Chapter 7

Multiphase Mass Transfer in Iron and Steel Refining Processes

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Additional information is available at the end of the chapter

http://dx.doi.org/10.5772/60800

Abstract

In the present chapter, a computational fluid dynamics (CFD) model for multiphase flow was developed, allowing the simulation of two different processes, the desulfurization of hot metal in a ladle mechanically agitated by an impeller (KR process) and the desulfurization of steel in a gas-agitated ladle. The model gives important information to characterize the fluid flow conditions, to define the velocity profiles of the phases involved, and to predict the evolution of the sulfur content during the desulfurization treatments. An expression for the rate of transfer of sulfur from the metal to the slag was proposed. This expression can be used in processes where sulfur is transferred from the metal to the slag phase. The predictions of the variations of sulfur content of the metal phase with time were validated based on experimental data obtained in a Brazilian industrial plant for steel desulfurization. After the validation, the model was used to simulate the effects of several parameters and to optimize the processes. Based on these simulations, it was possible to set up the best operational conditions to improve the productivity of sulfur removal in the primary and secondary metallurgy.

Keywords: Mass transfer, CFD, desulfurization, hot metal, steel

1. Introduction

With the growing demand for high quality steels, several processes for hot metal and steel refining were developed for various purposes, such as decarburization, removal of inclusions, narrowing the range of chemical composition, thermal homogenization, and production of steels with low levels of impurities.

The efficiency and the productivity of these processes depend largely on the kinetics of the chemical reactions. Since these processes are usually developed at high temperatures, the rate-controlling step of the reactions involved is usually a mass transfer step.

In iron and steel refining, mass transfer is always a multiphase phenomenon. Different situations occur, depending on the phases involved, as follows:

- Liquid–liquid mass transfer, in the case of reactions involving liquid hot metal and steel and slag
- Liquid–gas mass transfer, when a gas is injected into or onto liquid hot metal and steel
- Liquid–solid mass transfer, when solid particles are injected into liquid hot metal and steel to promote refining reactions

In the present chapter, the importance of multiphase mass transfer in hot metal and steel refining processes will be emphasized. The fundamentals of multiphase mass transfer will be addressed, including the different techniques that are usually adopted to evaluate the mass transfer coefficient and to analyze the effects of the variables that affect its value.

Finally, two case studies, analyzing multiphase mass transfer rate during the desulfurization of hot metal and steel, will be presented and discussed.

Steels with ultralow sulfur contents are used in the manufacture of pipes for transporting oil and construction of offshore platforms, which require high impact strength and resistance to lamellar crack formation by interaction with hydrogen and sulfide inclusions.

It is well known that the efficiencies of the desulfurization of liquid hot metal and steel depends on the setting of the kinetic and thermodynamic factors that must be adjusted simultaneously to provide theoretical and practical ways to enable the optimization of the process parameters. In these case studies, a computational fluid dynamics (CFD) model for multiphase flow was developed, allowing the simulation of two different processes, the desulfurization of hot metal in a ladle mechanically agitated by an impeller and the desulfurization of steel in a gas-agitated ladle. The model gives important information to characterize the fluid flow conditions, to define the velocity profiles, and to predict the evolution of the sulfur content during the desulfurization treatments.

The model was able to predict the sulfur contents of hot metal and steel as a function of time. The predictions of the model were validated based on experimental data obtained in an industrial plant for steel desulfurization. After the validation, the model was used to simulate the effects of several parameters and to optimize the processes.

2. Multiphase mass transfer

The desulfurization of hot metal and steel is a reaction that occurs at the interface between liquid metal and liquid slag. Due to the high temperatures involved, the reaction rate is usually controlled by mass transfer between these two phases. To enhance the mass transfer rate, different methods of agitation are used. The most common are gas injection and mechanical agitation using an impeller.

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The mass transfer rate of sulfur between metal and slag is usually determined by the following relationship [1]:

$$j_S = kA\left(C_S^m - C_S^i\right) \tag{1}$$

where

j_S is the sulfur transfer rate between metal and slag (kg/s),

k is the sulfur mass transfer coefficient (m/s),

A is the interface area (m^2),

C_S^m is the sulfur concentration in the metal phase (kg/m^3), and

C_S^i is the sulfur concentration at the metal–slag interface (kg/m^3).

To apply Equation (1) in the evaluation of the mass transfer rate, the sulfur mass transfer coefficient, the interface area, and the sulfur concentration at the metal–slag interface must be known. These parameters depend on the fluid flow patterns of both phases, on the geometry of the interface, and on the partition coefficient of sulfur between metal and slag. The sulfur concentration at the metal–slag interface is usually considered the equilibrium concentration since the chemical reaction at the interface is very fast at the temperatures at which these processes are developed. To estimate the equilibrium concentration, it is necessary to know the partition coefficient of sulfur between metal and slag. This partition coefficient is a thermodynamic variable that depends on the temperature and on the chemical composition of the slag and can be estimated using data available in the literature or using thermodynamic softwares (i.e., FactSage, Thermocalc, etc.).

The fluid flow patterns of the phases involved and the geometry of the metal–slag interface can be determined by simulations using computational fluid dynamics (CFD). Independent of the method of agitation, the procedure adopted to simulate the sulfur mass transfer during hot metal or steel refining followed the same sequence of steps:

- Evaluation of the fluid flow patterns of the phases involved, solving the turbulent form of the Navier–Stokes equations for multiphase systems. In this stage, the velocities and the turbulence parameters for all phases were determined. The geometry of the metal–slag interface was also obtained. Steady-state conditions are assumed in this step.

- Calculation of the partition coefficient of sulfur between the metal and the slag. The temperature and the slag composition must be specified. In the simulations presented here, the partition coefficients were determined based on information available in the literature.

- Simulation of the sulfur content variation in the metal during the desulfurization process. Using the results obtained on the previous steps, the transient mass conservation equation for sulfur was solved. The variation of sulfur concentration as a function of time and position in the simulation domain was calculated. The variation of the average sulfur concentration in the liquid metal was then compared to some experimental results obtained in an industrial plant for validation purposes.

In the next section, two case studies of metal desulfurization are present and discussed.

3. Case studies

In this section, simulations of steel desulfurization in a gas-stirred ladle and hot metal desulfurization in a mechanically agitated ladle (KR-process) are presented and discussed. The validation of the predictions of the models using industrial data is also included. Finally, the effects of process variables on the desulfurization rate are analyzed.

3.1. Steel desulfurization in a gas-agitated ladle

Steel desulfurization is usually developed in a gas-stirred ladle. Different configurations of the gas injection system are adopted and that can affect the efficiency of the process. In the present case, the configuration considered was used in a Brazilian steelmaking industry.

