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Evaluating the hot metal dephosphorization efficiency of different synthetic slags using phosphorus partition ratio, phosphate capacity and computational thermodynamics

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Abstract

Phosphate capability (C_p) and phosphorus partition ratio (L_p) are usually used to evaluate the thermodynamic efficiency of dephosphorization slags with different compositions in hot metal pretreatment. However, these parameters are only useful in equilibrium conditions, and they are not accurate when they are used to evaluate slag efficiency in industrial processes. The current study evaluated the hot metal dephosphorization efficiency of different synthetic slags in experimental tests at 1400°C. This evaluation was made by using C_p and L_p , and by the computational thermodynamic software FactSage 6.4. This software allows to calculate the amount of liquid and solid present in the slag, which directly affects kinetic reactions. Therefore, even the computational thermodynamic software calculations are from considering the equilibrium, which in these cases can be used to explain the kinetic behavior of the processes.

The obtained results allow concluding that C_p and L_p are valid, but only when they are used for equilibrium calculations; and that they should not be used to measure slag efficiency in industrial processes. A parameter named Dephosphorization Factor (F_{DeP}) was developed based on data determined by computational thermodynamics. It was developed to be related to the dephosphorization efficiency. The results obtained in the current study allow to conclude that the initial synthetic slags, which presented higher CaO activity value, higher liquid quantity and FeO activity, will be the most efficient ones. And allow to conclude too, that the higher the factor, the greater the slag dephosphorization efficiency.

Keywords: hot metal pre-treatment; dephosphorization; computational thermodynamics.

1. Introduction

Turkdogan (1996, 2000) and Heally (1970) showed that phosphorus oxidation may occur by the reaction between

the metal-dissolved phosphorus and the oxygen provided by the iron oxide found in the slag. The formed phosphorus pentoxide is fixed in the slag by the CaO found in it. This process forms tricalcium phosphate (3CaO.P₂O₅).

$$2P + 5(FeO) + 3(CaO) = 5Fe + (3CaO.P_2O_5) \Delta G^0 = -204450 + 83.55T (J/mol)$$
 (1)

$$\ln (\%P)_{eq} = \frac{1}{2} \ln (a_{3CaO.P205}) + \frac{\Delta G^0}{2RT} - \frac{5\mu_{02}}{4RT} - \frac{3}{2} \ln (a_{CaO}) - \ln (f_p)$$
(2)

In regards to the free energy standard, it is observed that the reaction is favored by low temperatures. They positively influence hot metal dephosphorization, since it presents temperatures lower than those of steel. The thermodynamics evaluation of the slag's dephosphorization efficiency is often performed according to the following parameters: phosphate capability (C_p) and partition ratio (L_p) . However, the computational thermody-

namic software used in the current study evaluates the proposed dephosphorizing slags, since this software identifies both the thermodynamics and the kinetic variables affecting the dephosphorization reaction. According to Campos (1984), hot metal dephosphorization does not reach equilibrium due to kinetic factors associated with the reaction, i.e., the phosphorus mass dissolved in the metal is transported to the metal-slag interface, reacts with the

oxygen provided by the oxygen sources in the dephosphorizing slag and enables phosphorus pentoxide (P_2O_5) formation. Young (1991) obtained several data in literature describing the equilibrium reached between metal and slag. Equation 3 shows his results. According to Young (1991), the term involving P_2O_5 in the equation refers to some slag data about this oxide's high concentration.

$$\log C_p = -18.184 + 35.84\Lambda - 22.35\Lambda^2 + \underline{22930\Lambda}_T - 0.06257.\% FeO - 0.04256.\% MnO + 0.359.\% P_2O_5^{0.3}$$
 (3)

