

EVALUATION OF PHOSPHORUS PARTITION IN STEELS USING COMPUTATIONAL THERMODYNAMIC AND COMPARISON WITH LITERATURE DATA *

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Abstract

Phosphorus content control at steel plants is frequently a challenge. This element, in general, is undesirable for steel, due to its deleterious effect in steel properties. Phosphorus partition ($L_p = \%P_{(slag)} / \%P_{(steel)}$) is widely used to analyze the dephosphorization efficiency. The thermodynamic program FactSage is largely used in the analysis of steelmaking slags. This study aims not only to evaluate the accuracy of the program FactSage v. 7.2, but also the contribution of the thermodynamics in the dephosphorization phenomena. A study about how the phosphorus partition behaves in the program calculations was done. This study is divided in two parts: the first part is an analysis of the effects that the different slag oxides, basicity, FeO and Al₂O₃, have in the L_p; the second part is a comparison between the literature values of phosphorus distribution and the results found by the program calculation. As the program considers that the system is in thermodynamic equilibrium, all the literature chosen was considered in equilibrium by its authors. For the first part it was found that the results behaved as expected by the literature, in the second part, the comparison resulted in average/good determination coefficient, R² above 0.6, for literature data.

Keywords: Dephosphorization; Phosphorus partition; Steelmaking slags.

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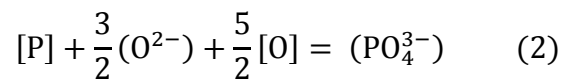
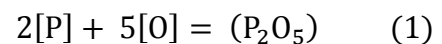
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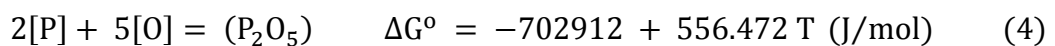
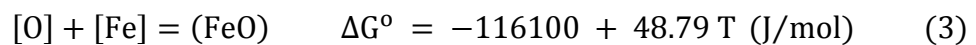
1 INTRODUCTION

Phosphorus content control at steel plants is frequently a challenge. Some steels application demand phosphorus content lower than 0.01%, where high deformation is the main requirement (pipe line, thin sheet and deep drawing steels). This element, in general, is undesirable for steel, due to its deleterious effect in steel properties [1,2]. Commonly, phosphorus is incorporated in steel through slag, iron ore or pig iron. It is known that phosphorus contents in iron ore increase according to the decrease of better ores, with lower P[3]. Therefore, it becomes more difficult to produce steel with lower phosphorus contents.

The dephosphorization (De-P) reaction can be represented by the molecular or ionic reactions given in equations 1 and 2, where [X] and (X) are species in liquid bath solution and species present in slag respectively.



Considering equations 1 and 2, phosphorus removal from iron and steel is promoted by high [P] activity, high oxygen potential [O] and high basicity (O^{2-})[4]. It is known that the direct removal of phosphorus with oxygen is not possible in steelmaking's conditions because of the instability of phosphorus pentoxide at the process temperatures. This can be better understood by equations 3 and 4[5].



These equations demonstrate that iron oxide is more stable compared to phosphorus pentoxide. The Gibbs free energy shows that FeO is more instable until temperatures close to 1000°C, and after that, stability is reversed: phosphorus pentoxide becomes more instable[6] and is reduced just after being oxidized. Thus, the P_2O_5 activity must be reduced through lime addition.

The compound formed with CaO ($3CaO.P_2O_5$) is more stable, preventing the phosphorus reversion[7]. In this way, the slags used in dephosphorization are basic, rich in $2CaO.SiO_2 - C_2S$, forming the compound $2CaO.SiO_2-3CaO.P_2O_5$ (C_2S-C_3P).

Balajiva *et al.* [8] were responsible for one of the experiments to study dephosphorization in the 40's. They used an indirect electric arc to heat metal and slag. Although they did not reach the equilibrium, they found that CaO, FeO and P_2O_5 were the main constituents that affect the dephosphorization reaction equilibrium.

Suito *et al.* studied the dephosphorization in the slag system CaO-SiO₂-MgO-FeO_x [9-13], using the phosphorus partition (L_p – equation 5) as parameter to evaluate the phosphorus removal efficiency.

$$L_p = \frac{(\%P)}{[\%P]} \quad (5)$$

Suito found that L_p increased with increasing content of CaO, and with decreasing temperature; the addition of CaF₂ influences the activity coefficient of P_2O_5 (decrease) and of FeO (increase) [9-13].

The phosphorus equilibrium reaction between carbon saturated liquid iron and MgO saturated slag with low basicity and low iron oxide content was analyzed by Lee and

Fruehan [14]. Some of their conclusions were that the $\log(L_p)$ increases with the $\log(\%FeO)$, with a slope of ≈ 2.5 . In the basicity range of 1.5–1.9, the activity coefficient of FeO decreases as the mole fraction of FeO increases.