3.1.1. CFD model

The ladle considered is schematically shown in Figure 1. Its main characteristics are presented in Table 1.

Parameter	Value
Height	3.3 m
Diameter at the bottom	2.32 m
Diameter at the top	2.50 m
Number of porous plugs	2
Diameter of the plugs	0.076 m
Nominal capacity	80 tons

Table 1. Main characteristics of the ladle.

Three phases were considered: liquid steel, liquid slag, and argon. The turbulent form of the conservation equations for mass, momentum, and turbulence quantities for each individual phase were solved using the commercial software Ansys-CFX. For turbulence, the standard k–ε model was used. One additional equation for sulfur mass conservation was included to simulate the transfer of sulfur between steel and slag. The transfer of sulfur to the gas phase was not considered.

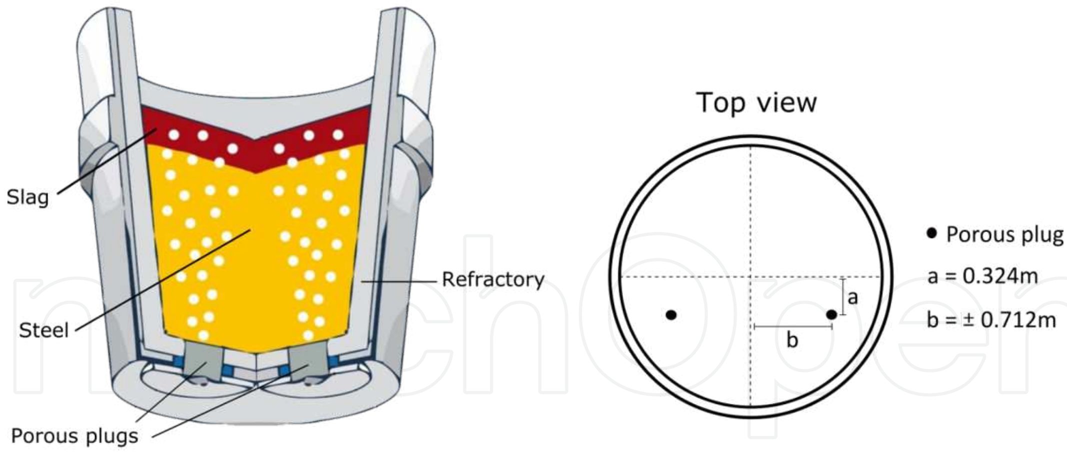

Figure 1. Schematic view of the ladle used for steel desulfurization with locations of the porous plugs at the bottom of the ladle.

Steel and slag were considered Newtonian fluids with constant density and viscosity. Argon was assumed as an ideal gas. Isothermal condition was considered in all simulations.

The diameter of the gas bubbles was considered uniform. It was evaluated using the equation proposed for liquid steel and argon [2]:

$$d_b = \left[\frac{3\sigma d_0}{\rho g} + \left(\frac{9\sigma^2 d_0^2}{\rho^2 g^2} + \frac{10Q^2 d_0}{g} \right)^{\frac{1}{2}} \right]^{\frac{1}{3}} \tag{2}$$

where

d_b is the diameter of the bubble (cm),

d_o is the diameter of the porous plug (cm),

Q is the gas flow rate (cm^3/s),

σ is the interfacial tension between liquid steel and argon (dyn/cm), and

ρ is the density of liquid steel (g/cm^3).

Nonslip conditions were assumed on all the solid walls of the ladle. At the porous plug, the gas flow rates were specified. Since the gas is injected at ambient temperature, the gas flow rates were adjusted considering the thermal expansion to the temperature of the domain. The top of the ladle was considered as an opening. The volumetric fractions of the three phases involved in the simulations were calculated in each of the control volumes of the domain. This enabled the evaluation of the contours of the phases and the interphase areas.

Grid-independent solutions were attained dividing the domain in approximately 1.5×10^6 volume elements. Figure 2 shows the mesh configuration adopted in the simulations.

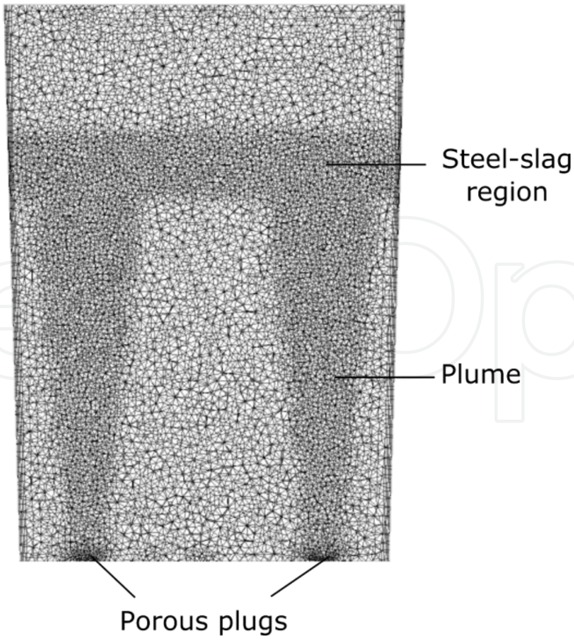

Figure 2. Mesh configuration considered in the simulations. Vertical plane passing through the porous plugs.

Nonuniform mesh was adopted, with smaller volume elements in the plume region and in the metal–slag interface.

3.1.2. Sulfur transfer model

An additional equation for sulfur transfer between metal and slag was incorporated into the model:

$$\frac{dM_S}{\mathrm{dt}} = -k_S A\left(C_{S,\mathrm{steel}} - \frac{C_{S,\mathrm{slag}}}{L_S}\right) \tag{3}$$

where

M_S the mass of sulfur transferred in the volume element interface(kg),

t is time (s),

k_S is the mass transfer coefficient for sulfur (m/s),

$C_{S,\mathrm{steel}}$ is the sulfur concentration in the metal (kg/m^3)

$C_{S,\mathrm{slag}}$ is the sulfur concentration in the slag (kg/m^3),

L_S is the partition coefficient for sulfur between slag and metal, and

A is the metal–slag interface area.

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This equation was applied in every volume element where metal and slag coexisted. The metal–slag interface area and the volume of metal in each control volume were calculated by the Ansys-CFX software.

The partition coefficient was determined according to an equation proposed by Gaye et al. [3]. The mass transfer coefficient was estimated based on the correlation proposed by Incropera and DeWitt [4]:

$$Sh = 0.029\mathrm{R}^{4/5} Sc^{1/3} \tag{4}$$

where

Sh is the Sherwood number,

Re is the Reynolds number, and

Sc is the Schmidt number.

