DEVELOPMENT OF PHOSPHORUS PARTITION MODEL USING EAF STEELMAKING DATA*

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Abstract
The research in steel dephosphorization were driven by the increasing demand for high-quality steel with fewer impurities. One of these impurities is the phosphorus. It is an undesirable element in steel, because of its deleterious effect in steel properties. In the last decades, researchers have studied dephosphorization to deduce expressions based in chemical composition and temperature. These expressions, deduced using equilibrium data, did not achieved a good correlation with industrial data. All expression can be written as a sum of a basicity term, an oxygen potential term, a temperature term and a constant of adjustment. This paper aims to create a new expression using, industrial data (slag and steel) and the results obtained by the thermodynamic program FactSage v. 7.2 to achieve a better correlation. The results given by FactSage were the Lp and viscosity of the slag, they were considered as an equilibrium term and kinetic term, resulting in another expression. Besides that, the expression will be tested using another group of industrial data. As a result, the new model had a good correlation with the industrial data R^2=0.89, and with the second industrial data, R^2 = 0.83. This fact evidences the importance of scrap load in dephosphorization**.**
Keywords: Dephosphorization; EAF Stelemaking; Steelmaking Slag.

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1 INTRODUCTION

The research in steel dephosphorization were driven by the increasing demand for high-quality steel with fewer impurities. Also, the steel industry is always searching ways to reduce costs and increase the profitable margin. These contrasting trends lead to an infinite search to obtain maximum quality using less noble raw materials.
Regarding to phosphorus, it is an undesirable element in steel, because of its deleterious effect in steel properties. Phosphorus removal (dephosphorization) can be explained by the reactions shown in equation 1 and 2:

Molecular reaction: $2[\mathrm{P}] + 5[\mathrm{O}] = (\mathrm{P_2O_5})$ (1)

Ionic Reaction: $[\mathrm{P}] + \frac{3}{2}(\mathrm{O^{2-}}) + \frac{5}{2}[\mathrm{O}] = (\mathrm{PO_4^{3-}})$ (2)

Where:
[X] = element in steel;
(X) = oxide/ion in slag.

The equilibrium quotient kp, is described in equation 3.

$$\boldsymbol{kp} = \frac{\%\boldsymbol{P_2O_5}}{[\boldsymbol{P}]^2(\%\boldsymbol{FeO})^5} \quad (3)$$

Considering equations 1 and 2, phosphorus removal from steel is promoted by high [P] activity, high oxygen potential [O] and high basicity (O^{2-}) [1]. Another important parameter that influences dephosphorization (de-P) reaction is the temperature. It is well known that phosphorus pentoxide is instable in temperatures above 1000 °C.
Thus, the P_2O_5 activity must be reduced through lime addition. The compound formed with CaO ($3CaOP_2O_5$) is more stable, preventing the phosphorus reversion [2]. As the slags used in electric arc furnace (EAF) are basic, and rich in $2CaOSiO_2$ – C_2S, they form the compound $2CaOSiO_2.3CaOP_2O_5$ ($C_2S.C_3P$).
Regarding dephosphorization, several studies have been made, but one of the first studies to found that CaO, FeO and P_2O_5 affect the de-P reaction was made by Balajiva et. al. [3]. Another important study, used the chemical composition of the experiment slag to propose an expression to the logarithm of the equilibrium quotient, log(kp) [4].
Since the study from Healy, researchers have been studying dephosphorization to deduce expressions based in chemical composition and temperature to predict the value of the equilibrium quotient. Table 1 show some mathematical models that predict the dephosphorization efficiency.
Table 1 show the following models:

- Healy's model (eq. 4), this model takes in consideration the following hypothesis: the conditions of the binary system $CaO-P_2O_5$ can be extrapolated to more complexes system without greater mistakes [4].
- Suito's model (eq.5) in this equation, Suito considered other oxides like MnO and MgO [5].
- Kunisada and Iwai in their study of phosphorus distribution in slags with Na_2O develop the model presented in eq.6 [6].
- Ide and Fruehan in 2000 [7] found differences when comparing their results with those of other authors, for this reason a new coefficient for MgO was calculated (eq. 7).

