Original Article

Sulfur incorporation assessment in hot metal from the blast furnaces at Usiminas

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Abstract

Sulfur is an element with strict specifications for certain steel grades, making its control in blast furnace produced hot metal essential. In this regard, the factors influencing sulfur incorporation in the small (BF-S) and medium (BF-M) blast furnaces at Usiminas were analyzed. The methodology included statistical analysis of the sulfur percentage in the furnaces, considering the correlated variables, with statistical comparison between the furnaces (t-test) and multiple regression models for sulfur content prediction in each reactor. Additionally, scenarios for sulfur reduction were simulated by partially replacing coke with charcoal and natural gas. The results indicated that the top pressure in BF-M was the determining factor for its lower sulfur content compared to BF-S, while quaternary basicity was the main explanatory variable for sulfur incorporation in both furnaces. In addition, assuming a constant desulphurisation efficiency, the sulphur content of the hot metal can be reduced by up to 8.8% by partially replacing coke or mineral coal with charcoal or natural gas.

Keywords: Blast furnace; Sulfur; Hot metal; Statistical analysis.

1 Introduction

Sulfur is an undesirable element in steel, as it reduces ductility and resistance to impact and corrosion, and can lead to the formation of non-metallic inclusions such as manganese sulfides (MnS), which act as initiation sites for hydrogen-induced cracking [1,2]. In a reducing environment, such as that of the blast furnace, sulfur transfer from the hot metal to the slag occurs in the form of sulfide ions (S^{2-}) [2]. However, the kinetic aspect of sulfur transfer between hot metal and slag is one of the limiting factors for desulfurization in the blast furnace [3]. The removal of sulfur from hot metal depends on the contact time between the metal and the slag, and equilibrium is not reached due to the high viscosity of the slags and the continuous tapping of liquids from the hearth, which hinders sulfide diffusion between phases [4,5].

From a thermodynamic standpoint, sulfur partitioning between hot metal and slag is directly proportional to oxygen partitioning in these phases. Therefore, reducing oxygen content in the hot metal and increasing it in the slag is necessary to lower the sulfur content in the metal [2,6]. Consequently, variables that affect oxygen activity in both hot metal and slag are directly related to sulfur behavior in the blast furnace. In this context, strategies to produce hot metal with low sulfur content include reducing sulfur input into the furnace, using metallic burden materials with high reducibility, employing more basic slags, and maintaining low oxygen activity in the hot metal [2,3].

Regarding furnace input, coke is the main sulfur carrier [7], accounting for approximately 90% of the sulfur charged into the furnace [2,8]. A previous study [9] showed that sulfur volatilization from coke increases with temperature, reaching losses of 15–31% at 1500 °C. Of the total sulfur, 54–69% originates from organic compounds and 31–46% from mineral sulfides, with complete volatilization occurring between 2100 and 2200 °C. However, another study [10] found that the sulfur content in coke upon reaching the tuyeres is about 60% of its initial value, with the remaining 40% being gasified in the upper furnace region and fully absorbed by the slag [11] and metallic burden, especially those containing basic oxides [10], and the content in metal is low. Sulfur gas absorption by sinter occurs above 1000 °C, with significant uptake by primary slag, followed by a decrease in concentration due to assimilation by the gangue in the sinter and renewed volatilization near the tuyeres [8]. In addition to coke, auxiliary fuels such as oil and pulverized coal can also increase sulfur content in the metal, as they contain significant amounts of the element [12,13].

Basic slags rich in CaO and MgO are effective in desulfurizing hot metal, as they solubilize S^{2-} ions by donating O^{2-} to capture sulfur [2,6]. Therefore, desulfurization efficiency

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increases with slag basicity, especially ternary and quaternary basicity [5,8,13,14]. Moreover, replacing SiO_2 with Al_2O_3 enhances the slag's sulfide capacity, but raises the liquidus temperature, which may hinder desulfurization [3,5]. In this context, MgO contributes to sulfur reduction in hot metal but is less effective than CaO [3, 5, 7], and its effect in the blast furnace should also be evaluated in terms of sinter quality, especially if it is added only during sintering [15]. FeO may promote desulfurization in acidic slags but hinders it in basic slags [2]. Na_2O is a good desulfurizer, but its use is limited due to crust formation on the furnace wall, which can affect the refractory lining [2,6].