The phosphorus partition ratio (L_p) is another evaluation method. This method expresses the equilibrium relation between the phosphorus concentration in the slag (% P) and the phosphorus concentration in the metal [%P]. The most used phosphorus partition ratio in

steelmaking is that suggested by Healy (1970), due to its simplicity and because its parameters are easily obtained. The author used ionic fractions to determine the phosphorus partition in the metal-slag interface. This correlation is shown in Equation 4. Suito (2006)

studied phosphorus transfer from a CaO-Fe_tO-P₂O₅-SiO₂ slag system to a CaO particle at 1400°C. Based on his experimental test, he developed an expression to estimate phosphorus partition in the metal-slag interface, shown in Equation 5.

$$\log \frac{(\%P)}{[\%P]} = \frac{22350}{T} + 0.08.(\%CaO) + 2.5.\log(\%Fe_{total}) - 16$$
(4)

$$\log \left\{ \frac{(\%P)}{[\%P]} \right\} = 0.072 \{ (\%CaO) + 0.3.(\%MgO) + 2.5.\log(\%Fe_{total}) + \frac{11570}{T} - 10.52$$
 (5)

The general equation to study the dephosphorization reaction rate is shown

in Equation 6 (WEI, 1993). Eq. 6 is known as the general equation for dephosphoriza-

tion reaction, whenever mass transportation within the metal controls the reaction.

$$-\frac{d[\%P]}{dt} = k' \frac{A\rho_m}{W_m} \cdot ([\%Pt] - [\%Peq])$$

(6)

Where: -d([%P])/d(t): dephosphorization rate (%P/min); k': global phosphorus mass transfer coefficient (m/s);

m: metal phase, respectively; ρ : metal density (kg/m³); A: interfacial area (m²); W_m : metal mass (kg), %Pt: phosphorus

content at time t and %Peq: phosphorus content at equilibrium.

Thus, the current study aims to evaluate hot metal dephosphorization efficiency using slags based on the CaO-FeO system. Therefore, the parameter used to evaluate synthetic slag dephosphorization efficiency must take into account the thermodynamic parameters shown in Equation 2, as well as the kinetic parameters shown in Equation 6. The computational

thermodynamics allows determining these parameters that are the amount of liquid and solid and the phases present in the slag. These phases directly interfere in the slag-metal interfacial area (A), from Equation (6). The computational thermodynamics allows determining, in addition to the equilibrium activities of the slag constituents, (GRILLO, 2013). There-

fore, even the computational thermodynamics software calculations take equilibrium into consideration, and in these cases can be used to explain the kinetic behavior of the processes. Oertel and Costa e Silva (1999) used computational thermodynamics to study equilibrium in steelmaking, and Heck (2007) to study stainless steel production.

2. Material and methods

Experimental procedures

Initially, solid pig iron was put into MgO-C crucibles and inserted into a MAI-TEC electric furnace, model 1700-FEE at 1400°C. Moreover, argon gas was blown onto the pig iron at the rate 10Nl/min to purge the environment and to prevent metal oxidation. The pig iron was completely melted after approximately 40 minutes. Then, an initial sample of this metal was removed by vacuum sampler, and the synthetic slag was added to the hot metal

through a stainless steel tube. After the synthetic slag addition, an alumina impeller was inserted into the bath at 500 rpm. Hot metal samples were removed after 0, 5, 10, 15, 20, and 30 minutes. The sulfur and carbon content in the metal was analyzed by the combustion infrared method, where the equipment was a simultaneous carbon and sulfur automatic analyzer, Quimitron QSC 7000 Plus. The phosphorus and silicon content in metal was analyzed by

iron-based ICP (plasma) spectrometry using a Spectroflame ICP spectrometer. The chemical analysis of the final slag generated during the dephosphorization tests was analyzed under an X-ray fluorescence (XRF) spectrometer. Table 1 presents the initial chemical composition and the hot metal mass used in the tests. Table 2 presents the initial chemical composition and the synthetic slag mass suggested and applied to the experimental tests.

