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THERMODYNAMIC ANALYSIS OF EAFD GENERATION USING THE Fe-O-Zn SYSTEM[1]

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Abstract

The steel production in semi-integrated mills generates several types of residues – among them, the electric arc furnace dust (EAFD). In addition to Fe and oxygen, the EAFD contains the element zinc which bears economic interests, capable to motivate its extraction. Aided by a 'computational thermodynamics' tool, theoretical considerations are made for the system Fe-O-Zn (most important elements in EADF) using the variables temperature, oxygen partial pressure and composition (Fe/(Fe+Zn) mass ratio) in order to determine which phases are stable in the conditions existing inside the electric arc furnace (EAF) during the production of steel. As a result, new phase diagrams for the systems Fe-O and Fe-O-Zn were drawn; they point to spinel as the most probable solid phase present in EAFD. Moreover, the possibility to transform franklinite into another oxide has also been addressed – this can be helpful in the hydrometallurgical processing of this waste.

Key words: EAFD; EAFD phases; Computational thermodynamics.

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1 INTRODUCTION

1.1 EAFD: Birth and Characteristics

This paper can be seen as an extension of a previous work(1) on the thermodynamic equilibrium between the components iron, oxygen and zinc aiming at the study of the electric arc furnace dust (EAFD) generation during the operation of electric arc furnaces (EAF).

Some diagrams that were only suggested in the previous article have been supplemented in this work and can now be used to clarify the existing conditions inside electric arc furnaces. These diagrams show the influence of temperature, gas composition (*oxygen* partial pressure), composition (Fe/(Fe + Zn) *mass* ratio) on the equilibrium state (stable phases) of the cited ternary system. The temperature ranged from 800 up to 1,700ºC; the oxygen partial pressure in the gas phase from $1\text{x}10^{-10}$ up to 1 atm, and the Fe to iron plus zinc mass ratio, from 0 to 1.

During the production of steel in semi-integrated plants, the generation of dust in electric arc furnace is unavoidable. This waste is constituted essentially by the solid oxides of iron; however, when using galvanized scrap in electric 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.,(2) the prevailing mechanisms of dust precursor emission appear to be the volatilization (27 wt.% of the EAFD) and the bursting of CO bubbles (60 wt.% of the EAFD); the direct dragging of solid particles takes place, but is very limited – if sufficient operating cautions are taken.

Because of raw materials, temperatures and pressures used, compounds containing *zinc* like zincite (ZnO) and franklinite – also called zinc ferrite – ($ZnFe_2O_4$) are commonly identified in EAFD. Besides franklinite, *iron* is present mostly as magnetite;(3) yet several other elements – metals and non-metals – are present. Accordingly, EAFD is considered a hazardous waste in most industrialized countries since it fails the toxicity test for *lead*, *cadmium* and *chromium*.(4)

Cost for EAFD disposal is not negligible (especially when considering the cost for transportation of hazardous waste). Alternatives to disposal have been discussed in previous studies.(5-7) The co-processing in the cement kiln during clinker production, *e.g.*, is currently underway; yet, some difficulties have been reported relating to its high chlorine content. In general, especially in industrialized countries, EAFD treatments commonly used aim mainly at the recovery of zinc (the accumulation of zinc in EAFD is related fundamentally to a higher zinc vapor pressure in relation to iron at the process temperatures).

1.2 Treatment Constraints

Several authors(4,8-10) agree to point out that, in hydrometallurgical processes designed to recover zinc from EAFD, both using alkaline or acidic milieu, the more reactive compounds, such as ZnO, do not rise obstacles for their dissolution in aqueous solution; franklinite, on the other hand, being a very stable mineral, requires harsher conditions.

Severe conditions are used in treatment processes making use of hydrochloric and sulfuric acids;(10) unluckily, they lead to greater iron dissolution – a clear drawback for these process routes.

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Xia and Pickles,(4) on the other hand, in their study of the recovery of zinc from EAFD in basic medium – a way out of the excess iron dissolution problem –, consider that processes employing a hybrid treatment – first a pyrometallurgical stage for primary *decomposition of franklinite*, followed by hydrometallurgical steps for the recovery of the nonferrous metals – may be more advantageous than the single-staged acid or alkaline waste treating processes.

In developing countries like Brazil, another problem can be added to the list. The amount of galvanizing steel is still low, ruling out the use of the major EAFD treatment technologies commercialized all over the world. In this case Takano et al.(11) suggest an EAFD *recirculation* or '*recycling*' step in the electric arc furnace before a zinc recovery process can be applied to the waste.

For proper implementation of any of these treatments, a good knowledge of the raw material is required. EAFD is commonly presented in the literature mainly using results of experimental mineralogy. This work, instead, intends to present the phases that can be present in this residue from the point of view of theoretical thermodynamics; despite of the distance between theory and experimental results, a fundamental theoretical insight is of importance in any practice or procedure, acting as a standard or reference for the material which is being processed.

2 METHODOLOGY

The state of the thermodynamic equilibrium for every system under study in the present work was determined using the *Gibbs energy minimization method* of *FactSage* software (version 6.2) – the software itself is described elsewhere by Bale et al.(12)

The following databases were employed:

- FSstel - FactSage steel intermetallic compounds and alloy solutions [2009];
- FToxid - FACT oxide compounds and solutions [2009];
- FACT53 - FactSage compound database [2009].