- Assis used his experimental data [8] , experimental data from Suito's study [5,9] and from Basu's research [10,11] to revise Suito's model, his model can be found in eq. 8.
- Assis et al [12] developed a new model for dephosphorization, presented in equation 9.
- Drain's model (eq. 10) [1], was developed using industrial data from 11000 heats, it can be seen that it is the only model to use [C] – carbon content of the steel.

Table 1. Mathematical model to predict dephosphorization

Eq.#	Equation	System / Conditions	Ref.
4	$logkp = 22350/T - 16.0 + 0.08\,(\%CaO)$	CaO-MgO-SiO_2-FeO_x T = 1550 – 1650°C	[4]
5	$logkp = 0.0720\,[(\%CaO) + 0.3\,(\%MgO) + 0.6(\%MnO) + 0.6\,(P_2O_5)] + 11570/T - 10.520$	CaO-MgO_{sat}-Fe_tO-SiO_2-P_2O_5-MnO T = 1550–1650 °C	[5]
6	$logkp = 0.092[(\%Na_2O) + 0.8(\%CaO) + 0.6(\%MnO) - 0.9(\%Al_2O_3)] - 3.54$	Na_2O-SiO_2-Fe_2O_3-CaO-MnO-Al_2O_3 T = 1600°C	[6]
7	$logkp = 0.0720\,[(\%CaO) + 0.15\,(\%MgO) + 0.6(\%MnO) + 0.6\,(P_2O_5)] + 11570/T - 10.50$	FeO-CaO-SiO_2-P_2O_5-MgO-MnO T = 1600 °C, B_2 = 2.5 – 3.2	[7]
8	$\log kp = 0.073[(\%CaO) + 0.148(\%MgO) + 0.8(\%P_2O_5) + 0.113(\%SiO_2)] + 11570/T - 10.403$	CaO-SiO_2-0.5<P_2O_5<5.5 -MgO-FeO T ~ 1600°C	[8]
9	$logkp = 0.073[(\%CaO) + 0.148(\%MgO) + 0.96(\%P_2O_5) + 0.144(\%SiO_2) + 0.22(\%Al_2O_3)] + \frac{11570}{T} - 10.46$	SiO_2-FeO-CaO-MgO_{sat}-P_2O_5-Al_2O_3 T = 1600°C	[12]
10	$log\frac{(\%P)}{[\%P]} = 0.242\,log(B_2 - 0.165(\%MgO)) + \frac{13536.1}{T} - 5.235 - 0.009\,log(\%Fe_t) - 0.010[C]$	CaO-SiO_2-MgO-Fe_tO-P_2O_5 T=1640–1680°C; B_2 = 1.08 – 1.22	[1]

All the models presented in table 1 has similarities that came from the equations 1 and 2. All models have a term to represent oxygen potential in steel, in the form of FeO, MnO or [C]; the models also have a term that represent the importance of temperature (the ratio between a constant and T). Besides that, the importance of basicity is shown in the term of CaO, SiO_2 and MgO. In general, the expressions can be written as the following:

$$\log kp = \sum A(\text{basicity term}) + \sum B(\text{oxygen potential term}) + \frac{C}{T} + D \quad (11)$$

The equation 11, exemplify all terms used in the literature expressions, where A, B and C are coefficients and D is a constant.

The authors have already studied the viability of the expressions presented in table 1 regarding industrial data of an electric arc furnace - EAF [13], and found that all models presented did not reached a satisfactory determination coefficient. Concluding that each empirical relation is limited to its temperature and chemical composition ranges. Besides that, another error source is the fact that the models were developed using equilibrium data, and it is known that EAF is far from the equilibrium.

The authors have already compared the thermodynamic program FactSage v. 7.2 results with industrial data [13] and with experimental data from other authors [14], concluding that FactSage did not achieved a high correlation. However, FactSage have the same behavior, as the literature, considering the effect of oxides, like FeO, CaO and SiO_2 regarding the phosphorus partition (equation 12) [14].

$$Lp = [\%P]/(\%P) \quad (12)$$

As the EAF is far from equilibrium, two new terms were added in equation 11, an equilibrium term, and a kinect term. These terms were obtained by FactSage calculations (Lp and viscosity of the slag). This paper aims to create a new expression using, industrial data (slag and steel), and the results obtained by the thermodynamic program FactSage v. 7.2, to achieve a better correlation. Besides that, the expression will be tested using another group of industrial data.