Basu *et al.* in their research [15,16] analyzed extensively the phosphorus partition between liquid steel and $CaO-SiO_2-MgO-FeO_x-P_2O_5$ with different FeO content. In their results it was found that increasing binary basicity above 2.0 had strong effect in L_p . However, any increase beyond 2.5 results in practically no effect, and the FeO has different effects on L_p depending on the basicity and temperature.

Recent experiments about phosphorus partition were conducted by Assis *et al.* [17,18] and Tayeb *et al.* [19]. These works, regarding BOF and EAF slag respectively, had similar methodology, where they reached equilibrium after 10 hours. They were aiming at finding correlations to predict the phosphorus partition equilibrium, and in Tayeb's case, to analyze the effect of alumina in L_p .

In some of these studies [15–19] the thermodynamic simulation program FactSage was used, but only to calculate the MgO saturation solubility, not to directly calculate the phosphorus distribution between iron and slag. As new versions of the program were released recently, this study aims to evaluate the accuracy of the program FactSage v. 7.2 in the dephosphorization phenomena. In order to evaluate that, a study about how the phosphorus partition behaves in the program calculations was done. This study is divided in two parts: the first part is an analysis of the effect that the different slag oxides have in the L_p ; the second part is the comparison between the literature values of phosphorus distribution and the results found by the simulation with the literature data.

2 METHODOLOGY

All the results were obtained through the thermodynamic simulation program FactSage v.7.2. This program was created in 2001, resulting from the merging of two well-known software packages in the field of computational thermochemistry: FACT-Win (formerly F*A*C*T) and ChemSage (formerly SOLGASMIX). According to Bale *et al.* [20–22], this software is under constant development, whether upgrading or adding new database. The last change occurred in 2018 changing from v. 7.1 to 7.2.

The following databases were chosen to the simulations:

- FactPS: contains data from standard compilations as well as most of the data for those compounds which have been evaluated/optimized to be thermodynamically consistent with other databases;
- FToxid: contains data for all pure oxides and oxide solutions (solid and liquid) formed among Al_2O_3 , CaO, FeO, Fe_2O_3 , MgO, SiO_2 , and various combinations of these oxides with P_2O_5 – responsible for the slag phase.
- FTmisc: a miscellaneous database for liquid Fe with dilute solutes Al, B, Bi, C, Ca, Ce, Co, Cr, Mg, Mn, Mo, N, Nb, Ni, O, P, Pb, S, Sb, Si, Te, Ti, V, W, Zn, Zr – responsible for the steel phase.

The flowcharts presented in figure 1, resume the methodology for the two parts of this study.

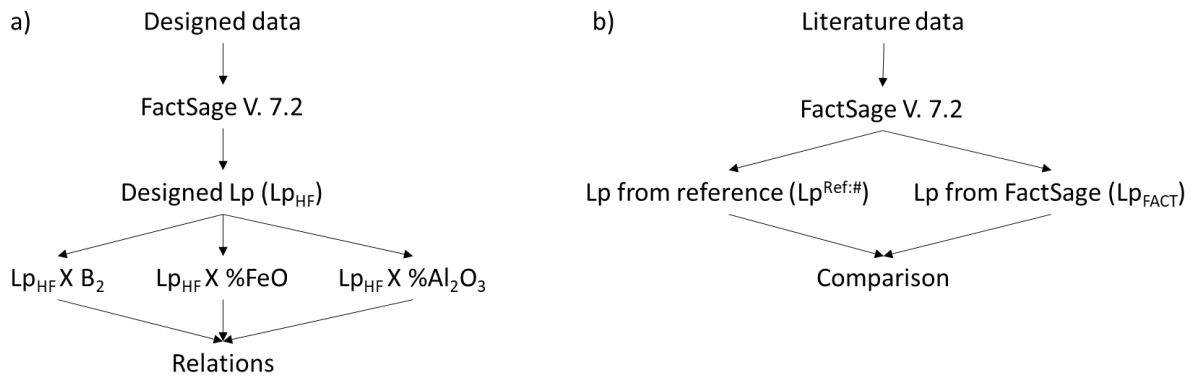


Figure 1: Flowchart for the two parts of this study: a) relation between slag oxides with the $L_{p_{HF}}$; b) comparison between L_p from literature and L_p from FactSage.

Figure 1:a is about the first part of this study. The equilibrium between a designed slag and iron was calculated for a temperature of 1600°C. In the first part of the study the slag/steel relation was 1:20. The chemical composition of these designed slags can be seen in table 1, this slag chemical composition is commonly used in electric arc furnace [30].