3 THEORETICAL CONSIDERATIONS

The three most abundant elements on EAFD – namely: Fe, Zn and oxygen – will be considered here for a fundamental, systematic, thermodynamic analysis.

Any analysis considering only few elements, *i.e.* taking into account a system that is far more simpler than a real one – like the intricate situation existing inside an EAF –, is subject to error. Yet, the very simplicity frequently is essential for clarity and understanding of complex problems. Moreover, knowledge of the phase relations in lower order (binary and ternary) systems is crucial for a better understanding of higher order (quaternary, quinary etc.) systems.

Up to now, investigations were carried out regarding mainly the *morphology* and *mineralogy* of the particles contained in EAFD. More research, including the type which is currently undertaken here, regarding *phases* and *phase genesis*, is needed.

In terms of *phases* – under the physical and chemical conditions of this study – the system is relatively straightforward and can display in the equilibrium state: hematite, spinel (for *spinel*, see considerations made later in this text), wustite, zincite, slag (*liquid phase* or *liquid oxide* in most metallurgical texts; according to the thermodynamic point of view of this text, the nomenclature *slag* suggested by the 'thermodynamic software' will be maintained here), plus the metallic (liquid and solid) phases of the Fe-Zn-O system.

These phases form extensive solid and liquid solutions and are stable over a wide range of compositions, temperatures and oxygen partial pressures. Accurate information on the stability of these phases is required in order to establish a thermodynamic basis for an advanced understanding of the EAFD mineralogy and genesis.

A preliminarily analysis, considering Fe/(Fe+Zn)=1, will be made first; the resulting system can be seen as the Fe-O binary system (zinc is absent). This introductory analysis will serve as background for a more inclusive Fe-O-Zn ternary system investigation.

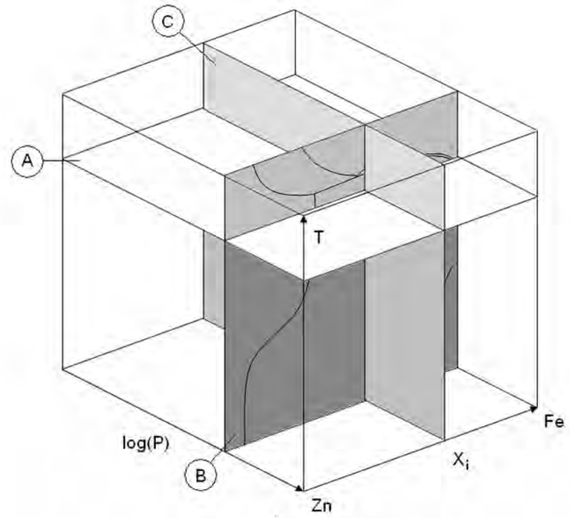

Figure 1. Schematic representation showing the general scheme used for plotting most of the phase diagrams (Fe-Zn-O system) that illustrate this work; the planes correspond to: (a) *isothermal*, (b) *isobaric* and (c) *isocomposition* sections; three coordinate axis are: temperature (T); logarithm of pressure (log(P)) and composition (in terms of Fe/(Fe-Zn) *mass* ratio).

3.1 System 'Fe-O'

The stable phases for the Fe-O-Zn system with mass ratio Fe/(Fe+Zn) = 1, in the temperature range between 800 and 1,700ºC and oxygen pressures ranging from $1x10^{-10}$ to 1 atm were determined.

The resulting Fe-O 'binary' phase diagram, Figure 2 (type 'C' diagram in Figure1), shows, in all, seven phases which can be found in some of the states of thermodynamic equilibrium – two are liquids, the other are in the solid state. Very important: *no* multiphase field exists. This comes directly from the fact that in this diagram stable phases are displayed as a function of two *thermodynamic potentials*: temperature and oxygen chemical potential (albeit, for simplicity sake, only oxygen *partial pressure* is denoted on the horizontal axis).

Even though this type of phase diagram is known since long time – *e.g.* see the works of Darken and Gurry(13-15) –, it is so far not quite well understood by researchers outside the group of termodynamicists.

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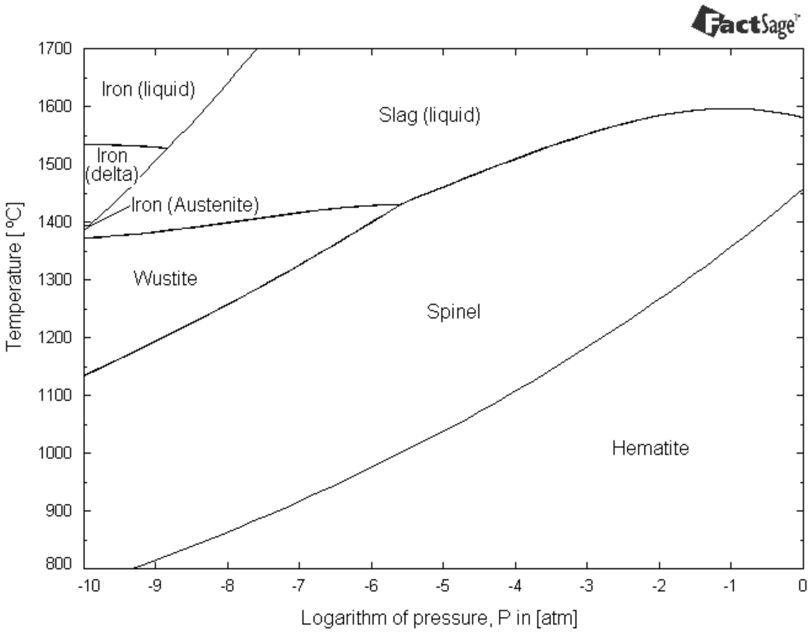

Figure 2. Fe-O phase diagram displaying stable phases as a function of temperature and logarithm of oxygen pressure (pressure in atmospheres); mass fraction Fe/(Fe + Zn) = 1; phases: Iron (liquid, delta and austenite) = liquid or solid solutions of iron with oxygen, Slag (liquid) = liquid iron oxide, Wustite = 'FeO' solid solution, Spinel = $FeO{\cdot}Fe_2O_3$ or magnetite (Fe_3O_4), Hematite = Fe_2O_3.