In this equation, most of the parameters depend on the physical properties of the liquid metal and are constant. Considering this, Equation (4) can be rewritten in the following form:

$$k_s = C v_i^{4/5} \tag{5}$$

where

C is the constant and

v_i is the velocity of the metal in the interfacial region with the slag. This velocity is calculated during the CFD simulation.

The value of constant C was determined using industrial data in which samples of liquid steel were taken from the ladle desulfurization.

3.1.3. Validation of the model

To validate the predictions of the model, data from three industrial treatments were obtained. The information collected in these treatments included the following:

- Variation of the sulfur content of the liquid steel during the process (samples taken at intervals of five minutes)
- Mass of liquid steel being treated
- Mass and chemical composition of the slag
- Temperature of the process
- Argon flow rate in each porous plug

3.1.4. Results and discussion

The conditions adopted in all the CFD simulations are presented in Table 2.

Parameter	Value
Height of liquid steel	2.48 m
Mass of liquid steel	80.84 tons
Thickness of the slag layer	0.12 m
Mass of liquid slag	1430 kg
Argon flow rate	
Porous plug 1	13.2 Nm^3/h
Porous plug 2	4.8 Nm^3/h
Viscosity of liquid steel	6.5 x 10^{-3} Pa.s
Viscosity of liquid slag	0.65 Pa.s

Table 2. Conditions considered in the CFD simulations of desulfurization in a gas-stirred ladle.

Figure 3 illustrates the velocity profile of liquid steel in a vertical plane passing through the porous plugs. This profile is similar to those presented by Patil et al. [5].

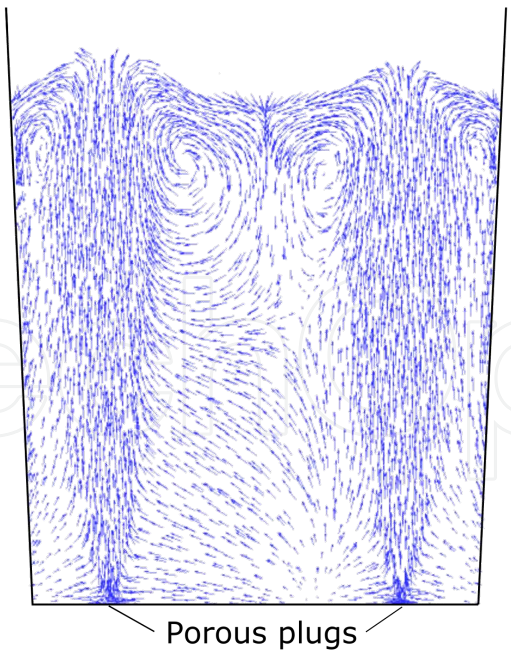

Figure 3. Velocity profile of liquid steel in a vertical plane passing through the porous plugs.

Figure 4 shows the regions of liquid steel, slag, and plume (argon) for the same vertical plane seen in Figure 3.

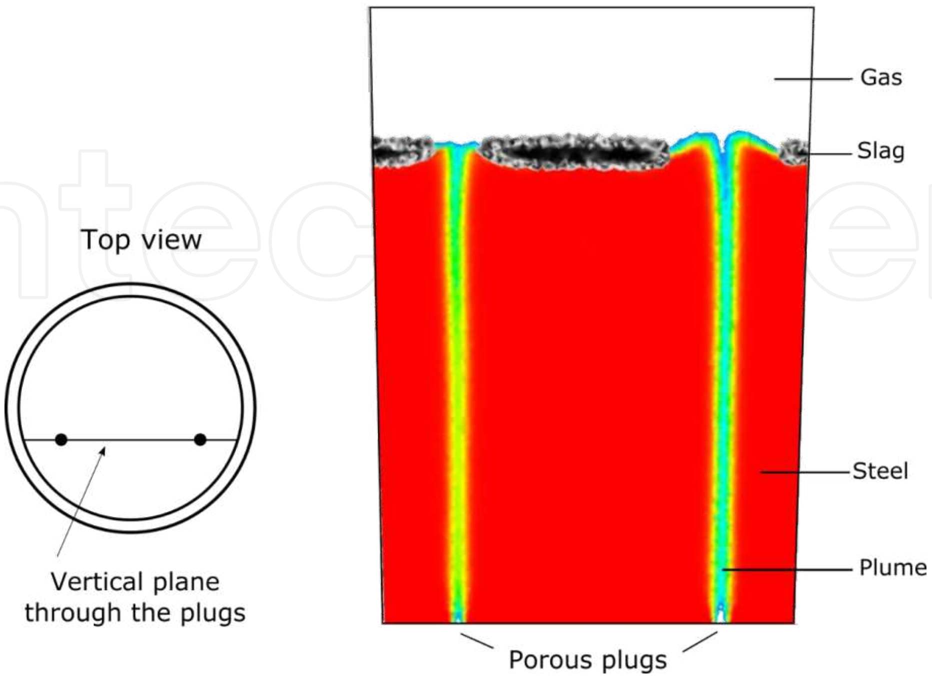

Figure 4. Regions of liquid steel, slag, and plume (argon) in a vertical plane passing through the nozzles.

For the conditions simulated, there is an “open eye” of liquid steel at the top of the ladle.

To estimate the value of constant C in Equation (5), samples of liquid steel were taken during a desulfurization treatment. The samples were taken at intervals of 5 minutes. Adjusting the simulation predictions to the experimental results, it was possible to determine the value of 1.45×10^{-3}. The composition of the slag used in this treatment was as follows (in weight percent): CaO, 54%; MgO, 5%; SiO_2, 19%; and Al_2O_3, 22%. The sulfur partition coefficient for this slag is 18.4 (according to the equation proposed by Gaye et al. [3]). With this value of constant *C*, it was possible to simulate the desulfurization process for other treatments. The results obtained for one of these treatments are illustrated in Figure 5. The experimental results are also included in the figure. In this case, the slag composition was as follows: CaO, 57%; MgO, 8%; SiO_2, 10%; and Al_2O_3, 25%. The partition coefficient was 52.

The model predictions reproduce very well the experimental data. Based on these results, the model was validated, and the effects of different parameters were analyzed. The effects of argon flow rate, slag composition, and slag thickness are presented here. These results were compared to the predictions using the conditions specified in Table2 (reference case).

Figure 6 shows the effect of the total argon flow rate on the variation of the sulfur content in the liquid steel. Only the last 15 minutes of the simulations are presented. In the range tested, there is only a slight effect of the argon flow rate on the sulfur contents. A significant increase in the argon flow rate does not have an important impact on the desulfurization but increases the area of the “open eye” of liquid steel, which can have deleterious effects on the steel quality.