Table 1: Initial chemical composition of slag and metal. Values in wt.%

	Variable	B_2 (%CaO/%SiO ₂)	FeO	Al ₂ O ₃	MgO	[P]	[O ₂](ppm)
Case 1	B ₂	1 – 5	27.5	7.50	6.00	0.02	314
Case 2	FeO	1; 2; 2.75	16 – 40	7.50	6.00	0.02	314
Case 3	Al ₂ O ₃	1; 2; 2.75	27.5	0 – 12	6.00	0.02	314

Table 1 shows that 3 variables were analyzed: binary basicity (B_2) in case 1, FeO (case 2) and Al₂O₃ (case 3). In case 1, the B_2 varied from 1 to 5 and the other oxides were constant for all the simulation. Nonetheless, for the two other variables, FeO and Al₂O₃, the values of CaO and SiO₂ changed in all calculus, however the relation between those two oxides (B_2) remained constant for all the desired range. The results were obtained for 3 values of B_2 , 1, 2 and 2.75. Then the L_p ($L_{p_{HF}}$) was calculated.

In the second part (presented in figure 1: b), the phosphorus partition found by other authors and the program results were compared. As the program considers the system to be in equilibrium, all literature data considered comes from experiments that reached the equilibrium according to its authors. A summary of the literature considered is presented in table 2.

Table 2: Summary of the literature considered

Ref.	Crucible	Furnace	Equilibrium time (h)	Temperature (°C)	Slag Mass (g)	Iron Mass (g)	Crucible size (O.D X Height – mm)
[9]	MgO	Vertical resistance	3 - 6	1550 - 1600	7 – 9	25	MgO (21 X 45)
[11,13]	MgO	Vertical resistance	3 – 6	1500 – 1600	7 – 9	25	MgO (21 X 45)
[14]	MgO	Vertical resistance	4	1550 – 1580	5	5	MgO (19.1 X 25.4)
[15,16]	MgO	Horizontal tube	8	1600 – 1650	10	10	MgO (25 X 30)
[17]	MgO	Horizontal tube	10	1600	6 – 8	12 – 18	MgO (25.5 X 31.7)
[19]	MgO	Horizontal tube	10	1600	6	12	MgO (25.5 X 31.7)

Some differences between the experiments appears in table 2. There were huge differences in the equilibrium time, it varies from 3 hours [9,11,13] to 10 hours [17,19], it can indicate that some literature experiments did not reach the equilibrium. Another important data from table 2 is the slag and iron mass relation (slag/iron), as De-P is ruled by mass transport, the differences in this relation should lead to great difference in L_p .

The chemical composition of the slag used in each of the experiments of table 2 is presented in table 3.

Table 3: Slag chemical composition range of the literature considered

Ref.	CaO	SiO ₂	MgO*	FeO	Al ₂ O ₃	MnO	P ₂ O ₅
[9]	1 – 40	1 – 35	6 – 30	10 – 80	0	0	0.1 – 1.2
[11]	8 – 29	0 – 27	6 – 16	21 – 68	0	0	4 - 6
[13]	0 – 37	0 – 30	7 – 29	10 – 70	0	2 – 5	0.1 - 3
[14]	33 – 45	32 – 40	13 – 22	3 – 10	0	0	0.3 - 4
[15]	38 – 60	18 – 30	3 – 16	6 – 15	0	0	3 – 6.5
[16]	30 – 53	12 – 25	3 – 17	16 – 32	0	0	2 – 7
[17]	34 – 52	12 – 20	3 – 6	24 – 43	0	0	1.3 – 2.5
[19]	28 – 49	9 – 32	3 – 18	10 – 38	0.1 – 16	0	0.8 – 1.1

*In all references its authors considered that the experiments were saturated in MgO

It can be seen in table 3 the variation in the chemical composition of the slag of each literature considered. As expected each experiment has its own peculiarities, Tayeb [19], that wanted to analyze EAF slags, considered Al₂O₃ in his experiment, Suito [13], that studied the manganese in the slag, considered the MnO. All the others authors considered the same oxides, but with different ranges, some considered very

low CaO and SiO₂ (references [9] and [13]), others studied only low FeO content slag (references [14] and [15]). However, all of the authors considered that their experiment were saturated in MgO.

With the literature data, the L_p for each reference (L_p^{Ref}) was calculated and compared with the FactSage result, L_p^{FACT}. The relation (interpolated line) was compared to the line Y=X, to analyze if the value calculated by FactSage is higher or lower than the literature value.

2.2 Results and discussion

As this research is divided in two parts, the results will be presented separately, first the effect of the oxides in L_p^{HF}, then the comparison between L_p^{FACT} and L_p^{Ref}.

2.2.1 Oxide effects

For this first part, the effect of binary basicity, FeO and Al₂O₃ are shown in the next sections.

Basicity effect

The relation between binary basicity and the phosphorus partition (case 1 from table 1) can be seen in figure 2.