2 DEVELOPMENT

For the development of this work, data of 3646 heats produced in an EAF (industrial data A), collected over one year were used. This data consists of slag chemical composition, steel chemical composition and temperature, besides that the heats were produced with different scrap loads. This data was already used in another study of the author [13].

In figure 1, is the methodology used for this study, figure 1a) regarding the statistical model, and figure 1b) about the FactSage simulation.

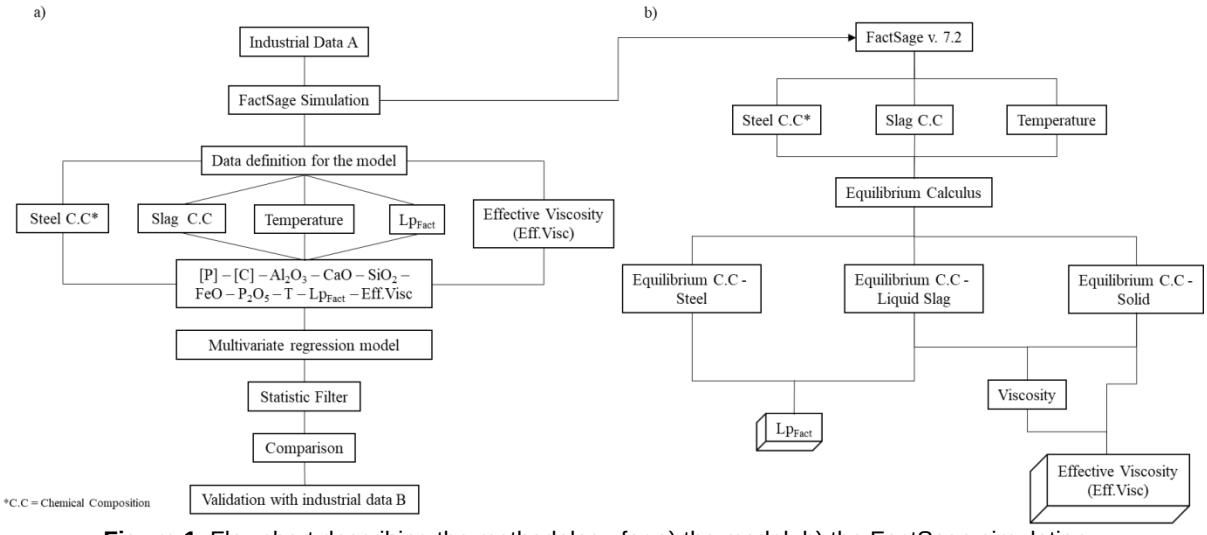

Figure 1. Flowchart describing the methodology for a) the model, b) the FactSage simulation.

The results obtained through the thermodynamic simulation program FactSage v.7.2 (ThermFact Inc., Montréal Canada and GTT-Technologies, Herzogenrath, Germany), described in figure 1 b), were firstly obtained through the Equilib module, selecting the FactPS, FToxid and FTmisc databases. The solution phases selected were FToxid-SLAGA, FToxid-MeO_A, FToxid-a-C2SA, FToxid-b-C2SA, FToxid-C2SP, FToxid-C3Pr, FToxid-C3Pa, FToxid-C3Pb, FToxid-M2Pa and FTmisc-FeLQ.

To calculate the Lp_{Fact}, the equation 12 was used, considering the phosphorus content of the steel and slag calculated by FactSage. To calculate the effective viscosity (Eff.Visc) of the slag, the viscosity modulus was used to obtain the viscosity of the liquid part of the slag, and with the solid particles the Eff.Visc of the slag was calculated using the Roscoe Einstein formula (equation 13).

$$\eta_{eff} = \eta(1 - \theta\rho)^{-2,5} \quad (13)$$

Where

η_{eff} = effective viscosity (poise);

η = liquid viscosity (poise);

θ = constant of packing ratios of the solids in suspension (equal to 1);

ρ = solid fraction.