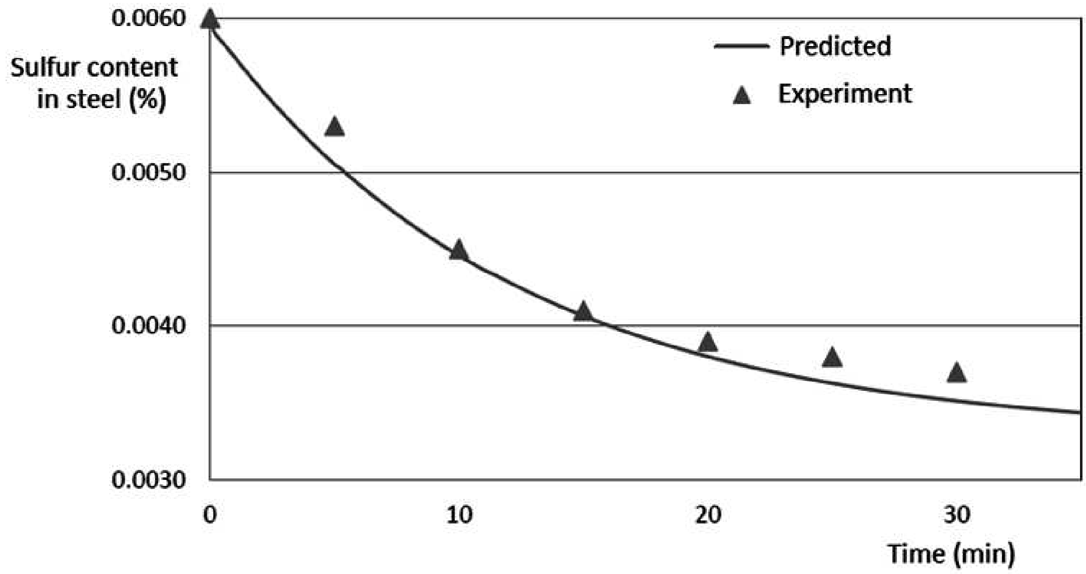

Figure 5. Comparison of the variations of sulfur content of the liquid steel as a function of time predicted by the model and determined in the experiment.

This effect of the argon flow rate can be explained in terms of the increase of velocities of the steel at the metal–slag interface, which increases the mass transfer coefficient, and in terms of faster homogenization of the sulfur content of the steel inside the ladle.

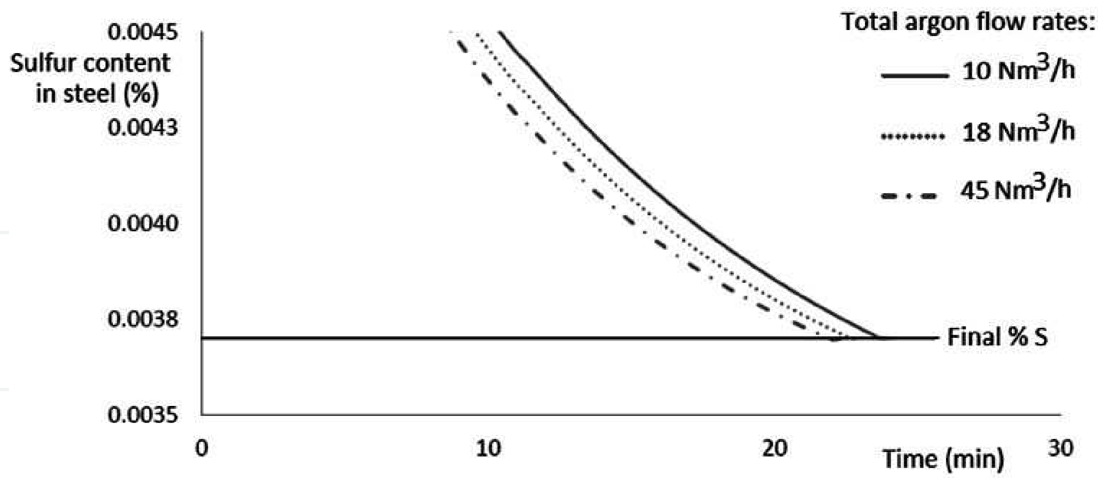

Figure 6. Effect of the total argon flow rate on the desulfurization process.

Figure 7 illustrates the effect of slag composition. Slag 1 is the one considered in Figure 5. Slag 2 has a partition coefficient of 52. An increase in the partition coefficient leads to a pronounced increase in the desulfurization rate. The time to reach a sulfur content of 35 ppm is 15 minutes for slag 2 and 24 minutes for slag 1. This could lead to a significant increase in productivity of the steel plant.

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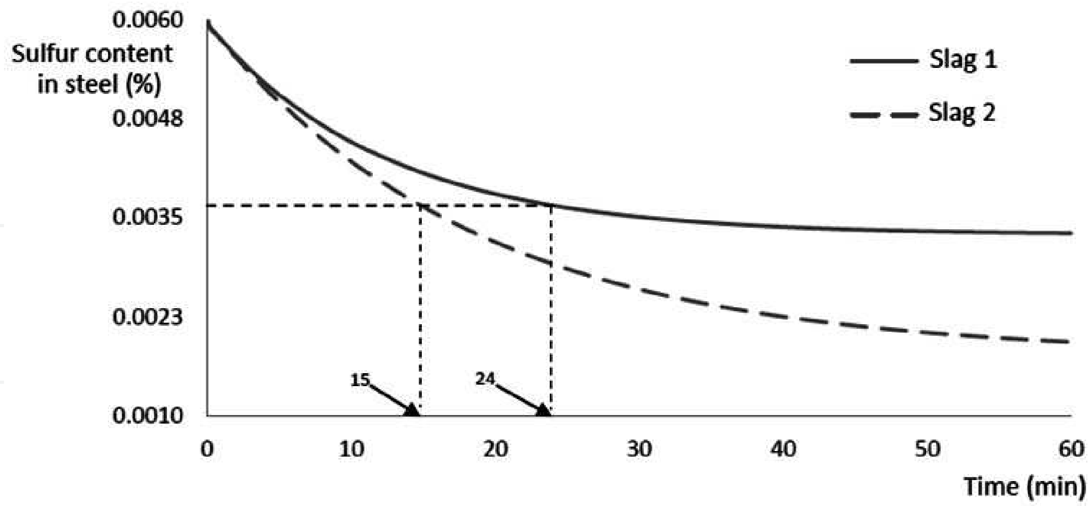

Figure 7. Effect of the slag partition coefficient on the desulfurization process.

Finally, the effect of the thickness of the slag layer is presented in Figure 8.