Beyond chemical composition and basicity, other slag properties such as process temperature, liquidus temperature, and slag volume (slag rate) also influence hot metal desulfurization. The liquidus temperature is the point at which the first crystal forms from a homogeneous liquid, and it increases slag viscosity, reducing desulfurization efficiency. Minor oxides (FeO, P_2O_5, etc.) can lower the slag's liquidus temperature by 30 to 50 °C [7]. Additionally, the sulfur absorption rate by slag increases with temperature, favoring desulfurization by reducing viscosity and improving sulfide transfer kinetics [3,11].

Desulfurization efficiency improves normally with lower slag rates, as higher slag volumes require more energy and coke although decreases the sulfur activity in the slag with same basicity, resulting in increased sulfur input into the furnace. On the other hand, sulfur solubility in hot metal decreases with rising temperature [8].

Oxygen activity in the metal is influenced by the reducibility of the metallic burden, bed permeability and reduction kinetic in coke bed zone. A decrease in these factors increases oxygen activity in the hot metal [3]. Permeability disturbances can lead to scaffolding and slips, transferring material with high oxygen activity to regions where it normally would not occur, thereby impairing desulfurization efficiency [3]. Furthermore, increased sulfur in hot metal reduces carbon saturation, hinders carburization, and interferes with contact between coke and hot metal [13,16].

Therefore, controlling the variables that affect sulfur transfer in the blast furnace is essential to reduce sulfur levels in hot metal, a key objective in the operation of Usiminas' blast furnaces. In this context, this study aimed to identify, through process data analysis and thermodynamic evaluations, the factors influencing sulfur incorporation in the hot metal produced by Usiminas' small-scale (BF-S) and medium-scale (BF-M) blast furnaces, which exhibited different sulfur contents.

2.1 Materials and methods

2.1.1 Data analysis

The individual evolution of the sulfur percentage in hot metal (S_hot metal) from Usiminas' small-scale (BF-S) and medium-scale (BF-M) blast furnaces was evaluated and statistically compared. The independent samples t-test was used to assess differences between the furnaces. In statistics, the t-test is a hypothesis test that allows for the comparison of means. The resulting p-value indicates whether the variables are statistically similar or different. The t-test is applicable only to normally distributed data. However, according to the Central Limit Theorem (CLT), when the data are continuous and the sample size exceeds 30, normality is not a strict requirement, as the data tend to approximate a normal distribution under these conditions [17].

Subsequently, other operational variables from both reactors were analyzed to identify correlations and understand the influence of key parameters on sulfur content in hot metal. Based on this, statistical models were proposed to predict sulfur levels using the most relevant variables. For BF-S, data were collected over a 28-month period, and for BF-M, over 23 months. Daily averages were used to correlate furnace operational data with hot metal quality. For BF-S, only days with production above 1600 tons were selected, and for BF-M, days with production above 5500 tons, in order to exclude periods with scheduled shutdowns or significant operational instability. These thresholds were chosen due to the different furnace volumes (885 m^3 for BF-S and 3163 m^3 for BF-M), and because BF-S exhibits greater thermal losses in the hearth, as shown in a previous study [18]. Additionally, only days with complete records of all monitored variables were considered.

More than 100 blast furnace variables were evaluated using the statistical software Minitab. Correlation matrices (heatmaps) were generated using Python, with the Matplotlib and Seaborn libraries, which offer superior graphical visualization. Statistical modeling was performed using multiple regression analysis, following a four-step methodology: (1) assess variable correlations (correlation matrix), (2) perform regression analysis including collinearity (VIF) and significance (p-value) checks, (3) verify model residual assumptions (normality, independence, and constant variance), and (4) develop and simplify the predictive model.