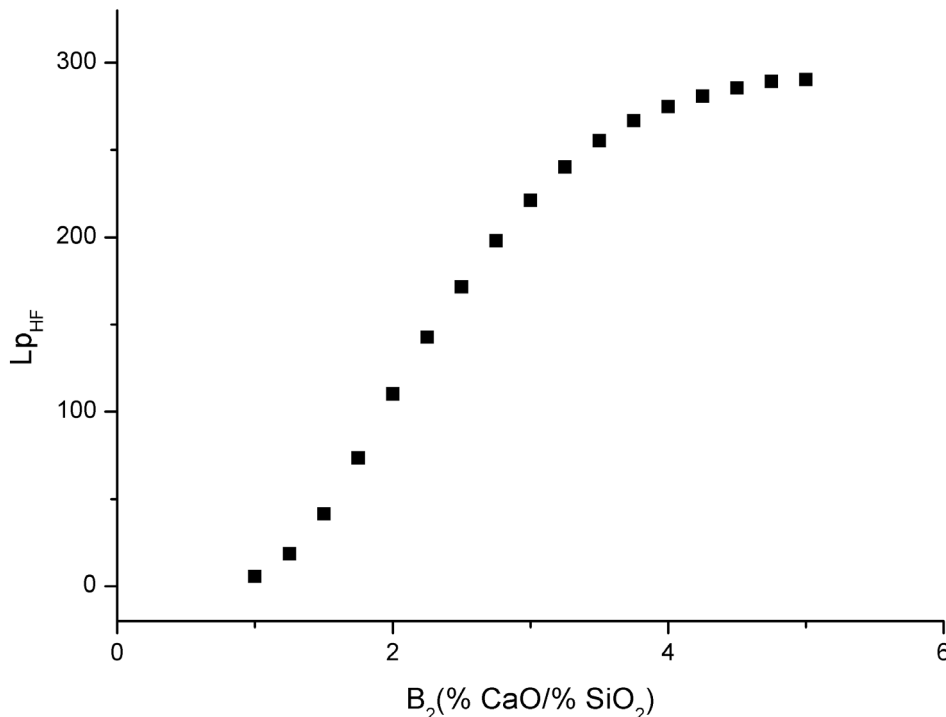


Figure 2: Relation between L_p^{HF} and binary basicity, T=1600 °C.

In figure 2 it is noted that with basicity between 1 and 2.5 the L_p^{HF} increases substantially (from 0 to 250), for values above that a slightly change in L_p^{HF} occurs and it remains almost constant for B₂ values near 5. Other authors found similar results for this relation, Basu [16] considered the same temperature but different FeO content (15 – 25%), Basu's and FactSage's results have similar L_p values. Urban [22] and Li [24] also found a similar trend, despite their L_p's values that were lower

and higher respectively. One of the reasons for the difference in L_p may have been the difference in temperature (1700°C for Urban and 1350°C for Li), as the dephosphorization reaction is very sensible to temperature.

FeO effect

Some authors [16,23,25 – 27] are in accordance that iron oxide can have either a positive or a negative effect on dephosphorization. At lower FeO content, increasing iron oxide in the slag, increases the phosphorous partition. This happens because this oxide increases the oxygen potential, and at higher levels of FeO, increases in iron oxide content decreases the L_p because it dilutes the CaO, an important oxide to form the main solid that contains phosphorus. The relation between FeO and $L_{p_{HF}}$ (case 2 from table 1) is presented in figure 2.