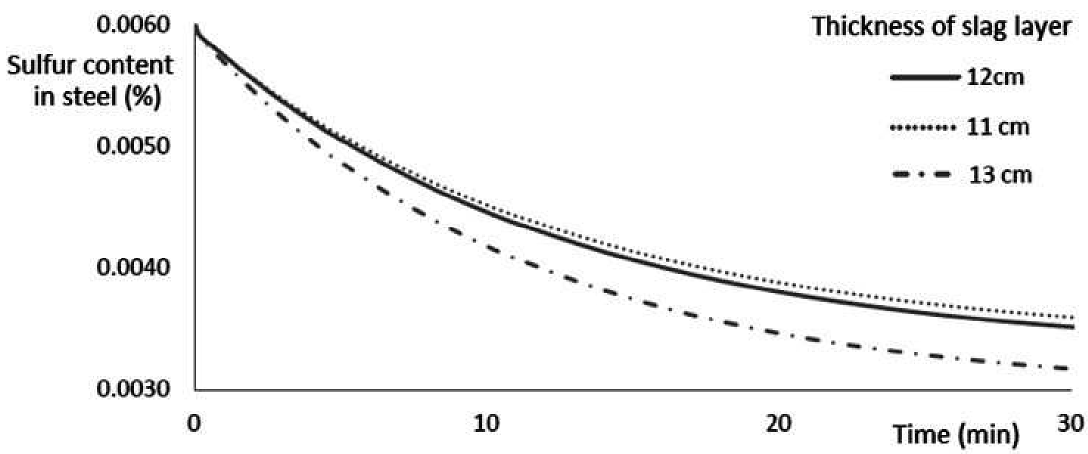

Figure 8. Variation of the sulfur content for different thicknesses of the slag layer.

As shown, an increase in the slag thickness has a positive effect on the desulfurization rate. With more slag, the sulfur concentration in the slag phase is smaller, and this leads to an increase in the driving force for sulfur transfer from steel to slag. Another benefit of a thicker slag layer is the reduction of the area of the "open eye."

3.2. Hot metal desulfurization in a mechanically agitated ladle

The KR process is largely used to promote hot metal desulfurization. In the process, an impeller is used to stir the liquid metal and to enhance the contact between metal and slag. Among the several variables that affect the desulfurization rate, the penetration in the liquid metal and position along the radius and the rotation speed of the impeller were chosen to be investigated.

The geometries of the ladle and of the impeller were specified according to the design currently being used in a Brazilian industry. The simulations followed the same steps described for the analysis of steel desulfurization in gas-stirred ladle.

3.2.1. CFD model

The geometries of the ladle and of the impeller are schematically shown in Figure 9. Their main dimensions are given in Table 3.

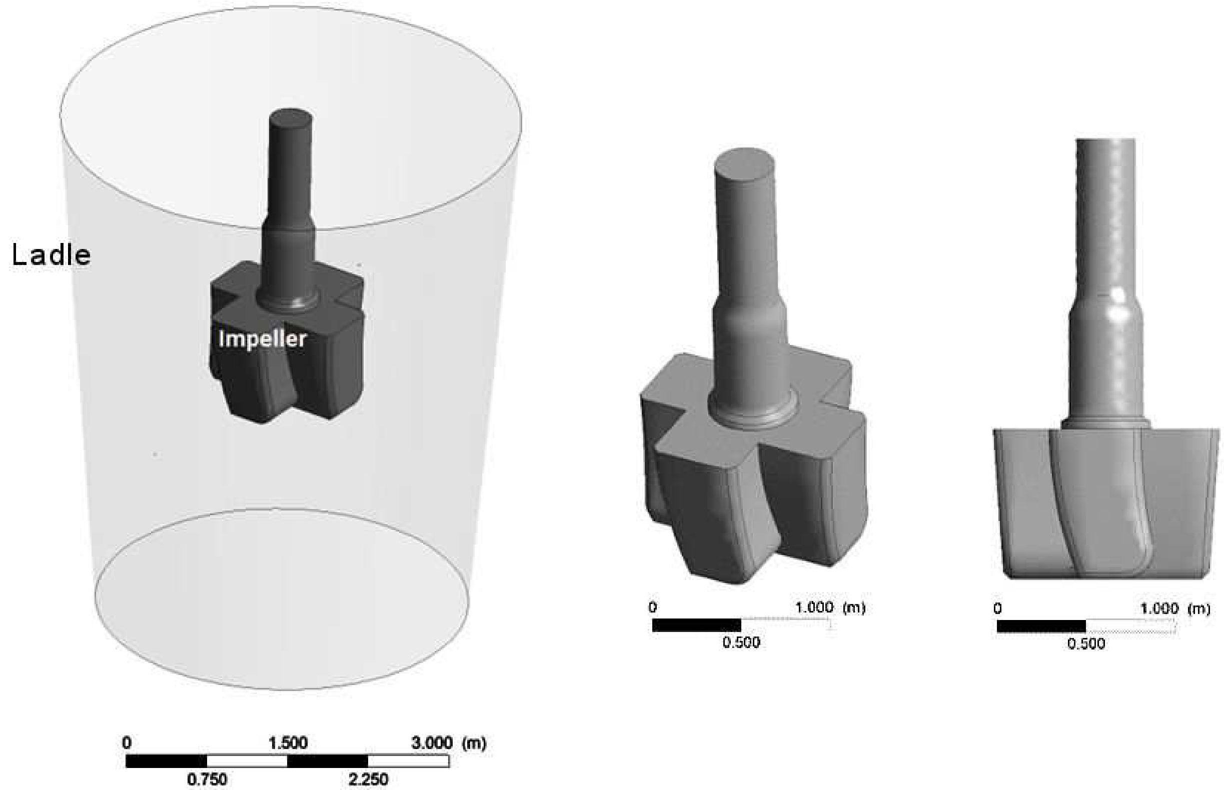

Figure 9. Schematic view of the ladle and of the impeller.

The CFD model is similar to that for steel desulfurization described previously. Since there is no gas injection in the KR process, the conservation equations were solved only for the hot metal and for the slag. A free surface model was implemented to identify the interfaces between the phases. The commercial software Ansys-CFX was used in all the simulations.

Hot metal and slag were assumed as Newtonian fluids with constant density and viscosity. These two phases were in thermal equilibrium.

For boundary conditions, nonslip conditions were considered on all the solid walls, including the surface of the impeller. The rotation speed of the impeller was specified.