As it can be seen in Figure 2, the greatest fraction of the researched area is covered by four oxide phases. Hematite (Fe_2O_3) is stable over a wide range of low temperatures combined with high oxygen partial pressures. For moderate to moderately high temperatures there is a strong tendency for spinel (Fe_3O_4) to form – especially at the lower oxygen pressure side of the phase diagram. And, for the higher temperature range, the slag (liquid iron oxide) forms at any pO_2: from that in equilibrium with liquid iron – at very low oxygen partial pressures – up to an atmosphere of pure oxygen. When the temperature drops, the slag may solidify as wustite – provided pO_2 is on the lower side of the range – otherwise the slag will transform itself into spinel.

For this composition, at pO_2 = 0.21 atm (Earth atmosphere), any stable oxide that will form (and later on will be find in EAFD) is a function of *temperature* only. Given that temperature prevailing in the furnace is high, the chances of finding magnetite (spinel) constituting the majority of EAFD are large. Furthermore, if slag forms because of a still higher temperature, it will solidify, later, probably as spinel as well.

Another situation can take place hypothetically in the system: oxygen partial pressure can not be longer guaranteed. Equilibrium, then, will be driven by the available *amount* of reactants in the thermodynamic system – including *oxygen*.

In this case, products follow a different type of equilibrium diagram – also described in the previously cited papers of Darken and Gurry – interesting to say, the majority of the binary phase diagrams seen in books are of this type (these do not fit into the scheme depicted in Figure 1), Figure 3.

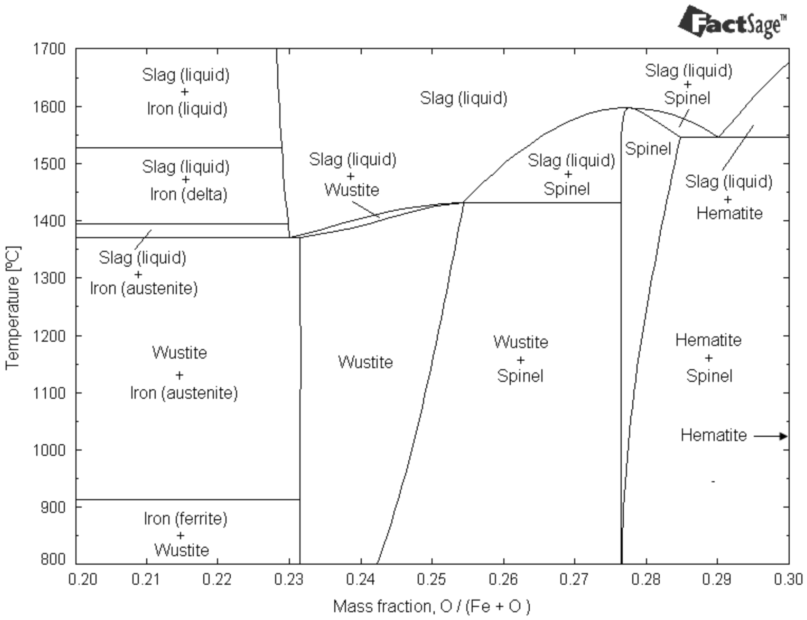

Figure 3. Fe-O phase diagram displaying stable phases as a function of temperature and mass fraction O/(Fe + O) = 0.2 - 0.3; (mass fraction Fe/(Fe + Zn) = 1).

Now, variables *temperature* and *composition* are controlling the phases that should take part in the equilibrium state. Relevant to this work is the fact that many *binary* phase fields are now present. Suppose, for instance, the amount of oxygen left to react with iron is close to 29 wt.% (of the system mass) and temperature is 1,500ºC, then, the stable phases in the system are hematite plus spinel. A biphasic EAFD will result.

For other compositions and temperatures, other single and binary phase fields – determining the type and number of phases participating in the equilibrium state – can be seen on the diagram.

3.2 System Fe-Zn-O

Given that EAF is an *open* furnace, a fixed oxygen partial pressure of 0.21 atm was used to determine the stable phases present in the thermodynamic equilibrium state for temperatures ranging between 800 and 1,700ºC, and variable composition – zinc mass fraction varying from zero to unity –, Figure 4 (type 'B' diagram, in Fig.1), in order to better understand the Fe-Zn-O system.

Hematite, spinel and slag – liquid slag phase forms just at temperatures above 1,600ºC while hematite is stable at lower temperatures only – are the three possible oxides present in the equilibrium state for systems of higher Fe/(Fe + Zn) mass ratio.

