the knowledge of the phase relations in lower order (binary and ternary) systems is often crucial for a better understanding of higher order (quaternary, quinary etc.) systems.

In terms of *phases* – under the physical and chemical conditions of the present study – the system is relatively straightforward and can display in the equilibrium state only hematite, spinel (for *spinel*, see considerations made later in this text), wustite, zincite, slag (*liquid phase* or *liquid oxide* in most metallurgical texts), plus the metallic (liquid and solid) phases of the Fe-Zn-O system.

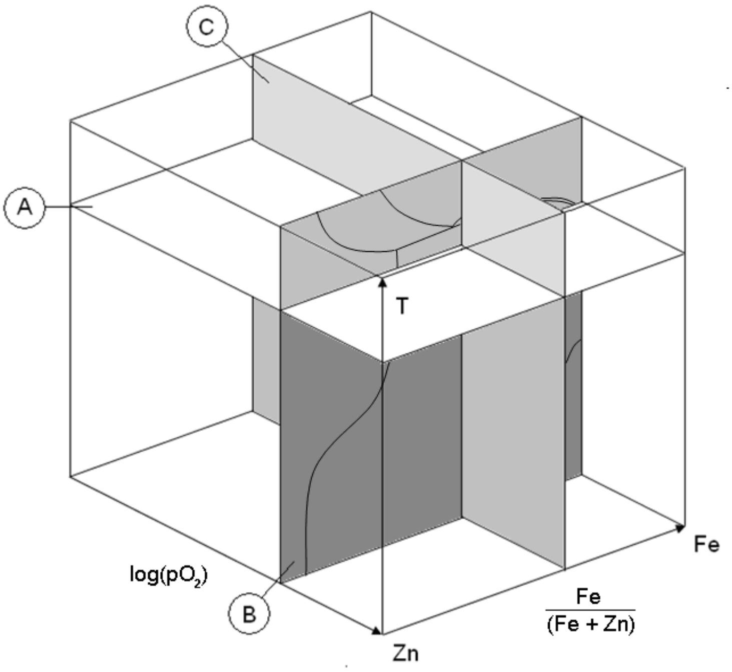

Figure 1. Schematic representation showing the three possible sections for phase diagrams within the Fe-Zn-O system; the planes correspond to: (A) *isothermal*, (B) *isobaric* and (C) *isophlet* section for a fixed Fe/(Fe + Zn) mass fraction; the three coordinate axis are: temperature; logarithm of oxygen partial pressure and composition.

These phases form extensive solid and liquid solutions which are stable over a wide range of compositions, temperatures and oxygen partial pressures. Accurate information on the stability of the abovementioned phases is required, in order to establish a thermodynamic basis for an advanced understanding of the EAFD – not only in terms of phases, but also its mineralogy and genesis.

For the ternary system Fe-Zn-O, a schematic representation in Figure 1 shows three fundamental types of phase diagrams which can be constructed: isothermic (plane A), isobaric (plane B) isophlet for the fixed composition in terms of Fe / (Fe + Zn) mass fraction (plane C). Due to the nature of the present analysis, in this work only diagrams of type B will be constructed.

4 RESULTS AND DISCUSSION

4.1 Thermodynamics of the Fe-O-Zn System

The stable phases present in thermodynamic equilibrium states for temperatures ranging between 800 and 1700ºC and a variable system composition – Fe weight

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fraction (Fe/(Fe+Zn)) varying from zero to unity – were determined for some selected oxygen partial pressures (all diagrams in this work are of type 'B', in Figure 1), in order to get a better understanding of the Fe-Zn-O system.

Since EAF is an *open* furnace, equilibrium phase diagram for a fixed oxygen partial pressure of 0.21 [atm] was determined first, Figure 2a.

Hematite, spinel, zincite, and liquid slag are the four oxide phases present in thermodynamic equilibrium states at this oxygen pressure. The liquid slag phase forms at a wide Fe/(Fe + Zn) range, just for temperatures above 1600ºC, while the stability field of hematite, Fe_2O_3, at Fe/(Fe + Zn) = 1, is a line, stable at lower temperatures only. Zincite and spinel are phase fields on the left and right portions of the phase diagram, respectively.