The equation for sulfur transfer between the hot metal and the slag is similar to that used for desulfurization. The partition coefficient was also calculated using the same method shown

	Parameter	Value
Ladle	Height	5.10 m
	Diameter at the bottom	3.45 m
	Diameter at the top	4.30 m
	Nominal capacity	315 tons
Impeller	Top radius	1.50 m
	Bottom radius	1.35 m
	Length	1.00 m
	Radius of the axis	0.25 m

Table 3. Dimensions of the ladle and of the impeller.

by Gaye et al. [3]. The mass transfer coefficient was estimated according to Equation (5), with a value of 1.45×10^{-3} for C.

Grid-independent solutions were attained dividing the domain in approximately 1.2×10^{6} volume elements. Figure 10 shows the mesh configuration adopted in the simulations.

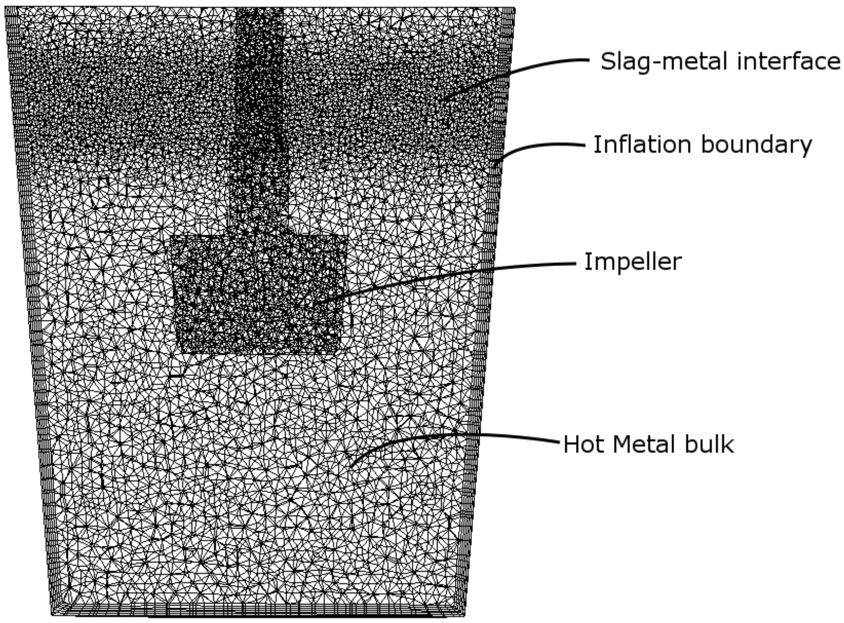

Figure 10. Mesh configuration considered in the simulations. Vertical plane passing the center of the ladle.

Nonuniform mesh was adopted, with smaller volume elements near the walls and in the metal–slag region.

3.2.2. Results and discussion

The conditions adopted in the CFD simulations are presented in Table 4.

Parameter	Value
Height of hot metal	4.20 m
Mass of hot metal	315 tons
Thickness of the slag layer	0.12 m
Mass of liquid slag	3500 kg
Viscosity of hot metal	6.5 x 10-3 Pa.s
Viscosity of liquid slag	0.65 Pa.s

Table 4. Conditions considered in the CFD simulations of desulfurization of hot metal in the KR process.

The impeller was located at the center of the ladle. The rotation speed was 40 rpm. The distance between the bottoms of the impeller and of the ladle was 2.5 m. These conditions are assumed as a reference to analyze the effects of some process variables.

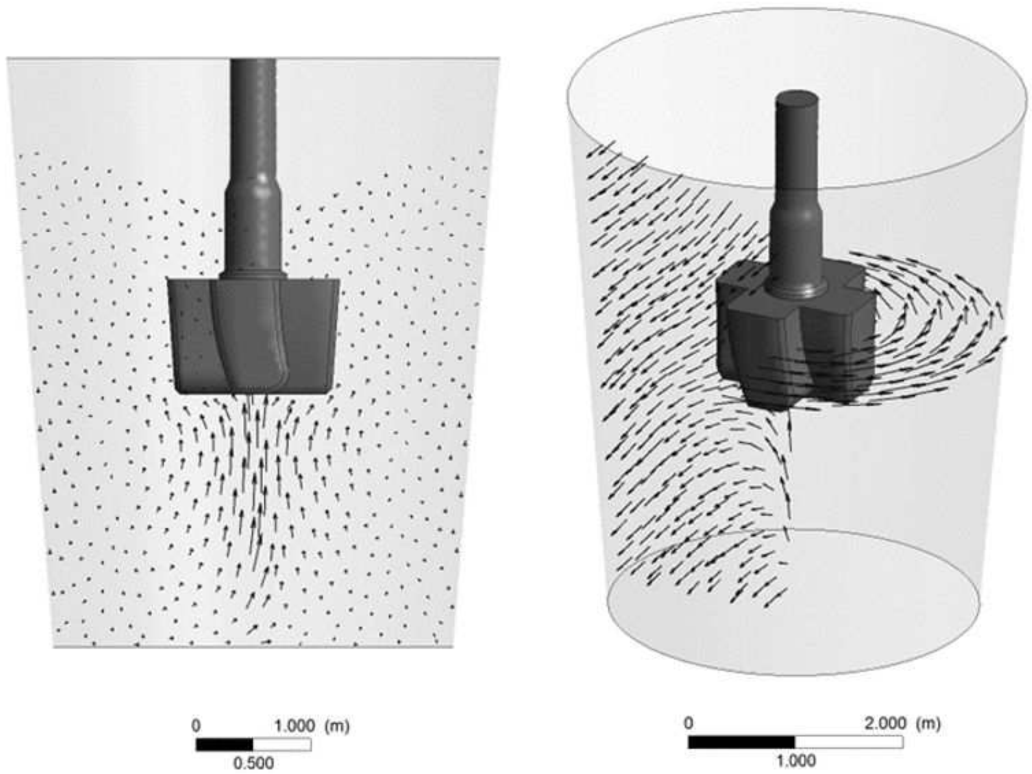

Figure 11. Velocity profiles of liquid hot metal in a vertical plane passing through the center of the ladle and in a horizontal plane passing through the impeller.

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Figure 11 illustrates the velocity profile of liquid hot metal in a vertical plane passing through the center of the ladle and in a horizontal plane passing through the impeller. This profile is similar to those presented by Shao et al. [6]. There is an upward flow underneath the impeller. The rotation of the impeller also induces a rotation flow of hot metal. This rotation affects the pressure field and leads to a reduction of the level of hot metal in the center and an increase near the wall of the ladle, as seen in Figure 12, which shows the regions of liquid hot metal and slag for a vertical plane passing through the center of the ladle. This variation in the level of the hot metal significantly increases the interface area between metal and slag and increases the desulfurization rate.