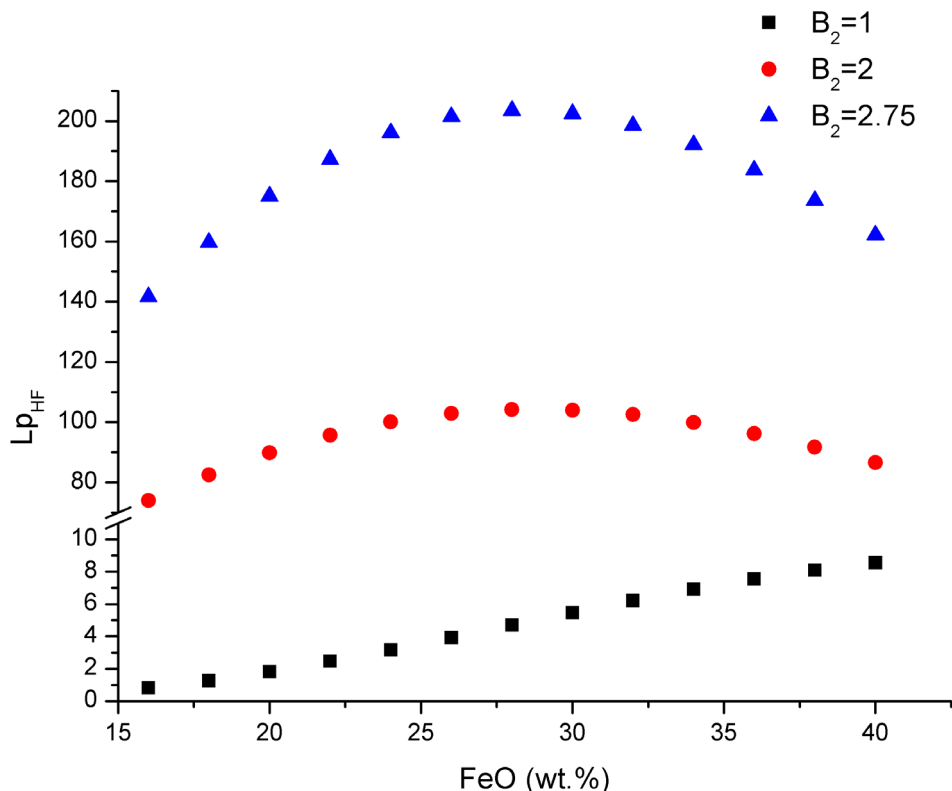


Figure 3: relation between $L_{p_{HF}}$ and the FeO content for different basicities, $T=1600^\circ\text{C}$.

The optimum value for $L_{p_{HF}}$ (figure 3) is very similar for the basicities 2 and 2.75, almost 27 % of FeO. In the case of $B_2 = 1$ the optimum FeO value was not reached. Other values of the optimum FeO content can be found in the literature, 23% [28] 17% [16] 25% [29]. The parabola behavior can be explained by the following statement: the L_p increases because of the increase in the oxygen potential (represented by FeO), and it starts to decrease due to the dilution of the basic oxides (such as CaO, reducing the formation of C_3P).

Chen [26] explains the behavior presented in figure 2 thermodynamically: when the FeO is very low, increasing FeO content would lead to a significant increase of FeO activity, and consequently increases the P participation in the slag phase. Further increases in FeO reduces the CaO content in the slag, which increases the activity coefficient of P_2O_5 ($\gamma_{P_2O_5}$) in the slag, slows down the increase of $a_{FeO}/\gamma_{P_2O_5}$, and finally the $a_{FeO}/\gamma_{P_2O_5}$ reaches a maximum point. After this critical point, increasing

FeO would decrease the $a_{FeO}/\gamma_{P_2O_5}$ in the slag, which in turn results in a decrease of the P distribution ratio.

Al₂O₃ effect

Alumina is a complex oxide that can be either a network breaker or a network former. In the CaO-SiO₂-MgO-FeO_x-Al₂O₃ slag system, alumina increases the liquid phase region [30], as it works as a network breaker, diminishing the viscosity of the slags. This effect of alumina in the slags should affect the Lp. Such relation is demonstrated in figure 4.

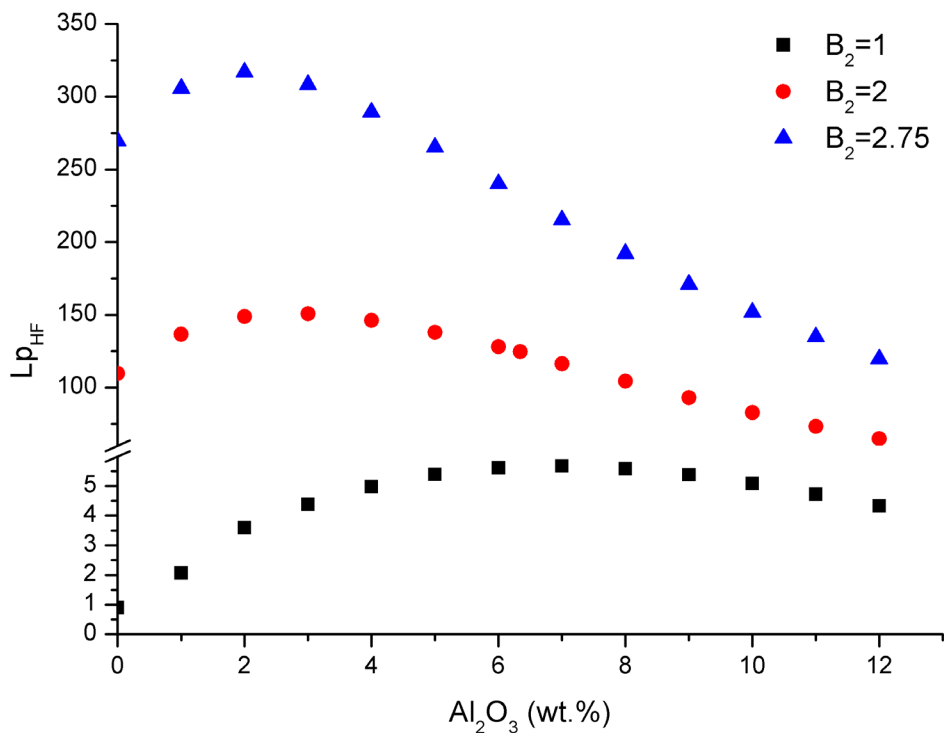


Figure 4: behavior of Lp_{HF} with the Al₂O₃ content for different basicities, T=1600°C.