Not so obvious in the phase diagram, but worth noting, is the fact that *spinel* phase is 'magnetite' on the *right* border of the phase field and 'franklinite' on the *left* – where it establishes equilibrium with zincite. As a result, where mineralogists see *two* minerals, for thermodynamicists there is nothing more than just a *single* phase.

In addition, from the shape of the borderline marking the equilibrium ZnO-spinel, one can observe that franklinite is *not* a true stoichiometric compound. Zn content in this mineral experiences a slight increase as the temperature drops – for instance, it shows ~67 wt.% Fe, at 1600 ºC but about 63 wt.% Fe at 800ºC.

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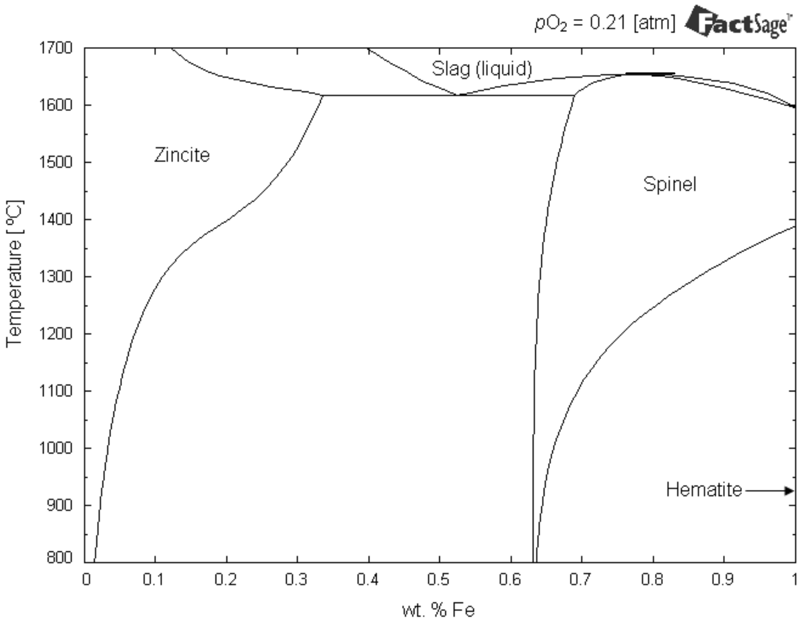

Figure 4. Fe-Zn-O phase diagram at pO_2 = 0.21 [atm] displaying stable phases as a function of temperature and mass fraction Fe/(Fe + Zn); only *single phase* fields are labeled; stoichiometric compound Hematite is represented by a vertical line on the right border of the diagram.

A feature can also be attributed to the shape of the borderline at the right of the spinel phase field. When spinel and hematite are present together in EAFD, the Zn content in spinel could be used as a sort of 'fingerprint' for determining the approximate temperature at which these species have been formed. Even though this aspect has only scientific importance, it can eventually be used as an evidence to support an experimental investigation.

Zincite – located in the opposite side of the diagram – is the oxide with the highest Zn content in the Fe-O-Zn system; it can be found in the equilibrium state within the full temperature range encompassed by this work.

The highest content in iron of this oxide rises with the increase in temperature – as it can clearly be seen in Figure 4. Thus, it may also be used as a 'temperature marker' – the same way suggested for determining the approximate temperature at which the spinel and hematite species have formed.

Moreover, in the central portion of the diagram, both phases zincite and spinel form a mixture. In order to have zincite phase taking part of the equilibrium state, though, Zn content must be greater than about 37 wt.% (this threshold value is a slight function of the temperature, decreasing by 3-4 percentage points at the highest temperature of the franklinite stability range).

Remarkably, the diagram also confirms that zincite and hematite can *not* be in equilibrium, *i.e.*, their presence in the same EAFD sample is not uncommon and can be seen as a clear sign of the coexistence of 'multiple equilibria sites' inside the EAF. A situation quite possible could be this: near a piece of galvanized scrap, ZnO will form – while, at a certain distance from that point, from the ferrous vapors evolved, only 'pure' iron oxides may form.

The diagram permits also the understanding of two possible Zn enrichment routes in the oxide phases – if this is to be used in waste treatments, in future, remains to be proven.

At first, the 'departing substance' must be an iron-rich spinel containing certain amount of Zn, formed at a temperature of 1,600ºC. If this phase is maintained a sufficient time at a temperature of 800ºC in air, it will eventually separate into 'pure' franklinite and hematite. For instance, for a starting spinel containing 90 wt.% Fe and 10 wt.% Zn, the franklinite produced will have 63 wt.% Fe and 30 wt.% Zn – which is the entire Zn content (given that hematite is composed only of iron and oxygen).

In the second of these routes, the same idea will be repeated with zincite. A numeric example may clarify the doubts. Consider a piece of ZnO mineral containing 35 wt.% Fe in equilibrium with franklinite at a temperature of 1600 ºC. When left sufficient time at 800ºC, it will decompose into franklinite and 'pure' zincite (containing not more than 2-3% Fe). Obviously, no physical phase separation will take place in view of the fact that all phases – in both cases – are in the *solid* state.

Under small oxygen pressures (depicted is pO_2 = $1x10^{-7}$ atm), the situation is somewhat different, Figure 5. While in the upper part of the temperature scale the stability field of the slag enlarges – it is stable now up to 1,400ºC – in the lower side all phase fields bellow the slag suffer a kind of 'flattening'.