With these results, the terms consisted by the major slag oxides (Al_2O_3, CaO, SiO_2, FeO, MgO, P_2O_5), steel [P] and [C] from industrial data, and the results from FactSage simulation (Lp_{fact} and Eff.Visc), were tested individually for its significance regarding kp.

With each individual significance, a multivariate regression model was calculated using only the terms that were significant individually.

As stated before, the models presented in table 1 did not achieved a good correlation with the industrial data. For this reason, another term that consider the industrial data as if it is in equilibrium (represented by $Log(Lp_{Fact})$) and a kinetic term (Eff.Visc) were added to the formula, resulting in equation 14.

$$\text{Log}(kp) = \sum A(basicity\ term) + \sum B(oxygen\ potential\ term) + \frac{C}{T} + D + \sum E(equilibrium\ term) + \sum F(kinetic\ term) \quad (14)$$

With the regression result, the industrial data A was filtered using the cook's distance [15].

After the data was filtered, another multivariate regression was calculated, another multivariate regression calculated with the filtered data. The two regressions, before and after the filter, were compared, resulting in a final model.

The industrial data B, consisted in 2000 heats produced one year later the industrial data A, was used in the validation process. There is a huge difference in the two data groups, industrial data A had a considerable content of pig iron in the scrap load, and industrial data B had a very low content of pig iron. The validation of the final model was done using the industrial data B in the final model and compare the result with $Log(k_p)$.

2.2 Results and discussion

As described before, the data consisted 3646 heats produced in an EAF. As the heats were produced with different scrap loads, the data has a great variation, this fact is shown in table 2.

Table 2. *Summary of the Industrial data A*

	Mean	S.D*	Minimum	Maximum
Al_2O_3 (%)	6.01	1.29	2.05	20.23
B_2	2.37	0.45	1.25	4.76

CaO (%)	32.11	4.33	16.25	49.41
FeO (%)	28.25	5.65	7.90	51.36
MgO (%)	7.97	1.64	3.46	40.49
P_2O_5 (%)	0.48	0.13	0.09	1.72
P_{metal} (%)	0.011	0.003	0.003	0.037
SiO_2 (%)	13.83	2.06	7.23	22.53
[C] (%)	0.0749	0.039	0.022	0.9370
T (K)	1898.49	24.01	1803.50	2004.00

**Standard deviation*

In table 2 there are values that were not fit for industrial data of an EAF, specifically maximum value of Al_2O_3 and MgO. These values are much higher than expected for an EAF [16], and probably are outliers.

To choose the data that will be used in the model, firstly the terms were tested individually, to see if they have a significant effect in the kp. The individual tests can be seen in table 3.

Table 3. Individual significance test for each term of the model

	Estimate	Std.Error	t-value	P-value
1/T	-7318.06	664.391	-11.015	<0.001
Al_2O_3	0.053501	0.003349	15.98	<0.001
CaO	0.053735	0.000522	103	<0.001
SiO_2	0.036681	0.002078	17.65	<0.001
B_2	0.24518	0.00909	26.97	<0.001
MgO	0.036943	0.002662	13.88	<0.001
Eff.Visc	0.95133	0.04604	20.66	<0.001
$Log(Lp_{Fact})$	0.45275	0.02177	20.8	<0.001
[C]	0.948733	0.112166	8.458	<0.001

Table 3 show that, each term chosen to the model of kp, is significant individually, and can be used in the general model. FeO was not tested for its significance, because it is part of kp (see equation 3).

The multivariate regression was calculated for all the terms shown in table 3, the final result can be seen in table 4.

Table 4. Results for the multivariate regression

	Estimate	Conf.Int (95%)	t-value	P-value
Constant	-9.827	-10.39 - -9,27	-34.424	<0.001
1/T	11963.47	10787.60 – 13139.34	19.941	<0.001
CaO	0.066	0.06 - 0.07	69.069	<0.001
MgO	0.027	0.02 - 0.03	17.685	<0.001
P_2O_5	0.513	0.48 - 0.54	34.792	<0.001
SiO_2	-0.006	-0.008 – - 0.004	-4.184	<0.001
Eff.Visc	0.058	0.00 - 0.11	2.007	0.0448
$Log(Lp_{Fact})$	-0.521	-0.57 - -0.47	-19.066	<0.001
[C]	-0.617	-0.70 - -0.53	-13.937	<0.001

In table 4, the absence of the Al_2O_3 and B_2 can be noted. Al_2O_3 was not significative when the multivariate regression was done, even though, it was significant individually.