2.1.2 Sulfur indicators in hot metal

Sulfur incorporation into hot metal was assessed using the following indicators: desulfurization index (ratio between sulfur input and sulfur removed via slag), Equation 1; natural logarithm of the actual sulfur partition between slag and hot metal (ln(S)/[S]); slag sulfide capacity (ability to solubilize sulfur), expressed as ln(C_S), Equation 2 [2]; and slag viscosity (μ), Equation 3 [19].

$$Desulfurization\ Index(\%) = \frac{S_{input} - (\%S_{HM} \times 10)}{S_{input}} \times 100 \quad (1)$$

Where S_{input} and $\%S_{HM}$ represent the sulfur input into the furnace (kg/t) and the sulfur content in hot metal, respectively.

$$\ln C_S = 2.303\left(\begin{array}{l} -5.57 + 1.39\dfrac{\%CaO + 0.7\%MgO}{0.94\%SiO_2 + 0.18\%Al_2O_3} + \\ 2.607\times10^{-3}\left(T_{slag} - 1500\right) \end{array}\right) \quad (2)$$

Where $\%CaO$, $\%MgO$, $\%SiO_2$, $\%Al_2O_3$ are the mass concentrations of the respective oxides in the slag, and T_{slag} is the slag temperature (°C).

$$ì = Exp\left(\begin{array}{l} -10.3469 + \dfrac{25144}{T_{esc}} - 9.6334x\dfrac{CaO}{100} - \\ 11.8176x\dfrac{MgO}{100} - 0.80216x\dfrac{Al_2O_3}{100} \end{array}\right)x10 \quad (3)$$

Equation (3) is valid within the range 35%< $\%SiO_2$ <45%, which corresponds to the slag compositions in Usiminas' blast furnaces. Under normal operating conditions, slag temperature inside the blast furnace is always higher than that of the hot metal [14], with an average difference of approximately 50 °C [7]. Since slag temperature is not directly measured, the hot metal temperature plus 50 °C was used.

2.1.3 Sulfur balance in the blast furnaces

A mass balance tool was developed to simulate scenarios for sulfur content in hot metal, based on the input materials and their compositions, slag rate, and the actual desulfurization efficiency of the reactors, calculated using the sulfur partition ratio (%S)/[S] (or L_S), obtained statistically. This ratio was determined for 687 operational days of BF-S and 734 days of BF-M. From these data, the mean and standard deviation of L_S were calculated, reflecting desulfurization efficiency. Using the input sulfur (Sinput), L_S values, and slag rate (SR), the sulfur content in hot metal can be estimated as shown in Equation 4:

$$\%S_{HM} = \frac{S_{input}}{\left(L_S \times SR + 1000\right)} \quad (4)$$

With the developed tool, scenarios were simulated to evaluate the impact of charcoal and natural gas, both low-sulfur fuels, on sulfur levels in hot metal. These materials align with ESG policies. In the simulations, a standard metallic burden and fixed slag rate were considered. The average sulfur value was represented by the mean L_S, while maximum and minimum values corresponded to positive and negative deviations from this mean, respectively.

2.2 Results and discussion

2.2.1 Comparison of sulfur in hot metal from the blast furnaces

A significant difference was observed in sulfur incorporation into the hot metal from BF-S and BF-M (p-value = 1.95×10^{-14}). In this context, BF-S exhibited a higher sulfur content than BF-M during the evaluated periods (Figure 1), with both the mean and standard deviation being greater for BF-S.

2.2.2 Small-Scale Blast Furnace (BF-S)

To identify the differences between the blast furnaces, it was necessary to evaluate the variables that most influenced the sulfur content in the hot metal (S_HM) from each furnace. For this purpose, the variables were ranked according to their Pearson correlation coefficient (R), with the ranking presented in the heatmap for BF-S in Figure 2, considering only variables with an absolute R value greater than 0.4, that is, those showing a moderate to strong effect on sulfur content in the hot metal.

Pearson correlation coefficients indicated that the desulfurization index had the strongest correlation with sulfur content in hot metal, followed by the ln(S)/[S] ratio. The sulfide capacity (ln Cs) also proved to be a good indicator of desulfurization efficiency. Additionally, sulfur incorporation into hot metal showed a negative correlation with hot metal carbon content (C_hot metal), slag basicities, and air temperatures, which aligns with previous studies that associate increased sulfur levels with reduced carbon