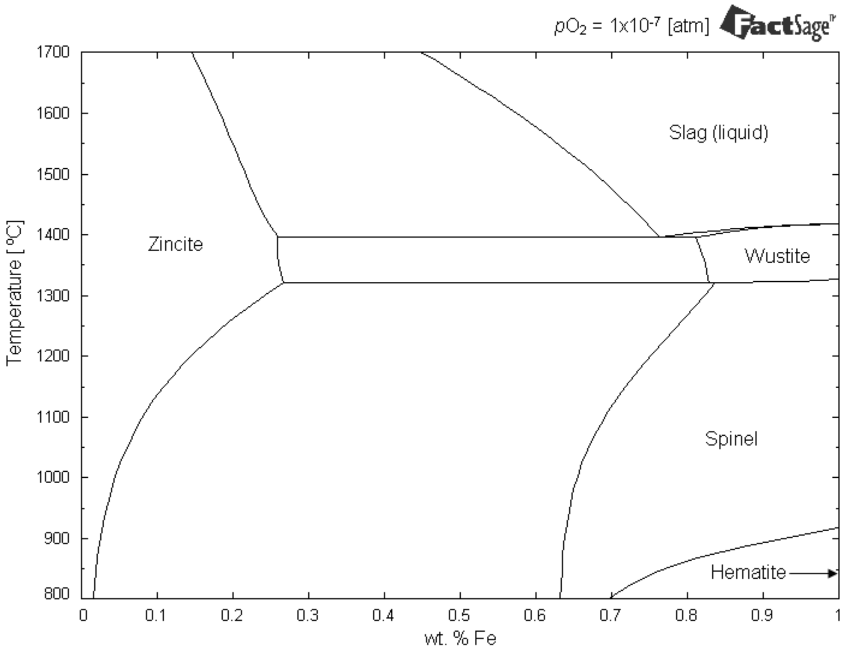

Figure 5. Fe-Zn-O phase diagram at pO_2 = $1x10^{-7}$ [atm] displaying stable phases as a function of temperature and mass fraction Fe/(Fe + Zn); only *single phase* fields are labeled; stoichiometric compound hematite is represented by a vertical line on the right border of the diagram.

For substances in form of slag, there is no possibility left to solidify as spinel phase – since wustite establishes an intermediate band between them.

The chances to find ZnO mixed with other solid phases (wustite and spinel) are larger, because, at this pressure, the stability field of zincite notably rises.

Unfortunately, the lower limit for the formation of ZnO is still high, and the raw material must contain at least ~20 wt.% Zn for this to happen.

The top view may shed some more lights into the fact that the chances of zincite formation are very favorable for a high temperature, especially if admitted in admixture with another phase, Figure 6 and 7 (type 'A' in Figure1).

It is notable, too, the shrinking of the spinel and the disappearance of the wustite phase at 1,550ºC with liquid slag filling their previous places.

To know what phases are stable at lower temperatures, two new phase equilibrium diagrams (of type 'C' in Figure1) were made having as variables pressure and temperature.

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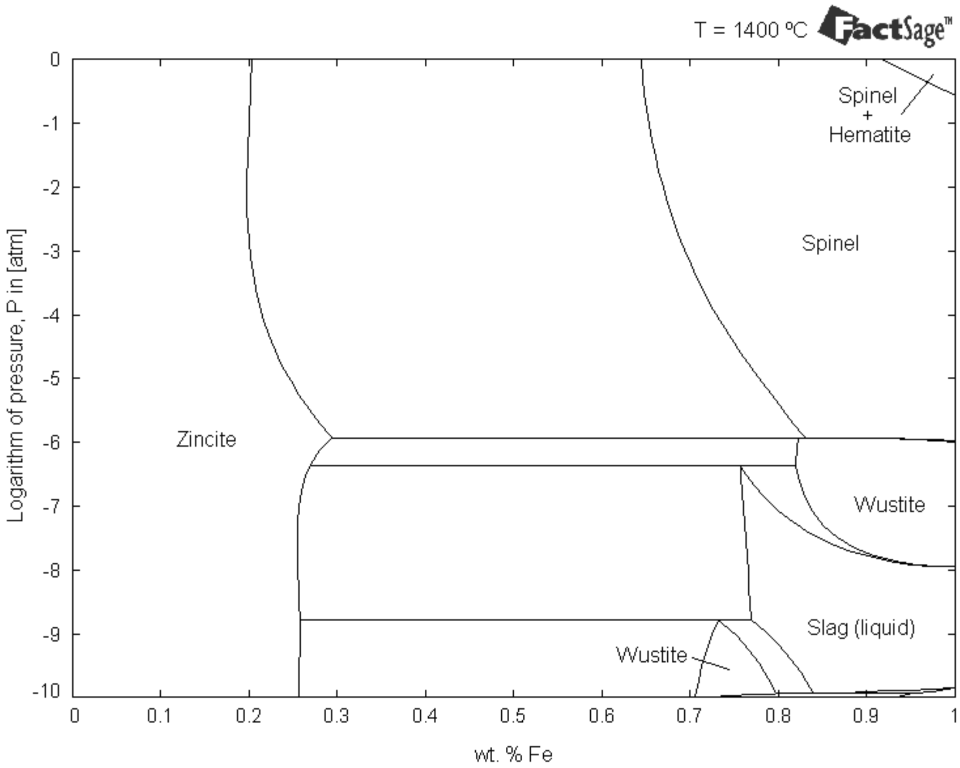

Figure 6. Fe-O-Zn phase diagram displaying stable phases as a function of wt.% Fe and logarithm of oxygen pressure (pressure in atmospheres); temperature 1,400ºC.