According to figure 4, in higher basicities (B₂ = 2 and 2.75) there is an increase in the Lp_{HF} for low Al₂O₃ content, less than 3%. From this point of maximum Lp on, it decreased greatly, by almost 70 % (from 320 to 120 for B₂ = 2.75, and from 150 to 65 for B₂ = 2). For the lowest basicity analyzed, the Lp_{HF} remained almost the same (changing slightly) from 1 to 5, having the highest value reached in Al₂O₃ equal to 7%. This result corroborates the statement from Ouchi [31] that “a small amount of Al₂O₃ can promote the CaO dissolution into the slags”. Tayeb’s [19] found a reduction, 50%, in the phosphorus partition when the Al₂O₃ increased from 0 to 9-11%. He also says that alumina decreases the phosphorus partition by reducing the availability of free oxygen necessary for phosphorus to be incorporated in the slag as phosphate.

2.2.2 FactSage X Literature data

This section will show the results of the calculations done with the literature data. Figure 5,6,7 and 8 demonstrate how the L_p from FactSage (L_{pFACT}) correlates with the L_p from different references ($L_{pRef:\#}$). The dot lines in the figures represents the LPL and UPL (lower and upper prevision limits), dots that are outside these lines, cannot be explained by the interpolated line (solid line) and are out of the 95% confidence interval. Also, the dash line represents the relation $Y=X$: if the interpolated line is below the dash line, the value given by the software is higher than the value from the literature.

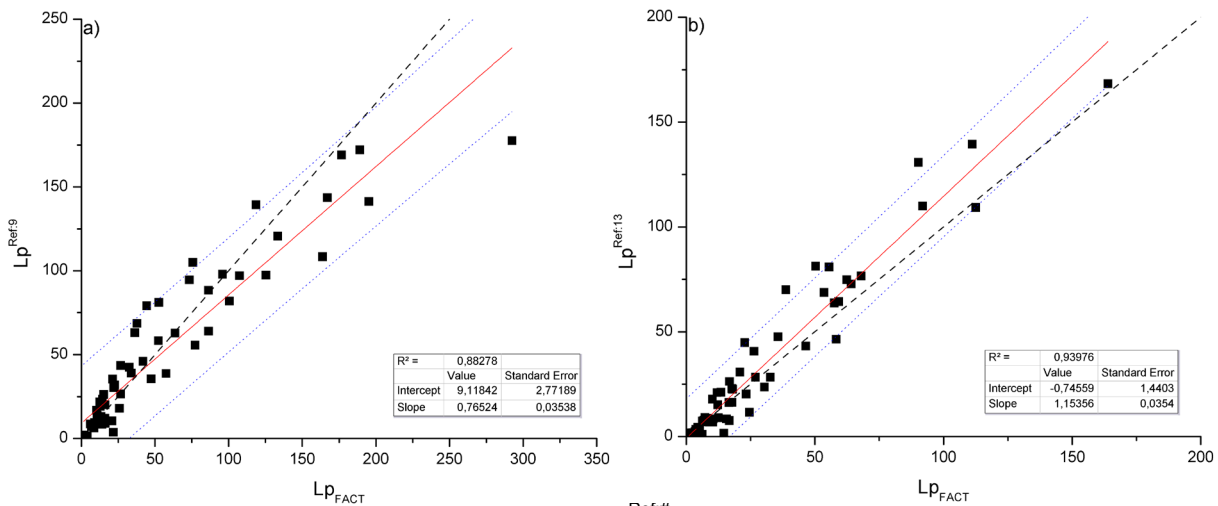


Figure 5: relation between $L_{pRef:\#}$ and L_{pFACT} ; a)Ref:9; b) Ref:13.

In figure 5 the references [9] and [13] not only had the most similar values between $L_{pRef:\#}$ and L_{pFACT} : more than 70% of the dots had a difference of 30% or less between the L_p 's values, but also, had high coefficient of determination (R^2). Even though these references had a good agreement with the program, some errors might be attributed to the **fact** that not all the heats were saturated in MgO in the FactSage calculations. However, the authors considered that all their heats were MgO saturated.