B_2 is not shown, because of multicollinearity between CaO, SiO_2 and B_2. This means that there is redundancy between predictor variables. As a result, CaO and SiO_2 were chosen for the model (they were not multicollinear). The data was filtered using

the cook's distance, resulting in a final industrial data A of 3453 heats and it was analyzed by another multivariate regression. It results can be seen in table 5.

Table 5. Results of the multivariate regression after the filter was applied

	Estimate	Conf.Int (95%)	t-value	P-value
Constant	-8.881	-9.50 - -8.26	-28.025	<0.001
1/T	10271.120	8995.92 – 11546.31	15.787	<0.001
CaO	0.064	0.06 - 0.07	63.738	<0.001
MgO	0.022	0.02 - 0.03	13.593	<0.001
P_2O_5	0.536	0.51 - 0.56	39.258	<0.001
SiO_2	-0.009	-0.011 – - 0.006	-6.234	<0.001
Eff.Visc	0.337	0.24 - 0.43	6.796	<0.001
$Log(Lp_{Fact})$	-0.521	-0.57 - -0.47	-19.482	<0.001
[C]	-0.676	-0.77 - -0.58	-13.819	<0.001

There main differences between table 4 and 5 (before and after the filter, respectively) are: the estimate value for the constant, that is higher in table 5 (from -9.8 to -8.8), the estimate value of temperature, *1/T*, that is lower (from 11963 to 10271) and the effective viscosity term, Eff.Visc, that the estimate had a great increase (from 0.058 to 0.337) and also its significance is higher – the term P-value <0.001.

The comparison between the model before and after the filter can be seen in figure 2.

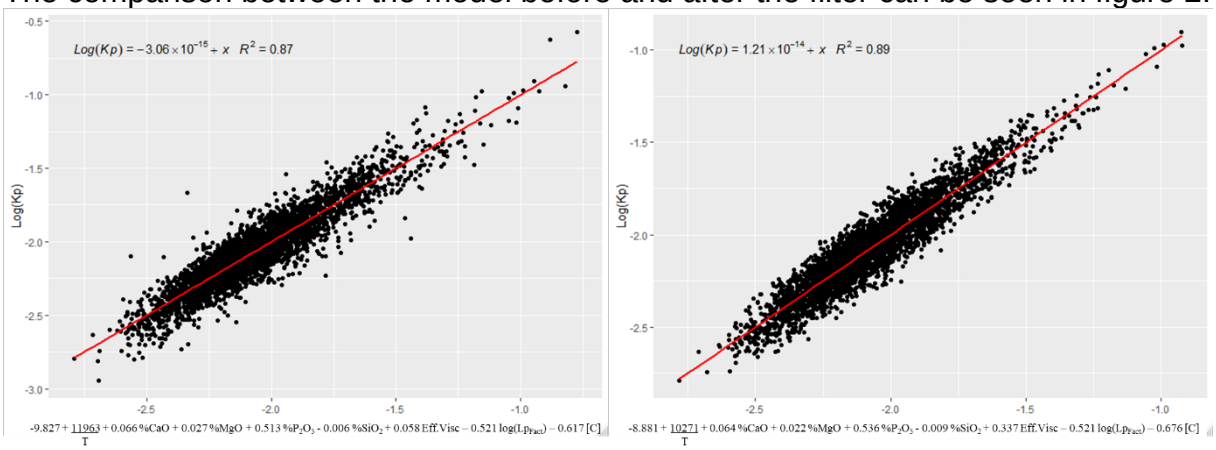

Figure 2. Comparison between the regression model a) before and b) after the filter.

After the filter, the determination coefficient (R^2) had a slightly increase in its value, from 0.87 to 0.89. Showing that the control limits of the model are closer to zero, this can be seen in figure 3.