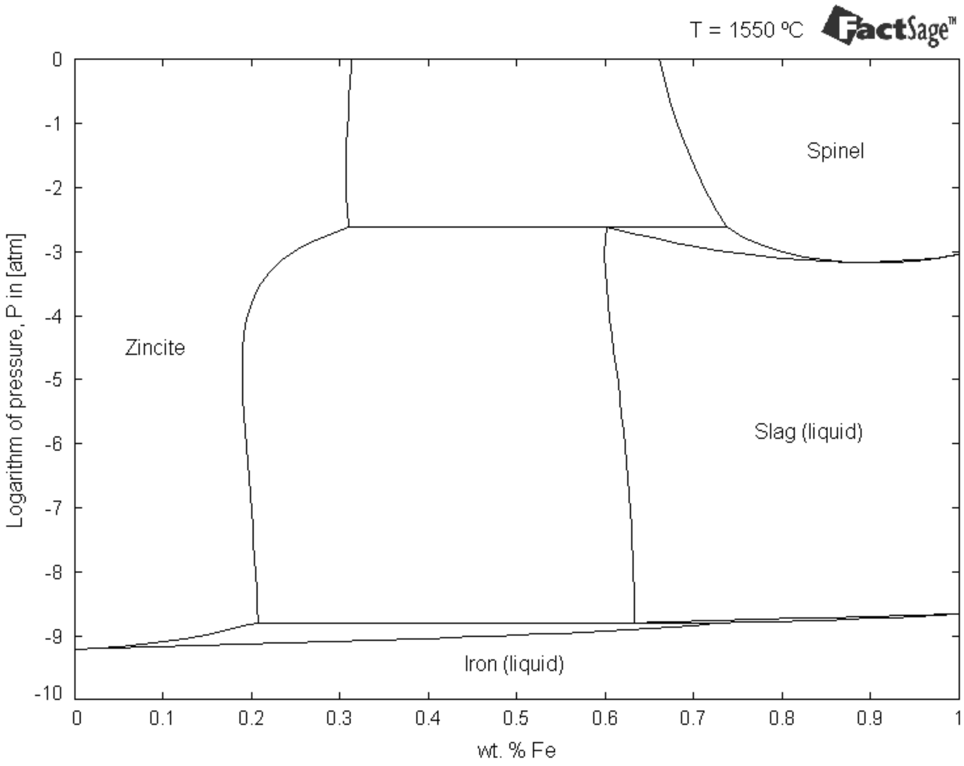

Figure 7. Fe-O-Zn phase diagram displaying stable phases as a function of wt.% Fe and logarithm of oxygen pressure (pressure in atmospheres); temperature 1.550ºC.

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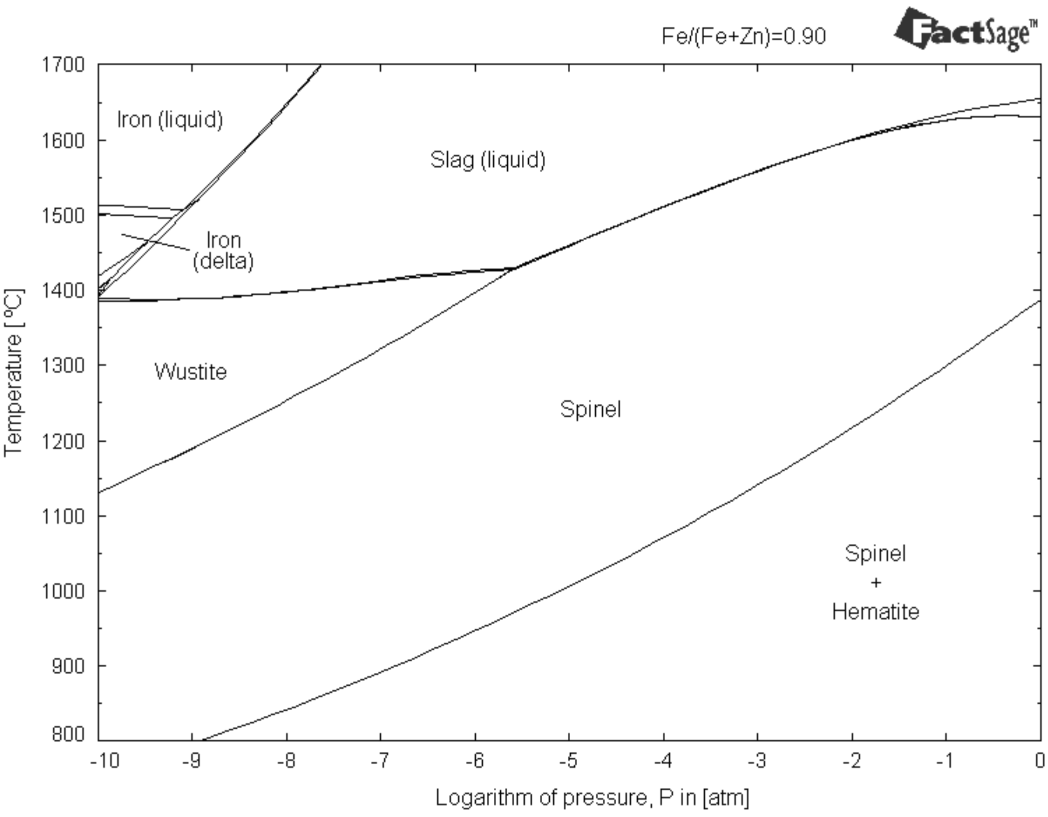

Figure 8. Fe-O-Zn phase diagram; mass fraction Fe/(Fe + Zn) = 0.9.