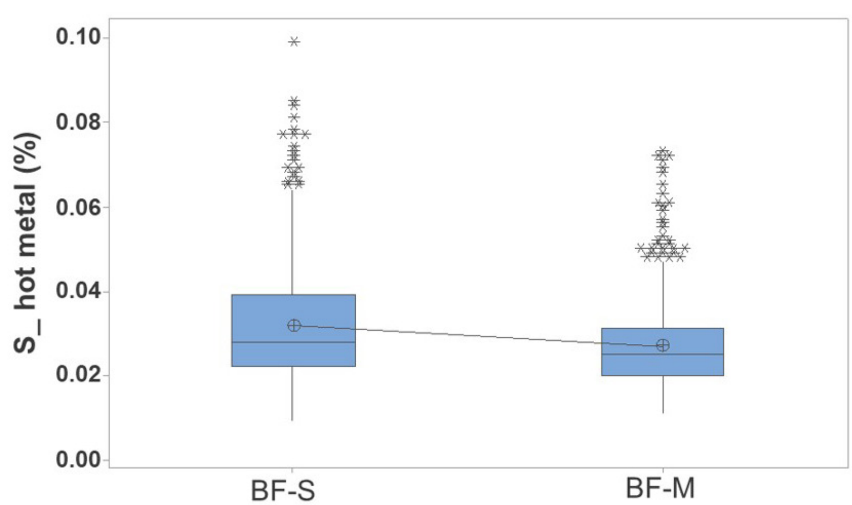

Figure 1. Comparison of sulfur content in Hot Metal from BF-S and BF-M.

saturation, lower process temperatures, and decreased basic oxide content in the slag. Among the basicity indicators, quaternary basicity (B4_slag) was the most effective for sulfur control, also highlighting the importance of maintaining CaO (CaO_slag) within an optimal range.

On the other hand, slag viscosity and FeO content (FeO_slag), as well as the percentage of yard coke usage (which is richer in ash), showed a positive correlation with sulfur content in hot metal. This supports the literature, which suggests that higher sulfur levels are related to increased sulfur input from coke and slower transfer to slag due to higher viscosity. Furthermore, increased alumina content in the slag (Al_2O_3_slag) also raised sulfur levels in hot metal, as observed by Lanna et al. [3].

Based on the correlations shown in Figure 2, a multiple regression model was developed to predict sulfur content in hot metal from BF-S, using the sulfur input into the reactor (S_{input}), furnace thermal condition (Hot metal temperature – $Temp_{HM}$), and slag chemical composition (Quaternary basicity – $B4_{slag}$). The results of the multiple regression are given by Equation (5):

$$\%S_{HM} = 0.636 + 0.0117 \times S_{input}\left(\frac{kg}{t}\right) - 0.264 \times B4_{slag} - 0.000254 \times Temp_{HM}\,(°C) \quad (5)$$

The model indicated that 64.21% (adjusted R^2) of the variability in sulfur content in hot metal is explained by these three variables. The multiple regression analysis showed that quaternary basicity had the greatest impact on sulfur content (~26%), followed by sulfur input (~12%) and hot metal temperature (~9%).

2.2.3 Medium-Scale Blast Furnace (BF-M)

The variables that most influenced sulfur content in hot metal from BF-M were also ranked according to their Pearson correlation coefficient (R), as shown in Figure 3, considering only those with an absolute R value greater than 0.4, that is, variables with a moderate to strong effect on sulfur content.

The desulfurization index was the variable most strongly correlated with sulfur content in hot metal, demonstrating its effectiveness in process control. The slag's sulfide capacity (ln Cs) and the actual ln(S)/[S] ratio also stood out as excellent indicators for sulfur control, showing stronger correlations in BF-M than in BF-S. As in BF-S, variables such as temperature, hot metal carbon content (C_hot metal), and slag basicities (B2, B3, and B4_slag) showed inverse correlations with sulfur content. Quaternary basicity (B4_slag) proved to be the most efficient indicator, emphasizing the importance of maintaining CaO (CaO_slag) at appropriate levels.