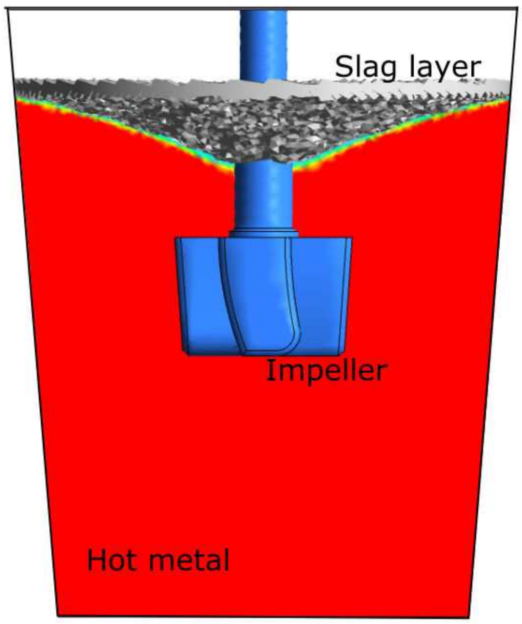

Figure 12. Regions of liquid hot metal and slag in a vertical plane passing through the center of the ladle.

Figure 13 shows the results of the simulation of the desulfurization of the hot metal for the reference conditions. The reduction of sulfur content is much faster than that observed with steel. The main reason for that is the higher initial sulfur content of the hot metal, which gives a more significant driving for sulfur transfer to the slag. As seen, a 5-minute treatment is sufficient to reduce the sulfur content from 300 to approximately 50 ppm. This result is consistent with data available in the literature [7–9].

Using the mathematical model, the effects of some process variables were investigated. Figure 14 shows the effect of positioning the impeller with its axis of rotation 40 cm off center. Compared to the impeller located at the center of the ladle, there is a slight reduction in the desulfurization rate. Although it was not considered in the model, locating the impeller off center might also increase refractory wear due to higher velocities near the wall of the ladle.

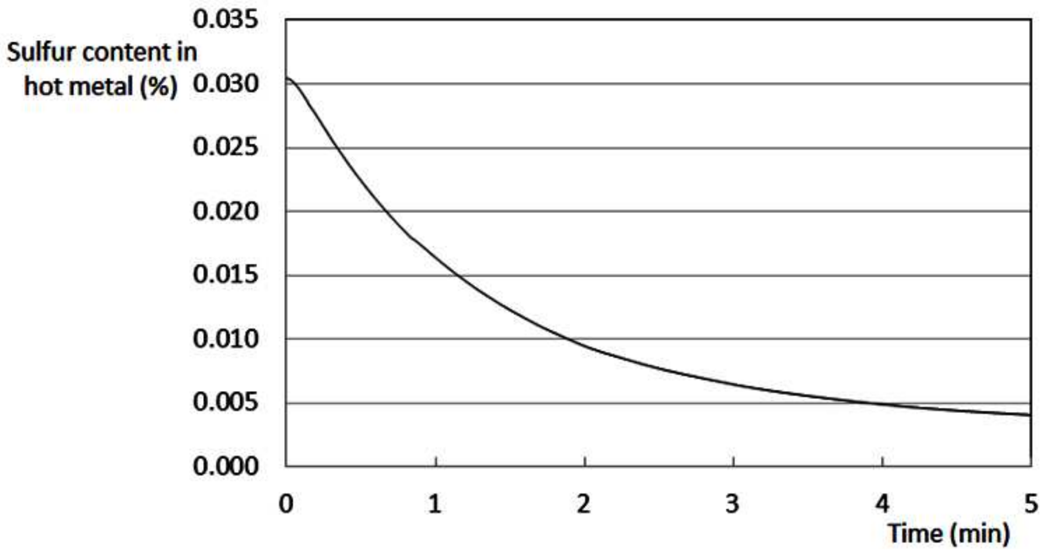

Figure 13. Variation of the sulfur content in the hot metal during the desulfurization. Reference conditions.

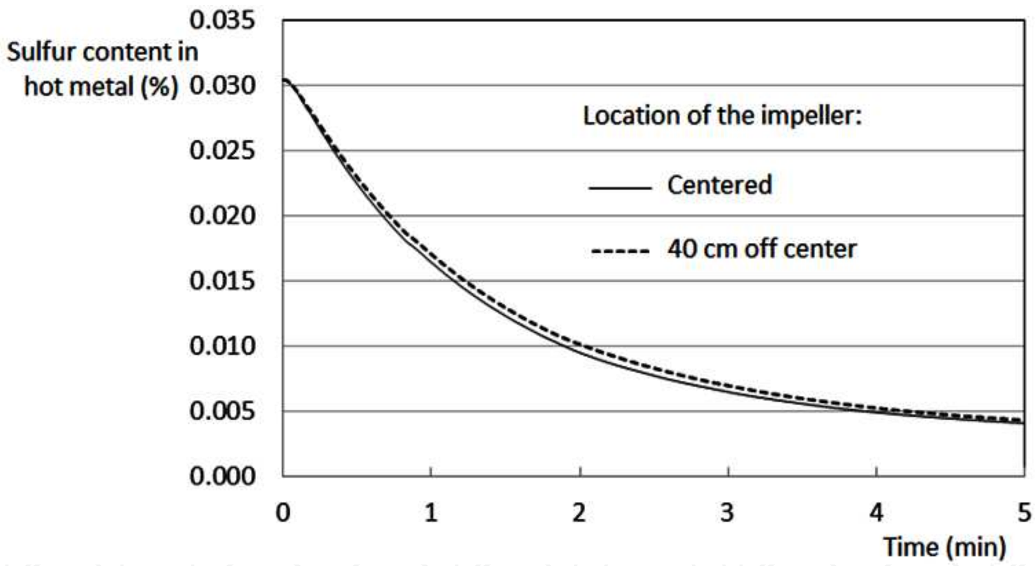

Figure 14. Effect of the location of the impeller on the variation of sulfur content of the hot metal.

The effect of the immersion depth of the impeller is presented in Figure 15. In this case, the immersion depth was increased in 40 cm (the distance between the bottoms of the impeller and of the ladle was reduced to 2.1 m). Increasing the immersion depth reduces the velocities in the region close to the interface between the hot metal and the slag. This leads to a decrease in the sulfur mass transfer coefficient and has a negative effect on the desulfurization rate.