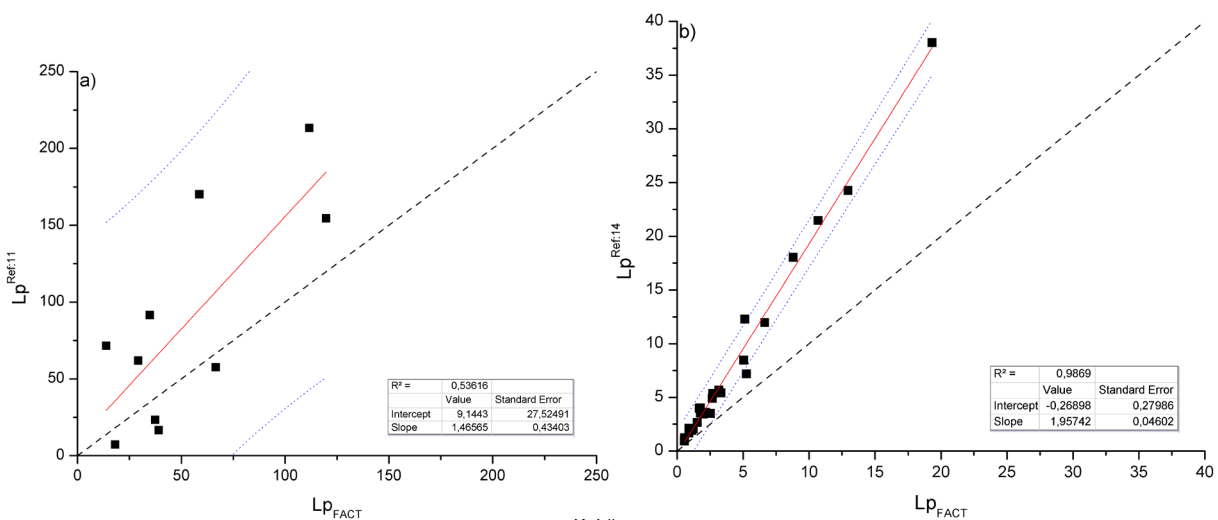


Figure 6: relation between $L_{pRef:\#}$ and L_{pFACT} ; a)Ref:11; b) Ref:14.

In figure 6: a, the R^2 was considered low ($R^2=0.53$), this reference contained huge amounts of FeO and P_2O_5 (21 – 68 % and 4 – 6% respectively), this was the only reference that have these two characteristics together. Lee and Fruehan's work [14]

showed the best correlation ($R^2=0.98$, figure 6: b). Their study was about De-P for slags with low basicity and low iron oxide content (less than 10%). Although the good correlation, there was a great difference between the Lp's. In 90% of the data the $Lp^{Ref:14}$ is 2 times higher the Lp_{FACT} .

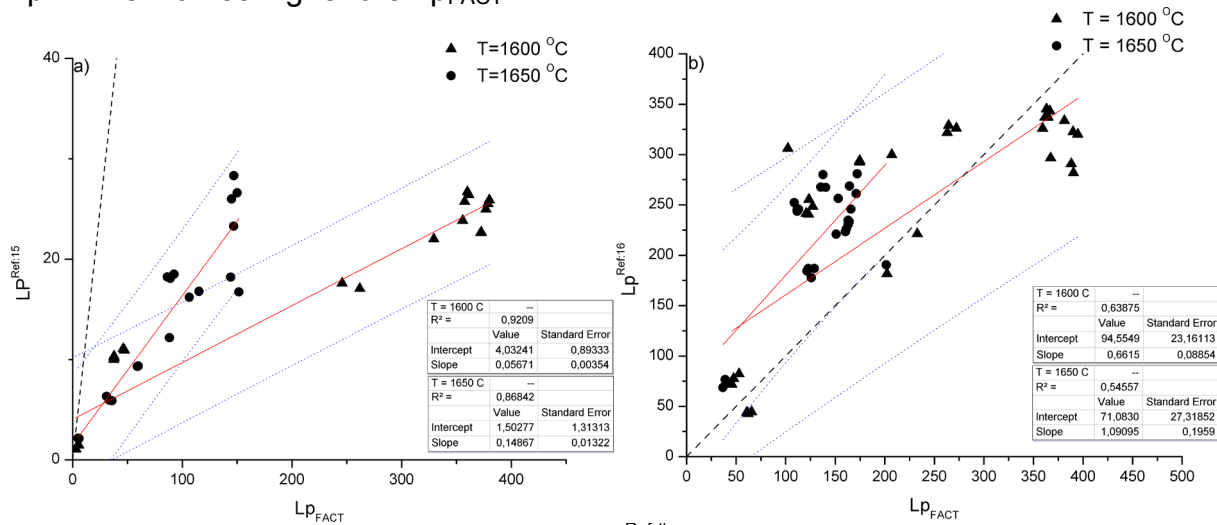


Figure 7: relation between $Lp^{Ref. \#}$ and Lp_{FACT} ; a) Ref:15; b) Ref:16.