Evaluating synthetic slags using partition ratio and phosphate capacity

The hot metal dephosphorization thermodynamic analysis was initially done

using thermodynamic models, phosphate capacity and partition ratio. The chemical

composition of the MD1 to MD4 synthetic slags was used to calculate these parameters.

Evaluating synthetic slags using Computational Thermodynamics Software

Computational thermodynamic simulations were performed in FactSage 6.4 with FTOXID databases (Bale, 2008) according to the hot metal and

to the dephosphorizing synthetic slag's chemical compositions presented in Tables 1 and 2. This procedure was used to determine the phosphorus concentra-

tion equilibrium in the metal, as well as the phases presented and the amount of solid and liquid in the synthetic slags at the beginning of the process.

Synthetic	Rotation		Mana (1.0)				
′ slags	(rpm)	С	C Si P		S	Mass (kg)	
MD1	500	3.94	0.34	0.096	0.0104	1.0	
MD2	500	3.99	0.33	0.089	0.0088	1.0	
MD3	500	3.94	0.27	0.102	0.0116	1.0	
MD4	500	3.90	0.31	0.103	0.0079	1.0	

Table 1
Hot metal initial mass and chemical composition.

Synthetic		Cher	nical Cor	mpositio	n (%)		C-0/5-0	CaO/SiO ₂	Mass (g)
′slags	FeO	CaO	SiO ₂	Al ₂ O ₃	MgO	MnO	CaO/FeO		
MD1	71.6	25.6	1.65	0.68	0.20	0.5	0.36	15.5	65.0
MD2	65.8	31.5	1.60	0.64	0.20	0.4	0.48	19.7	65.0
MD3	60.0	37.4	1.51	0.63	0.20	0.4	0.62	24.7	65.0
MD4	56.1	41.3	1.49	0.61	0.20	0.4	0.73	27.7	65.0

Table 2 Initial chemical composition of dephosphorizing synthetic slags.

As for the current study, the liquid and solid phases formed within the synthetic slags were determined through Fact-Sage applications. Fact53 and FTOXID

databases were used in the analysis. Fact 53 determined the formed solid compounds, whereas FTOXID evaluated the compounds formed in the liquid

phase, the CaO and FeO activity of the synthetic slags and in the final slag. These parameters were then used to evaluate the slag efficiency.

3. Results and discussion

Experimental results

Table 3 shows the hot metal chemical composition variation over time and the dephosphorization efficiency (η) obtained in the experimental tests by using the MD1,

MD2, MD3 and MD4 synthetic slags produced in laboratory using pellet feed and lime. The efficiency was calculated by Equation 7. Figure 1 shows the [%P.]/[%P₀]

ratio variation in hot metal according to time. $[\%P_t]$ is the phosphorus concentration in the hot metal at time (t), and $[\%P_0]$ is the initial phosphorus concentration.

$$\eta(\%) = \frac{([\%P_{initial}] - [\%P_{final}])}{[\%P_{initial}]} \times 100$$
 (7)

Time	Time (min)		10	20	30	η(%)
	%C	3.9396	3.4925	3.4474	3.3801	14.20
MD3	%P	0.1026	0.0219	0.0166	0.0107	89.57
	%Si	0.2639	0.1443	0.1342	0.1310	50.36
	%C	3.9032	3.5197	3.4986	3.4768	10.92
MD4	%P	0.1037	0.0237	0.0218	0.0213	79.46
	%Si	0.3061	0.1433	0.1409	0.1405	54.10
	%C	3.9896	3.4937	3.4314	3.3480	16.08
MD2	%P	0.0897	0.027	0.023	0.0205	77.15
	%Si	0.3293	0.1482	0.1439	0.1447	56.06
	%C	3.9421	3.5067	3.4494	3.4067	13.58
MD1	%P	0.0961	0.0282	0.0259	0.0251	73.88
	%Si	0.3431	0.1759	0.1729	0.1671	51.30

Table 3
Hot metal chemical composition variation over time in dephosphorization. Experimental tests.