Zincite is a solid solution of Zn, O and Fe, yet, it is represented normally as the stoichiometric compound ZnO – in this form (at the left border of the diagram) it is the oxide with the highest Zn content in the Fe-O-Zn system. The iron increases to the right of the phase field and the highest Fe content of this oxide is not constant, but rises with the increase in temperature). Zincite can be found in equilibrium states within the full temperature range encompassed by this work.

Spinel is a phase that, from the stand point of mineralogy, spans from $ZnFe_2O_4$ (franklinite or 'zinc ferrite') up to magnetite, Fe_3O_4, at Fe/(Fe + Zn) = 1. From the curved shape of the left borderline, one can observe that franklinite is *not* a true stoichiometric compound; instead, the Zinc content in this mineral experiences a slight increase as the temperature drops – for instance, it shows ~23 wt.% Zn, at 1600ºC but about 27 wt.% Zn at 800ºC.

The rest of the diagram area is filled with 'two phase mixture' fields. In the central portion of the diagram, the mixture is composed of zincite and spinel. In order to have zincite phase taking part of the equilibrium state, Fe/(Fe + Zn) must be smaller than about 63 wt.% (this threshold value is valid for the lower temperature range only). In the right, the mixture is composed of hematite plus spinel.

The diagram also confirms that zincite and hematite *cannot* be in equilibrium. Experimentally, however, their presence in the same EAFD sample is not uncommon; this might be seen as a clear sign of the coexistence of 'multiple equilibria' (or 'local equilibria') sites inside the EAF. For instance, near a piece of galvanized scrap, ZnO could form – while, at a certain distance from that point, from the ferrous vapors evolved, only 'pure' iron oxides may form.

According to Guézennec *et al*. [1], large CO bubbles may form within the metal bath or the slag as a result of processes that occur naturally in the furnace or are the result of intentional operation (steel decarburization and slag foaming). Thus, one can assume that locally an atmosphere may exist whose partial pressure of O_2 is less than 0.21 [atm]. Moreover, according to these authors, bubble burst at the liquid steel surface is claimed to be the principal source of EAFD emission.

The possible influence of this low O_2 pressure in the equilibrium states of the Fe-Zn-O system was evaluated next. In order to do this, two additional equilibrium diagrams were effected, each one under a given fixed oxygen pressure: $pO_2 = 1x10^{-4}$ (Figure 2b) and $pO_2 = 1x10^{-7}$ [atm] (Figure 2c).