Figure 16 illustrates the effect of an increase of 10 rpm in the rotation speed of the impeller (from 40 to 50 rpm). Among the variables analyzed, the rotation speed presented the most significant effect. With a rotation speed of 50 rpm, the velocities near the metal–slag interface increase and so does the mass transfer coefficient. Together with a faster homogenization of

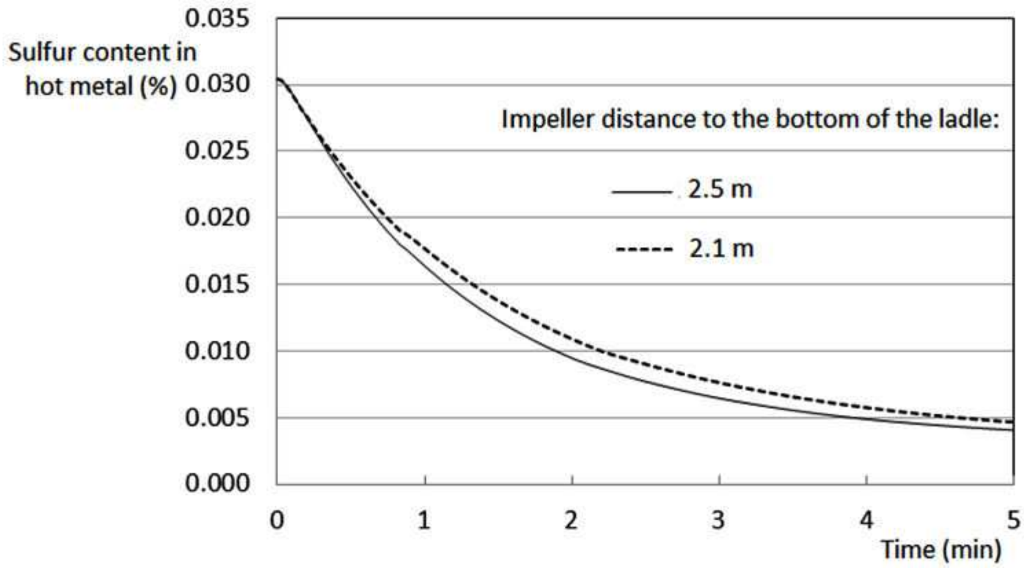

Figure 15. Variation of sulfur content of the hot metal for two immersion depths of the impeller.

the sulfur of the hot metal, the consequence is a faster desulfurization. Considering a final content of sulfur of 50 ppm, it is observed that an increase in the rotation speed to 50 rpm can lead to approximately a 1-minute reduction in the treatment time, with possible increase of productivity.

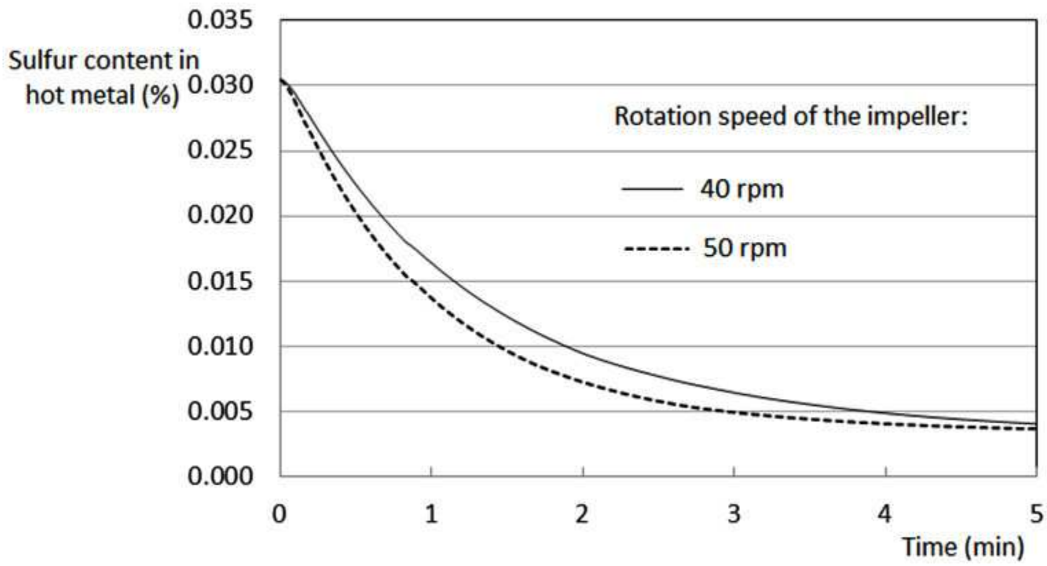

Figure 16. Effect of the rotation speed of the impeller on the variation of sulfur content of the hot metal.

Based on factor analysis, an equation to predict the sulfur content after a treatment of five minutes was determined:

$$S_{ppm} = 300.13 - 87.45d_B - 4.47r_S + 9d_{off} + 1.45d_Br_S \quad (6)$$

where

S_{ppm} is the sulfur content after 5 minutes of treatment in the KR process (ppm),

d_B is the distance between the bottoms of the impeller and of the ladle (m),

r_S is the rotation speed of the impeller (rpm), and

d_{off} is the off-center distance of the axis of the impeller (m).

4. Conclusions

Mass transfer plays a significant role in the kinetics of steelmaking processes. In these processes, mass transfer is usually a multiphase phenomenon, and its rate is affected by the flow conditions of the phases involved.

In the production of high-quality steels, especially those with very low sulfur content, desulfurization must be implemented. The desulfurization of hot metal and liquid are both commonly used. In most processes, desulfurization is promoted by transferring sulfur from the metal to a refining slag.

In the present chapter, the desulfurization of steel in a gas-stirred ladle and of hot metal in the KR process was studied by mathematical modeling. The model developed involved the simulation of the flow field of the different phases involved, coupled with a mass conservation equation for sulfur, which included an expression for the rate of transfer of sulfur from the metal to the slag. The commercial software Ansys-CFX was used to solve the turbulent form of the Navier–Stokes equations for multiphase flows. The predictions of the model were validated using industrial data of steel desulfurization.

For the desulfurization of steel in a gas-stirred ladle, it was shown that increasing the sulfur partition coefficient and the thickness of the slag layer at the top of the ladle have both positive effects on the desulfurization rate. The gas flow rate, in the range tested, presented a minor effect, but very high gas flow rates lead to an increase in the "open eye" area of liquid steel at the top of the ladle, which can have deleterious effects on the steel quality.

For the desulfurization of hot metal in the KR process, the effects of variables related to the impeller position and rotation speed were investigated. The predictions of the model indicated that, in the ranges tested, the rotation of the impeller has the most significant effect. Increasing the penetration depth of the impeller and locating its axis of rotation off center in the ladle have slight negative effects on the desulfurization rate.

Acknowledgements

The financial support of FAPEMIG in the form of a research grant to R. P. Tavares (Process No. TEC-PPM-00118-13) is gratefully acknowledged. The authors also acknowledge the

http://dx.doi.org/10.5772/60800

financial support of CAPES/PROEX to the graduate program. The scholarship from CNPq to the first author is also gratefully acknowledged.

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