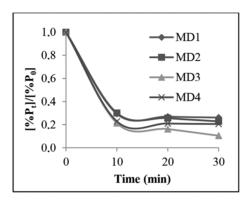


Figure 1 [%P₁]/[%P₀] ratio variation in hot metal according to time.

Based on Table 3, it is possible to primarily conclude that the used synthetic slags are able to remove silicon and phosphorus and to reduce the concentration of these elements in the hot metal. Furthermore, the MD3 synthetic slag presented the highest dephosphorization capacity with 89.6% efficiency. MD2 and MD4 synthetic slags presented a performance similar to that of MD3, with 77.1% and 79.5% efficiency, respectively. The MD1 synthetic slag was less effective in phospho-

rus concentration reduction in hot metal than the others, presenting a 73.9% efficiency. Figure 1 shows the kinetic curves of the dephosphorization process using the 4 different slags, which can be represented by Equation (6). It can be noted that MD3 had the lowest final value of P in the same time interval, and therefore a greater dephosphorization rate (%P/min). Figure 1 shows too that, when the synthetic slags were used, there was considerable phosphorus concentration reduction in hot metal in

the first ten minutes of the experimental tests. The synthetic slags used in the experimental dephosphorization tests were evaluated using thermodynamics software in order to obtain further information and conclusions about their behavior. By use of the software Fact Sage, the amount of the liquid and solid phases formed, the CaO and FeO activity was determined. These properties were then used to measure slag efficiency. In the next section this item will be discussed.

Thermodynamic evaluation of synthetic slags using partition ratio and phosphate capacity

Figure 2 shows the correlation between the efficiency obtained in the experimental tests and the dephosphorization parameters such as phosphate capacity (CP) and phosphorus partition ratio (LP). Figures 2a and 2b show that, increased phosphate capacity (CP) and phosphorus partition ratio (LP) results in a dephosphorization efficiency increase. However, the correlation between the thermodynamic parameters and the efficiency obtained in the experimental tests was relatively low, approximately 0.4.

This is due the fact that these parameters are not sufficient to explain the process, since formation of the solid phases changes the slag behavior, by influencing the amount of liquid generated and, consequently, the kinetics. These parameters should only be used for thermodynamic calculations, and do not allow an accurate evaluation of which slag will be the most efficient, and it is necessary to develop a new model that allows a more accurate analysis.

Therefore, a new parameter was developed to obtain a better correlation between this Factor and the slag dephosphorization efficiency. That is, a factor was developed so that the higher the factor, the greater the slag dephosphorization efficiency. This factor was named Dephosphorization Factor (FDeP).

The development of this factor was

based on the dephosphorization thermodynamic equation (Equation 2), in the dephosphorization kinetic equation (Equation 6), and in the data obtained by Fact Sage.

From the analysis of these equations, it is possible to conclude that the most efficient slag will be the one with: CaO activity equal to 1 and simultaneously the highest FeO activity and amount of liquid phase possible, as long this does not reduce the activity of CaO for values less than 1. In other words, the slag should only have a CaO content needed to saturate the slag. The Dephosphorization Factor (FDeP) and that whichwas initially defined according to Equation 9, where Nliquid,

NSolid and kDeP are the liquid mass fraction, the solid mass fraction, and the

dephosphorization constant respectively.

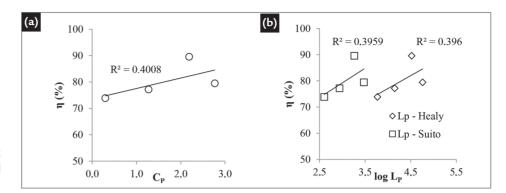


Figure 2 Relation among η (%) and (A) CP and (B) LP of the synthetic slags at 1400°C.

$$F_{DeP} = a_{CaO} + a_{FeO} + (N_{liquid} - N_{solid}).k_{DeP}$$
 (8)

Where: $\mathbf{a}_{\mathsf{CaO}}$: CaO activity; $\mathbf{a}_{\mathsf{FeO}}$: FeO activity; $\mathbf{N}_{\mathsf{Liq}}$: Liquid mass fraction and $\mathbf{N}_{\mathsf{Sol}}$: Solid mass fraction; $\mathbf{k}_{\mathsf{DeP}}$: the dephosphorization constant.