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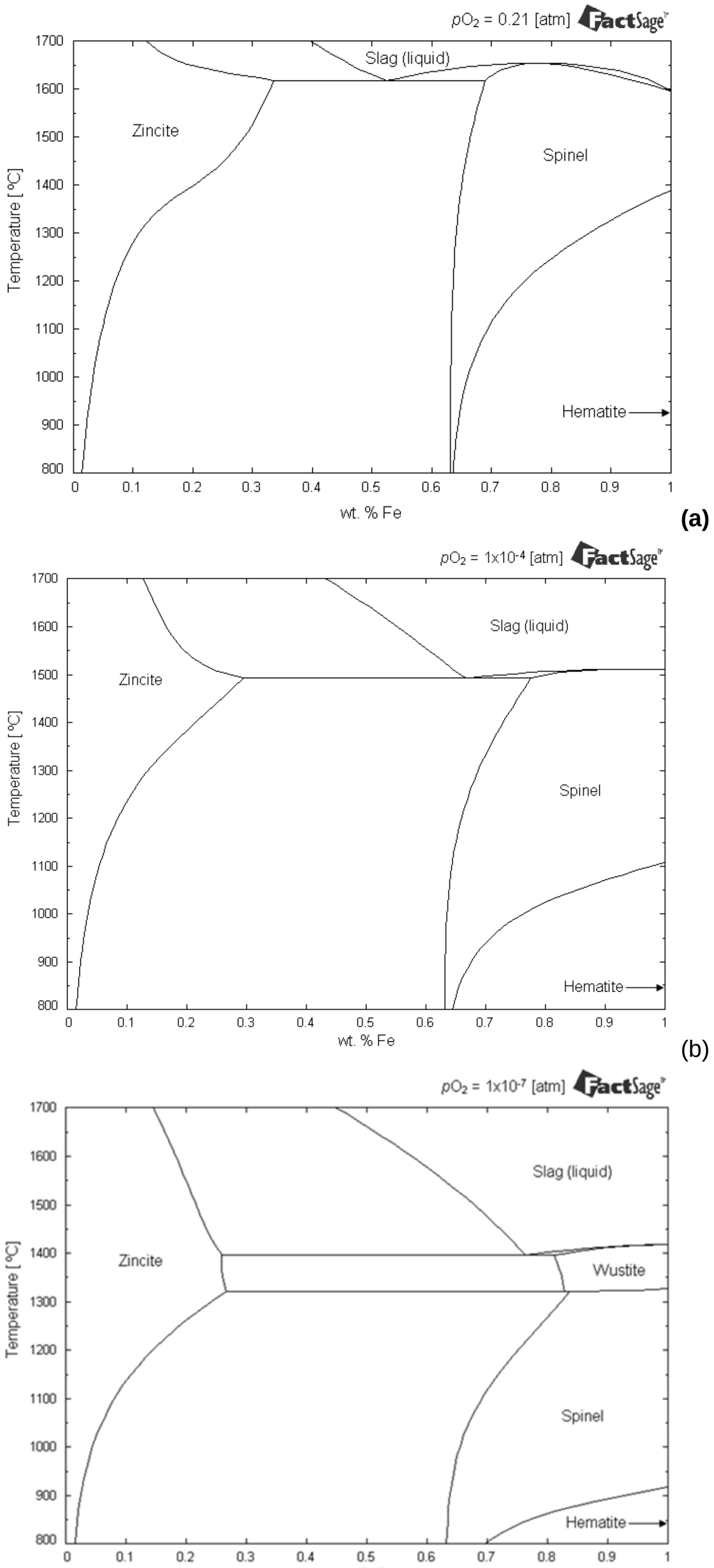

Figure 2. Fe-Zn-O phase diagram at pO_2 = 0.21 (a), 1x10^{-4} (b) and 1x10^{-7} [atm] (c) displaying stable phases as a function of temperature and mass fraction Fe/(Fe + Zn); only *single phase fields* are labeled; stoichiometric compound hematite is represented by a vertical line on the right border of the diagram.

Results for the three cases observed in sequence show that there is a lowering of the eutectic temperature, from 1600 to ~1400 °C, with the decreasing pressure. This means an increasing stability of the liquid slag phase.

There is also a more marked variation in the Fe content in franklinite as a function of the temperature, for the decreasing oxygen pressures. And this can be noticed by the slope of the line from the left boundary of the spinel phase, which is becoming less steeply as the temperature rises. This phenomenon has as an end result the fact that the ZnO phase becomes more likely to be found at equilibrium.

A new feature appears in the diagram corresponding to the partial O_2 pressure equal to a $1x10^{-7}$ [atm]: the reducing conditions are sufficient to cause the appearance of wustite solid solution phase.

If these low oxygen pressures come to establish themselves within the EAF and if they can affect the dust that is being formed, remain to be proven.

4.2 Alternative EAFD Treatment

Considering EAFD which is collected on plants outside the area covered economically by a Waelz-process operating plant, plus the ‘low Zn content’ dust, the processing of EAFD by hydrometallurgical processes becomes credible. However, as mentioned above, the importance of the presence of zinc metal in zincite phase is clear for a good usage of these processes for the recovery of Zn contained in the dust.

For this, according to the equilibrium phase diagrams calculated in this work, a Zn concentration much greater than the threshold value of 37 wt.% Zn on Zn/(Zn + Fe) is required (that figure is the Zn mass fraction in franklinite at a low temperature). Or, conversely, a Fe / (Fe + Zn) lower than 63 wt.% Fe.

An analysis, based on data provided by the literature, is presented in Table I, and shows values for both Zn and Fe mass fractions. In this table the EAFD are classified in SZ or ZH; SZ dusts are those having Zn/(Zn + Fe) values higher than 37 wt.%. This classification can be seen as a generic and preliminary criterion, indicative of suitability for hydrometallurgical treatment for zinc recovery.