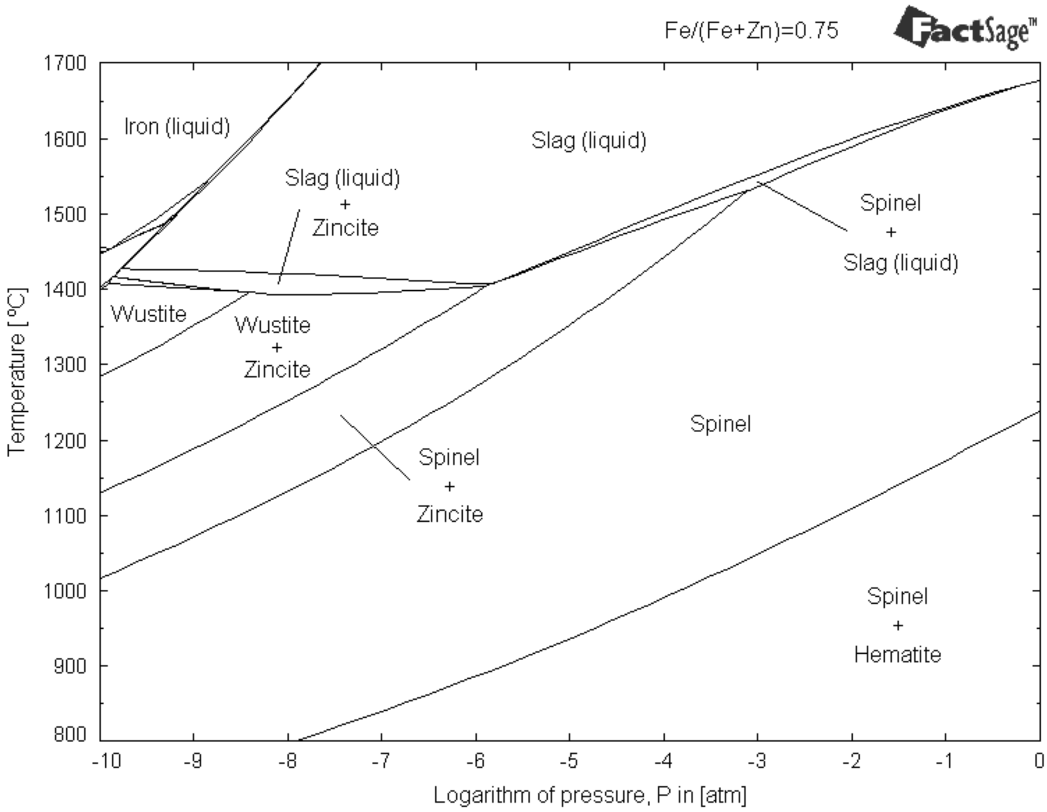

Figure 9. Fe-O-Zn phase diagram; mass fraction Fe/(Fe + Zn) = 0.75.

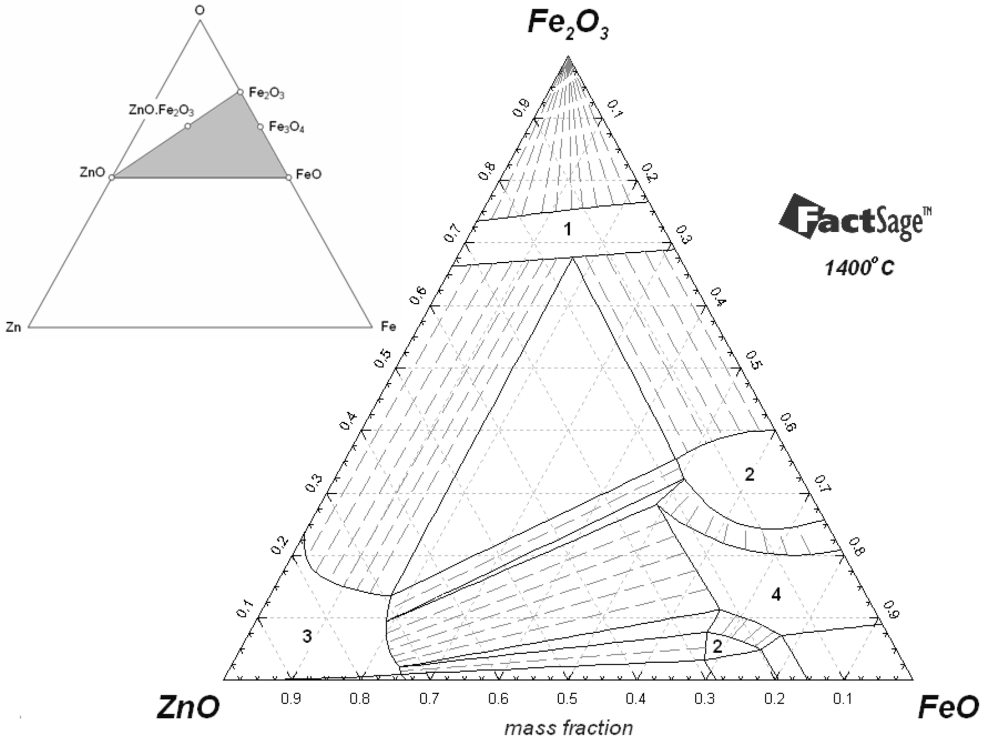

Figure 10. Pseudoternary FeO-Fe_2O_3-ZnO system; left: Fe-O-Zn (schematic); single-phase fields: 1 = spinel; 2 = wustite; 3 = zincite; 4 = slag (liquid); T = 1,400ºC.