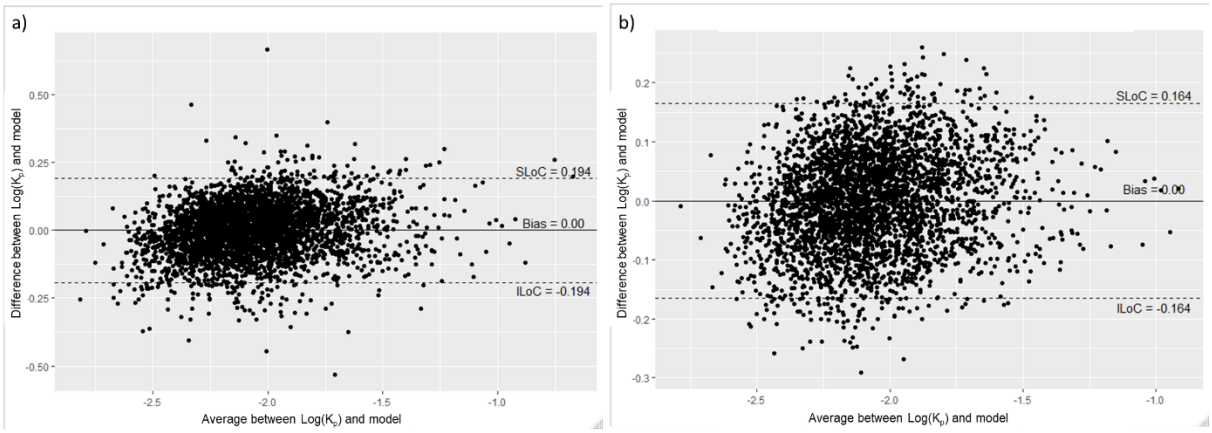

Figure 3. Difference plot showing the superior and inferior limits of control and the mean difference for a) results before the filter and b) results after the filter.

The scatter diagram, in figure 3, show the differences between the values of Log(kp) and the model result, plotted against the averages of the two measurements. It shows that the superior and inferior limit of control (SLoC, ILoC respectively) are closer to zero after the filter, resulting in the greater R^2 showed in figure 2. The final model, calculated with industrial data A after the filter is show in equation 15.

$$\mathbf{Log(kp)} = +\mathbf{0.064} * \mathbf{(CaO)} + \mathbf{0.022} * \mathbf{(MgO)} - \mathbf{0.009} * \mathbf{(SiO_2)} + \mathbf{0.536} * \mathbf{(P_2O_5)} - \mathbf{0.676} * \mathbf{[C]} + \frac{\mathbf{10271.12}}{\mathbf{T}} + \mathbf{0.337} * \mathbf{Eff.Visc} - \mathbf{0.521} * \mathbf{Log(Lp_{Fact})} - \mathbf{8.881} \quad (15)$$

The validation of the final model shown in equation 15, was done plotting the model results using another data group (industrial data B), the variation of industrial data B is shown in table 6.

Table 6. Summary of the industrial data B

	Mean	S.D*	Minimum	Maximum
Al_2O_3 (%)	5.84	1.14	2.45	10.97
B_2	2.15	0.29	1.52	6.25
CaO (%)	29.98	3.42	18.90	45.21
FeO (%)	29.42	5.88	10.72	54.03
MgO (%)	9.57	1.85	4.83	21.38
P_2O_5 (%)	0.40	0.09	0.14	0.86
P_{metal} (%)	0.006	0.002	0.000	0.015
SiO_2 (%)	14.07	1.84	6.22	20.27
T (K)	1941.26	16.96	1723.00	2001.00

The major's differences between industrial data B (table 6) and industrial data A (table 2) are the maximum value of Al_2O_3 and MgO. In table 2 these values were 20.23 and 40.49 respectively, much higher than the values in table 6 (10.97 and 21.38). The validation of the final model is shown in figure 4.