Similarly to BF-S, slag viscosity and FeO content (FeO_slag), along with sulfur input from fuel (Fuel rate), the main source of sulfur in the reactor, showed strong positive correlations with sulfur content in hot metal. These results support previous studies that associate increased sulfur levels with higher input from fuels, slower transfer to slag due to elevated viscosity, and

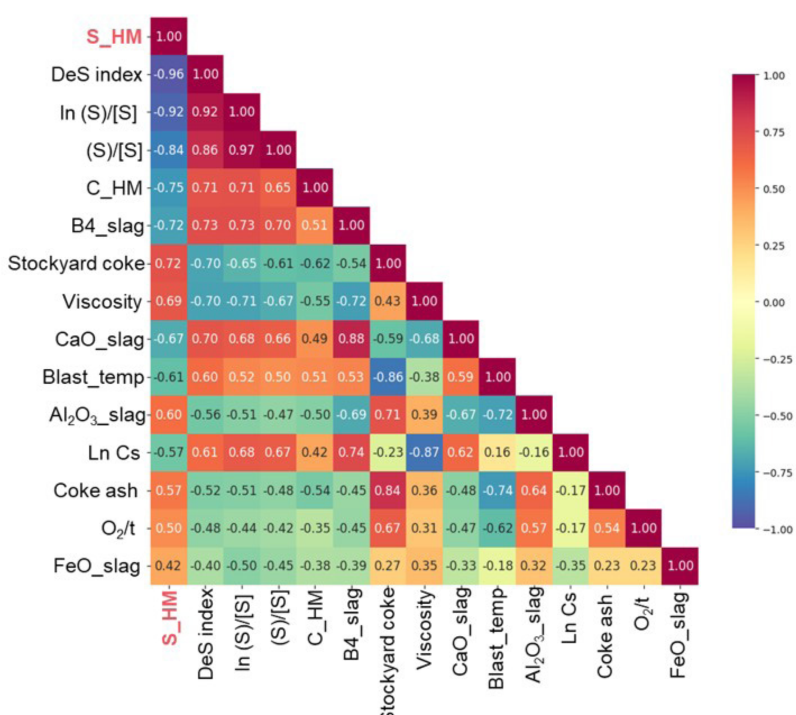

Figure 2. Heatmap of variables correlated with sulfur content in hot metal from BF-S

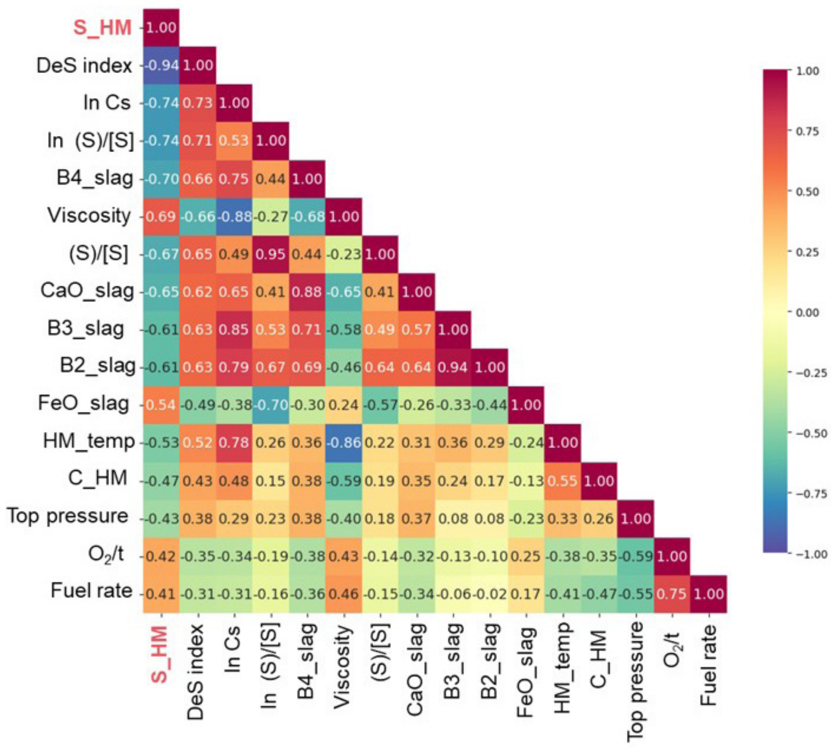

Figure 3. Heatmap of variables correlated with sulfur content in hot metal from BF-M.

increased oxygen activity in hot metal caused by higher FeO levels in basic slags.