The dephosphorization constant

 $(k_{DeP)}$ was created to increase the correlation between the dephosphorization efficiency and the dephosphorization factor $(R^2 \text{ value})$. It is an adjustment factor for these correlations. The value for (k_{DeP})

was equal to 1.66 in the current study

The next section will demonstrate the computational thermodynamics used in the synthetic slag evaluation suggested in the current study.

Thermodynamic evaluation of dephosphorizing synthetic slags using computational thermodynamics software Table 4 shows the results of synthetic slags, %Liquid, %Solid, a_{CaO} and a_{FeO} determined by FactSage.

Table 4
Phases formed in the synthetic slags generated at the beginning of the process and CaO and FeO activity using FTOXID database.

Synthetic slags	%Liquid	%Solid	a _{CaO}	$a_{_{FeO}}$	η (%)
MD3	99.33	0.67	1.0	0.38	89.57
MD4	93.22	6.78	1.0	0.38	79.46
MD2	100.0	-	0.63	0.51	77.15
MD1	100.0	-	0.40	0.62	73.88

According to Table 4, except for the MD1 and MD2 synthetic slags, which are completely liquid at 1400°C, the other slags show a formed solid phase, which consisted of CaO only.

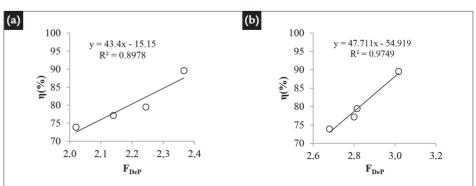
Table 4 shows that the slag generated at the beginning of the process using MD3 and MD4 synthetic slags was saturated with CaO. However, the solid CaO concentration in the MD3 slag is less than that in the MD4, 0.67% and 6.78%, respectively. By analyzing Table 4, it is possible to see that the most effective synthetic slags were those that had the highest CaO activity value and CaO solid

enough to keep the initial synthetic slag saturated at the beginning of the dephosphorization process. In addition, among the synthetic slags presenting higher CaO activity values, MD3 was the one showing the highest amount of liquid, thus it was the most efficient. Analyzing these results, from Equation 6, this synthetic slag had better dephosphorization efficiency and consequently a higher dephosphorization rate (%P/min). It was the one that had the greatest amount of liquid and consequently a greater value of A, and CaO activity equal to 1.

Therefore, it can be stated that -

based on the results found in the current study – the synthetic slags showing CaO activity equal to 1 had better performance, since their liquid amount was higher than 93%. Besides influencing the equilibrium phosphorus concentration, the CaO activity influenced P_2O_5 activity and, consequently, the P_2O_5 concentration in the slag.

For a better understanding of the results, the desulfurization efficiency was correlated with the Desphosphoration Factor. The dephosphorization factor was calculated according to the data shown in Table 4. These results are shown in Figure 3.



 $\label{eq:Formula} \begin{array}{c} \text{Figure 3} \\ \text{Correlation between (a) } F_{\text{DeP}} \text{ and } \eta(\%) \\ \text{without } k_{\text{DeP}} \text{ and (b) Correlation between} \\ F_{\text{DeP}} \text{ and } \eta(\%) \text{ using kDeP correction factor.} \end{array}$

The analysis showed that the best synthetic slag had the greatest CaO activity and liquid amount (since it does

not reduce CaO activity) and the smallest number of solid phases. Initially the dephosphorization factor (FDeB) was then defined according to Equation 10 without k_{DeP} .