In the comparison between FactSage and Basu's researches [15,16] (figure 7) there were differences in the results. When low FeO was considered (figure 7: a), a good correlation was found for the two temperatures ($R^2 = 0.92$ and 0.86), but when the FeO content was increased, figure 7: b), the R^2 was reduced to 0.63 and 0.54 for the same temperatures. Besides the FeO, other difference between the two literatures data is the oxygen dissolved in steel, that was not considered in their study with low FeO.

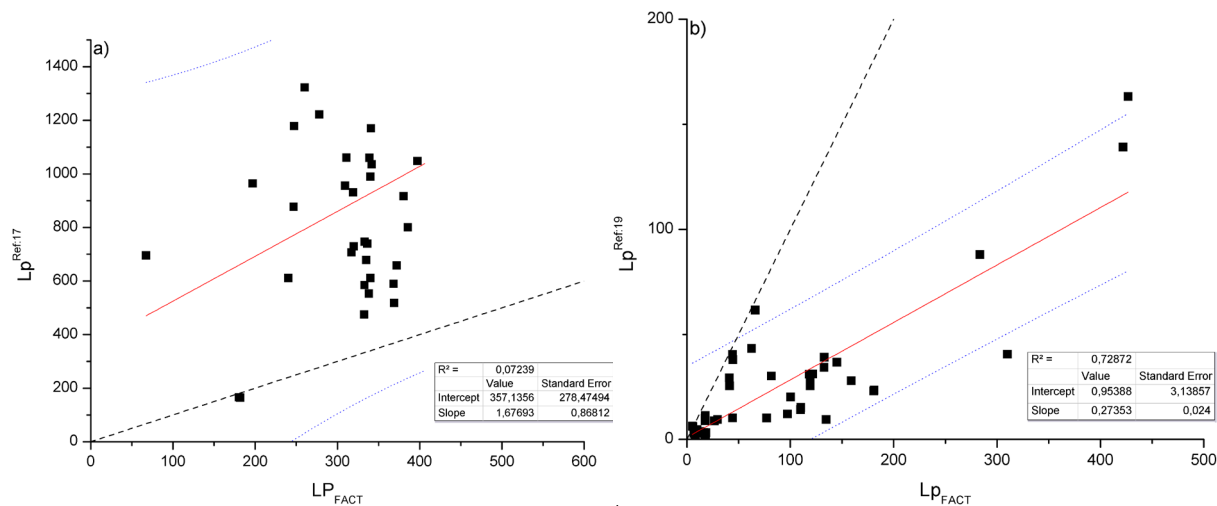


Figure 8: relation between $Lp^{Ref. \#}$ and Lp_{FACT} ; a) Ref:17; b) Ref:18.

Figure 8 shows the relation between Lp_{FACT} and the references [17] and [18]. These references had the same methodology, but very different results. The worst relation was found in reference [17] - figure 8:a – in which the values found by the authors were 3 times (or more) higher than the ones found by FactSage v. 7.2. One source of error may be connected to the fact that the authors did not specify the slag and iron mass for each experiment; as De-P is ruled by mass transport, any difference in this relation (slag/iron) should lead to great difference in Lp. Another problem was found

in the solid particles: not all their slags were saturated in MgO, indicating that the amount of slag changed during the experiment because of the crucible dissolution. The authors stated this fact in their study, and also concluded that the experimental MgO saturation was higher than the one predicted by FactSage [17,32].

3 CONCLUSION

Regarding the oxides effects in FactSage v.7.2 calculations, it can be concluded that:

- The basicity has similar trend comparing to the literature, increases L_p with basicity and for higher basicities ($B_2 > 4$) L_p remained almost constant;
- About FeO, the only difference between literature and Factsage L_p 's was the FeO optimum content, near 27% for FactSage and for literature: 23%[28], 17%[16], 25%[29], for basicities 2 and 2.75. For $B_2=1$ the optimum FeO was not reached.
- Al_2O_3 , for basicities higher than 2, has in general a deleterious effect in L_p , from 0 to 12 % of Al_2O_3 the L_p decreased by more than a half, though it showed a small increase in L_p for low alumina content.

Concerning the comparison between the L_p from literature data ($L_p^{Ref:\#}$) and FactSage's (L_{pFACT}), it can be concluded that:

- From the 8 references analyzed 4 of them ([9], [13], [14] and [15]), had a good coefficient of determination (R^2 above 0.8);
- The two references (11 and 16) that contained 20% or more of FeO and more than 2% of P_2O_5 simultaneously, had an R^2 below 0.7;
- Reference [17] was the only one that did not demonstrate any relation, $R^2=0.07$.

The main reason for these differences is that, all authors considered that their experiments reached the equilibrium, and were MgO saturated, however FactSage calculations showed that not all their slags were saturated in MgO and that they may not have reached equilibrium.

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