Additionally, for BF-M, top pressure showed an inverse correlation with sulfur content in hot metal. This is due to BF-M having a counter-pressure system that maintains top pressure at a level 25 times higher than that of BF-S. This increase in top pressure improves heat recovery from the gas and makes burden reduction more efficient, resulting in lower oxygen activity in the hot metal and, consequently, lower sulfur concentration. Thus, technological differences between the furnaces, such as top pressure, are determining factors for variations in sulfur content in hot metal, something also observed for silicon in a previous study [18].

Based on the correlations shown in Figure 3, a multiple regression model was developed to predict sulfur content in hot metal, using sulfur input into the reactor, furnace thermal condition, and slag chemical composition. The results of the multiple regression are given by Equation (6):

$$\%S_{HM} = 0.4733 + 0.003164 \times \text{S}_{\text{input}} \left(\frac{\text{kg}}{\text{t}}\right) - 0.1695 \times \text{B4}_{\text{slag}} - 0.000188 \times \text{Temp}_{HM}\,(^{\circ}\text{C}) \tag{6}$$

The model indicated that 58.87% (adjusted R^2) of the variability in sulfur content in hot metal is explained by these three variables. The multiple regression analysis showed that quaternary basicity had the greatest impact on sulfur content (~22%), followed by hot metal temperature (~12%) and sulfur input (~5%). Thus, in BF-M, temperature had a greater influence than sulfur input, when compared to BF-S.

2.2.4 Alternatives for reducing sulfur in hot metal

In the mass balance performed for BF-S, the metallic burden considered consisted of 82% sinter, 7% pellet, and 11% lump ore (NPO). In the baseline scenario, the average values for slag rate, coke rate, and PCI rate were 336, 406, and 132 kg/t, respectively, with a sulfur partition of 32.15 ± 12.83, considering the mean hot metal temperature of 1491 °C.

In this base case, it was observed that approximately 80% of the sulfur input originated from coke and 20% from pulverized coal. The sulfur content in the metallic charge is relatively low, with 0.016% in the sinter, 0.029% in the pellet, and 0.086% in the lump ore.

Aligned with ESG policies, substitution scenarios were evaluated, via small-scale injection, replacing coke with charcoal (CV) in BF-S, the only viable reactor due to its lower fuel strength requirements. The substitution, in increments from 10 to 40 kg/t of hot metal (Figure 4), progressively reduced the sulfur input, resulting in a 7.3% decrease in sulfur content in hot metal in the scenario with 40 kg/t of charcoal.

Additionally, further reductions could be achieved, though with less impact, by replacing mineral coal with

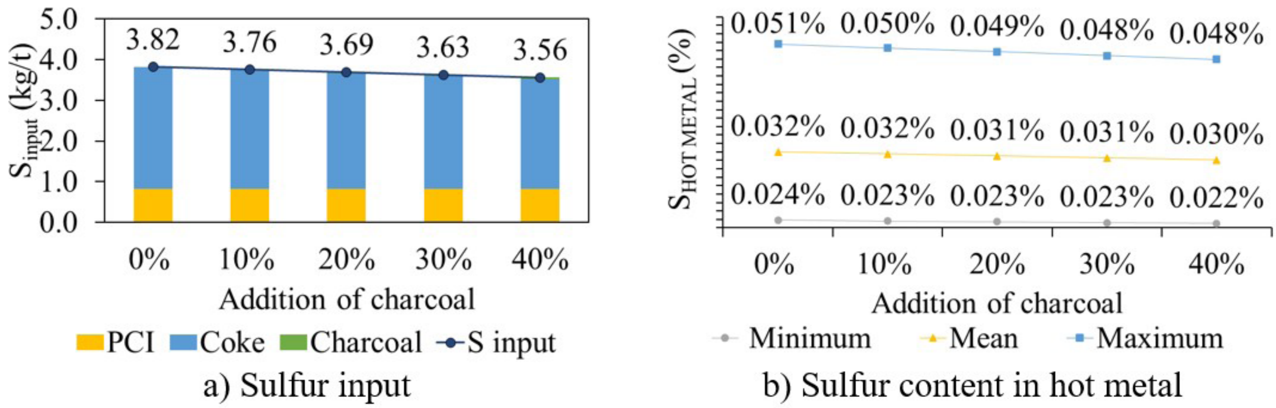

Figure 4. Impact of coke replacement with charcoal on sulfur in hot metal from BF-S