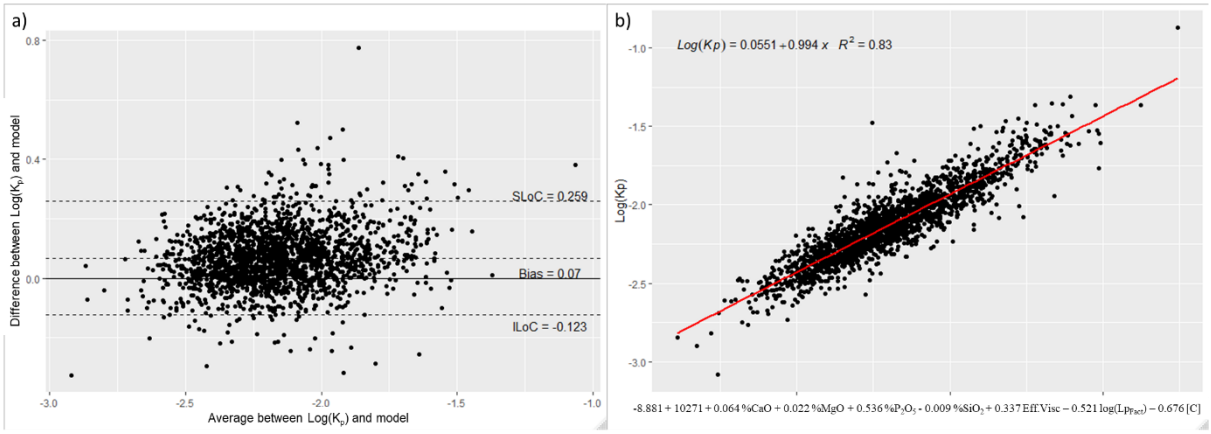

Figure 4. Validation for the model calculated using industrial data B, a) Difference plot showing the superior and inferior limits of control and the mean difference, b) comparison between the regression model with log(kp).

Figure 4 show two different graphs, figure 4 a) show the difference between the final model and Log(kp). In this graph that probably the final model is not fit to be used in this data. The reason for that is: the control limits of the model (SLoC and ILoC) are displaced. The value of this displacement is shown by the Bias, that is the average difference between the final model and Log(kp), it value is 0.07, even though this difference is small, it is not negligible. This difference affected the determination coefficient, figure 4 b), that is 0.83. Comparing this R^2 with the ones found in figure 2 b), a reduction of 0.07 was observed. The reason of this difference probably is related to the scrap load of each data. In industrial data A, the scrap load had a high content of pig iron (>1**5**%), however scrap load B had none or a very low (<5%) pig iron content. As pig iron is the main source of phosphorus in EAF [12], this difference had affected the behavior of dephosphorization.

3 CONCLUSIONs

This study showed that the two terms calculated by FactSage v. 7.2, $Log(Lp_{Fact})$ and Eff.Visc, had a significative effect in the Log(kp) of industrial data. It can also be concluded that the use of these two parameters helped to achieve a higher correlation between the regression model and Log(kp) of industrial data, as they had a very low p-value, showing its significance in the multivariate regression, resulting in the final model.

$$\mathbf{Log(kp)} = +\mathbf{0.064} * \mathbf{(CaO)} + \mathbf{0.022} * \mathbf{(MgO)} - \mathbf{0.009} * \mathbf{(SiO_2)} + \mathbf{0.536} * \mathbf{(P_2O_5)} - \mathbf{0.676} * \mathbf{[C]} + \frac{\mathbf{10271.12}}{\mathbf{T}} + \mathbf{0.337} * \mathbf{Eff.Visc} - \mathbf{0.521} * \mathbf{Log(Lp_{Fact})} - \mathbf{8.881}$$

Al_2O_3 content is omitted from the final model, as it was not considered significative in the multivariate regression, even though it was significative individually.

The final model had a high R^2 (0.89) for the industrial data A after the filter, however it did not have the same success in the validation using industrial data B (R^2=0.83), probably, due to the differences in the scrap load (pig iron content in industrial data A was much higher than industrial data B).

This fact evidences the importance of scrap load in dephosphorization, as different types of iron sources (scrap, pig iron, reduced iron) had different content of phosphorus. Depending on the amount that is added, it will influence the phosphorus content in the steel, and the dephosphorization behavior.

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