From the table, something less than 50% of the dust fall within the SZ class – and, among them, one is close to the limiting value suggested.

As this result suggest, a hydrometallurgical processing can be a good alternative only when considering an enrichment of Zn in EADF, for this will increase the amount of Zn contained in the zincite phase.

As suggested by Takano *et al.* [6] and tested by Buzin *et al.* [8-10], an EAFD *recirculation* or ‘*recycling*’ step in the electric arc furnace before a zinc recovery process could be a possible treatment to be applied to the waste.

An adverse factor which comes about with the reintroduction of EAFD into the EAF must be taken into consideration, which is the concentration of undesirable impurities like chlorides and other metals such as cadmium and lead. This tendency, however, can be controlled by monitoring and, if appropriate, by applying a pre-treatment on the EAFD before the recirculation. More studies are currently being undertaken to explore these possibilities.

In this same table, the joint concentration of iron and zinc (Fe + Zn) found in different EAFD samples can also be seen. They make up together at least 50 wt.% of the mass of each sample. Further, ‘other’ (elements) found in the dust reach at most the mark of ~1/4 of the mass of the samples.

Table 1. EAFD classification as a function of Fe/(Fe + Zn) mass fraction; Spinel-hematitic – SH; Spinel-zincitic – SZ, and, Spinel only – S (all figures given in wt.%) [1-10]

Ref.	Fe	Zn	O	Other	(Fe + Zn)	Fe/(Fe + Zn)	Zn/(Fe + Zn)	Class
Zabett [11]	32.6	21.6	27.45	18.35	54.2	60.1	39.9	SZ
Tsubouchi [12]	24.9	26.1	26.49	22.51	51.0	48.8	51.2	SZ
Tsubouchi [12]	41.4	10.8	31.24	16.56	52.2	79.3	20.7	SH
Tsubouchi [12]	24.4	35.9	14.2	25.5	60.3	40.4	59.6	SZ
Jeong Yoo [13]	37.46	22.03	27.77	12.74	59.49	63.0	37.0	S
Bruckard [14]	27.1	23.1	29.22	20.58	50.2	54.0	46.0	SZ
Machado 1[5]	48.96	9.24	-	-	58.2	84.1	15.9	SH
Rizzi [16]	38.9	11.7	-	18.45	50.6	76.9	23.1	SH
Sófilic [17]	45.23	5.24	-	18.73	50.47	89.6	10.4	SH
Martins [18]	36.46	28.47	19.62	15.45	64.93	56.2	43.8	SZ
Vargas [19]	42.0	13.3	-	15.82	55.3	75.9	24.1	SH
Telles [20]	58.3	14.2	-	-	72.5	80.4	19.6	SH

5 CONCLUSIONS

Focus of this work are *phases* and conditions for *phase generation* in EAFD by means of thermodynamic simulations, in order to shed some light into the understanding of how this complex mixture of oxides and other compounds is formed and how it behaves.

EAFD can present itself as a mixture of several phases of oxidized Fe and zinc, depending on the conditions prevailing inside and outside the furnace like composition, temperature and oxygen pressure. When conditions inside the furnace are not uniform, one must expect the EAFD being a mixture of some of these phases outside the equilibrium state (*local equilibria*).

In order to be able to recycle the material or to lower recycling costs, the recovery of zinc in EAFD by Waelz-type process has become a standard. Unfortunately it cannot be applied to all EAFD generated due to low Zn content or long distance between plant and treatment station, therefore, hydrometallurgical processes must come into action. An analysis in this work, based on data provided by the literature, introduced an EAFD classification as a generic and preliminary criterion, indicative of suitability for hydrometallurgical treatment for zinc recovery. Since most EAFD fail to pass this criterion, a preliminary Zn enrichment step must be introduced.

If feasible, as a result of the reintroduction of EAFD in the furnace, an increase of zinc content in EAFD may be obtained. This effect can translate into lower costs for disposal or recycling of this waste, because it enables hydrometallurgical process for the recovery of zinc and other metals from EAFD be performed close to the steel plant, with low capital-intensive process and lesser operating costs.

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