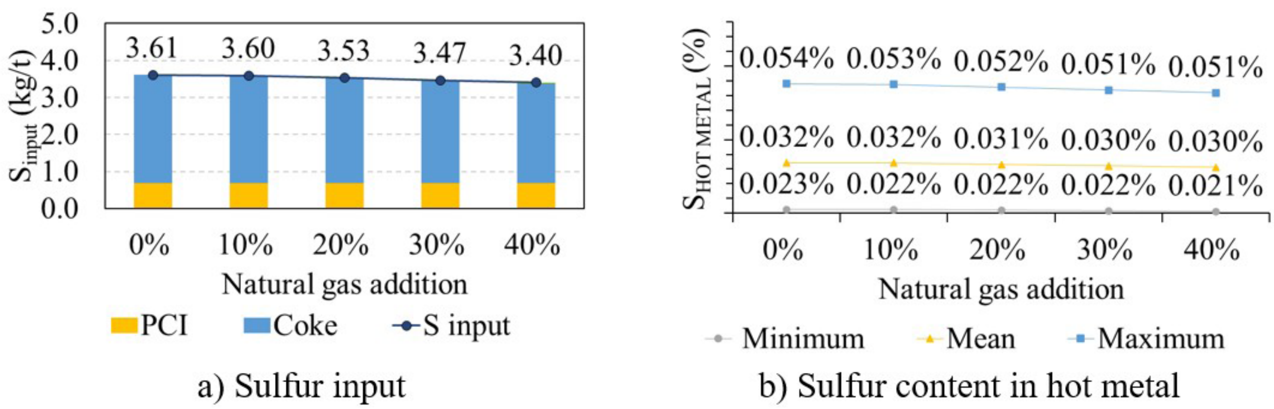

Figure 5. Impact of coke replacement with natural gas on sulfur in hot metal from BF-M

charcoal in the PCI blend, since mineral coal has a lower sulfur content (~0.65%) compared to coke (~0.75%), and charcoal is even lower (~0.10%). In this regard, replacing 40 kg/t of mineral coal with charcoal in PCI would result in an approximate 6.0% reduction in sulfur content in hot metal.

For BF-M, the metallic burden considered consisted of 84% sinter, 5% pellet, and 11% lump ore (NPO). In the baseline scenario, the average values for slag rate, coke rate, and PCI rate were 326, 403, and 110 kg/t, respectively, with a sulfur partition of 31.80 ± 14.22, considering the mean hot metal temperature of 1508°C. In line with ESG policies, coke replacement with natural gas, free of sulfur, was evaluated to analyze its impact on sulfur content in hot metal. The substitution, in increments from 10 to 40 kg/t of hot metal (Figure 5), progressively reduced the sulfur input, resulting in an 8.8% decrease in average sulfur content in hot metal with 40 kg/t of natural gas. The sulfur reduction was more significant with natural gas compared to charcoal.

3 Conclusion

Among the factors that influenced sulfur incorporation into hot metal from Usiminas' small-scale (BF-S) and medium-scale (BF-M) blast furnaces, top pressure in BF-M was a determining factor in achieving lower sulfur content. This finding reinforces that technological differences between furnaces affect the quality of the produced metal, and therefore, furnaces should be operated differently when aiming for higher-quality hot metal.

Other variables affecting sulfur incorporation were related to slag composition, furnace thermal condition, and the amount of sulfur input. Statistical models were developed to predict sulfur content in hot metal for both reactors, based on three key variables, which explained 64% and 59% of the sulfur variation in BF-S and BF-M, respectively. In this context, quaternary basicity accounted for more than 20% of sulfur incorporation in both reactors. For BF-S, sulfur input had a greater effect than hot metal temperature, whereas the opposite was observed in BF-M.

Sulfur input into the blast furnaces was primarily from fuels (80% from coke and 20% from pulverized coal). In alignment with ESG policies, the adoption of 40 kg/t of charcoal in BF-S, partially replacing mineral coal or coke, and the partially replacement of 40 kg/t of PCI with natural gas in BF-M would lead to estimated reductions of 6.0%, 7.3%, and 8.8% in sulfur content in hot metal, respectively, assuming constant desulfurization efficiency.

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