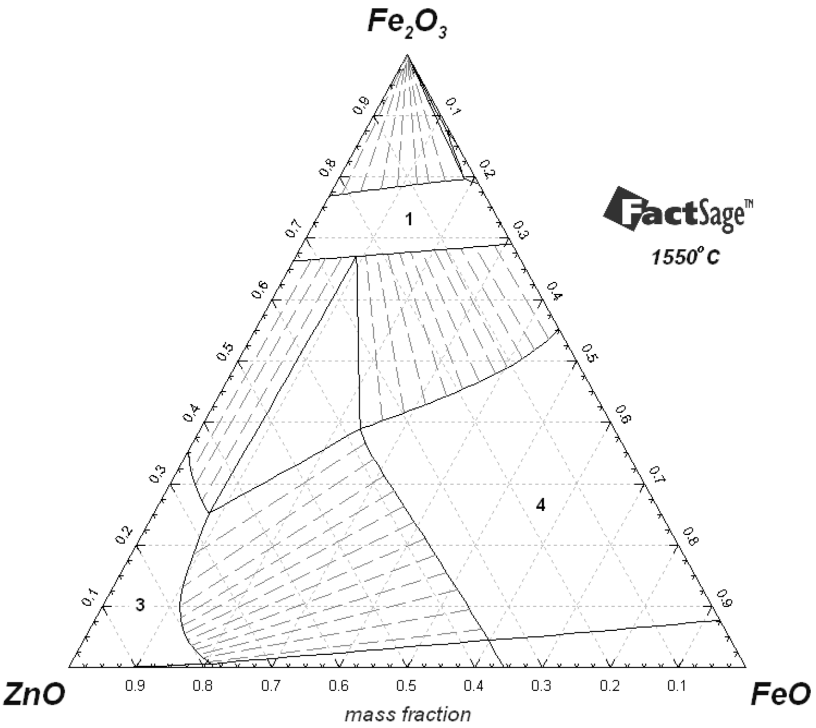

Figure 11. Pseudoternary FeO-Fe_2O_3-ZnO system; single-phase fields: 1 = spinel; 3 = zincite; 4 = slag (liquid); T = 1,550ºC.

The first, with the composition Fe/(Fe+Zn) = 0.9, Figure 8, shows much resemblance to the diagram Fe/(Fe+Zn) = 1 (0 wt.% Zn), Figure 2. The second, at weight fraction Fe/(Fe+Zn) = 0.75, Figure 9, shows at median pressure range the solid phase wustite and a mixture of spinel and zincite.

The suggestion for franklinite decomposition – by maintaining EAFD in a temperature of 1,150ºC under an atmosphere of pure nitrogen – in zincite, magnetite and hematite, as suggested by Stopić, and Friedrich,(16) can be partially seen through these diagrams.

Now, the case of equilibrium determined by the *composition* and *temperature* (disregarding pressure) is described next using the three component system: Fe_2O_3, FeO and ZnO.

In this case, it is no longer possible to construct a phase diagram like that of Figure 3 – as this is a *pseudoternary* system. It was then decided by the presentation of isothermal sections of the pseudoternary diagram. Two sections, one at temperature of 1,400ºC and the other at 1,550ºC can be seen in Figures 10 and 11, respectively.

Ternary phase fields – missing in previous diagrams – will now appear in several sectors of the phase diagram – a situation equivalent to the presence of *binary* fields in the Fe-O system. The most important of them are those which display equilibrium between spinel, ZnO and FeO. The presence of wustite in the state of thermodynamic equilibrium was hindered, in the former diagram, by oxygen chemical potential.

4 CONCLUSIONS

The thermodynamic simulation on EAFD phases/genesis – considering the limited temperature, pressure and composition ranges of the simulation in the system Fe-O-Zn – shows that primarily iron oxides – mostly spinels of iron and zinc – have great chances to be generated at the conditions existing inside the FEA, according to the following proportion of components: high Fe, low Zn content in the charge, and pO_2 = 0.21 atm.

Nevertheless, the EAFD generated can present several other phases, depending on the particular conditions of temperature, composition and pressure prevailing inside the furnace. Whether these conditions are not uniform, one must expect in the EAFD a mixture of several types of these phases.

Indeed, given that hematite and ZnO are reported in the same EAFD sample, it is undoubtedly clear that several oxygen potentials and/or temperatures do exist in the electric arc furnace (or ducts by which the EAFD crosses in its way to the baghouse), which correspond to two or more thermodynamic equilibrium conditions.

The possibility of phase change – even partially – with the conversion of an iron oxide initially formed in others (if the temperature is sufficiently high as to guarantee the supply of energy required to overcome the energy barriers presented by the activation energy of reactions or transformations), is suggested as pretreatment for the conversion of franklinite into more treatable oxides of iron and zinc. More studies should be made to explore these possibilities.

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