$$F_{DeP} = a_{CaO} + a_{FeO} + (N_{Lia} - N_{Sol})$$

Figure 3(a) shows the correlation between F_{DeP} and dephosphorization efficiency, which was determined, using Equation 8 without k_{Dep} . Figure 3(b) shows the correlation between

F_{DeP} and dephosphorization efficiency using the k_{DeP} , shown in Equation 9. This dephosphorization constant (k_{Dep})

It is possible to note that the use of the k_{Dep} increased the correlation between F_{DeP} and $\eta(\%)$, from 0.90 to 0.97. In this case, the overall equation for F_{DeP} calculation is:

$$F_{DeP} = a_{CaO} + a_{FeO} + (N_{Lig} - N_{Sol}).kDeP.$$

(10)

(9)

The k_{DoP} correction factor was used here instead of the 1.66 value, because k may or may not take other values into account under different processing conditions.

Then, the F_{DeP} Equation for the present study is:

$$F_{DeP} = a_{CaO} + a_{FeO} + (N_{Lig} - N_{Sol}).).1.66$$

(11)

Thus, a model was developed to predict dephosphorization efficiency for slags with the same composition range and efficiency used in this study, based on the trend line equation:

$$\eta(\%) = 47.711(F_{DeP}) - 54.919$$
 (12)

Table 5 shows the final chemical composition of the final slag obtained in the dephosphorization tests. Table 6 shows the formed phases and the CaO and FeO activities in the final slag; both data were obtained by means of FactSage.

Final	Chemical Composition (%)									
slags	FeO	CaO	SiO ₂	Al ₂ O ₃	MgO	MnO	P ₂ O ₅	Cao/FeO	CaO/SiO ₂	η (%)
MD3	27.7	52.3	11.5	2.11	1.86	0.30	4.23	1.89	4.55	89.6
MD4	24.4	57.1	12.2	0.90	1.17	0.30	3.93	2.34	4.68	79.5
MD2	35.2	41.1	13.8	2.84	2.14	0.55	4.37	1.17	2.98	77.1
MD1	42.4	32.3	14.7	1.4	3.91	0.60	4.69	0.76	2.20	73.9

Table 5 Final concentration of the final slag generated during the dephosphorization tests.

Final slags	%Liq.	Solid phase (%CaO)	a _{CaO}	a _{FeO}	η (%)
MD3	93.8	6.2	1.00	0.29	89.6
MD4	86.7	13.3	1.00	0.28	79.5
MD2	100	-	0.27	0.59	77.1
MD1	100	-	0.11	0.75	73.9

Table 6 Phases formed within the final slag generated at the end of the dephosphorization process and the CaO and FeO activities at 1400°C, using the FT OXID database.

Table 6 shows that the final slags generated by the MD3 and MD4 synthetic slags were CaO-saturated at the end of the dephosphorization process. However, the concentration of solid CaO in the MD3 slag was lower than that in MD4, 6.2% and 13.3%, respectively. The MD1 and MD2 slags were not CaO-saturated at the end of the dephosphorization process. According to Table 6, it was noticed that the most effective slags presented the highest CaO activity value at the end of the dephosphorization process. Among the slags that presented the higher CaO activity values (MD3 and MD4), MD3 was the one with the highest liquid amount; thus, it was the most efficient.

4. Conclusions

According to the obtained results, it is possible to conclude that: CP and LP have low correlation (R2) with dephosphorization efficiency and they are not accurate for predicting it; and the initial

synthetic slag presenting the highest CaO activity value, the highest liquid amount and FeO activity will be the most efficient one. Therefore, the synthetic slag presenting the highest F_{DeP} value will be the most efficient one; It was possible to develop a model to predict dephosphorization efficiency according to the CaO and FeO activities, and to the amount of solid and liquid: η (%)=47.711(F_{DeP}) – 54.919.

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