A New Numerical Model for Predicting Carbon Concentration during RH Degassing Treatment

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A new decarburization model for the RH system was constructed to calculate the decarburization reaction using a three-dimensional fluid dynamics program. The behavior of Ar gas and the entire fluid flow in the RH process were considered. On the basis of this fluid flow, the decarburization reaction could be calculated by establishing the decarburization reaction model, which considers thermodynamics, reaction kinetics, and the decarburization sites (Ar bubble surface, bath surface, and the CO bubble formation at inner sites). The model showed good agreement with two RH operation results. Using this numerical model, the contributions of each reaction were investigated.

KEY WORDS: decarburization; reaction model; reaction site; reaction area; reaction rate.

1. Introduction

The RH process plays an important role in the steel refining process in terms of degassing, mixing and decarburization before continuous casting process. In particular, decarburization is the main function of the RH process for producing the ultra low carbon steel.

Although the RH system can treat a larger quantity of melt compared with other refining processes, the decarburization rate of a steel melt with low carbon content has a tendency to be low. Therefore, the decarburization rate has been one of the main issues in the RH process for increasing the steel productivity.

Because many parameters affect the decarburization rate, estimating the effect of each parameter is difficult. Investigations on the decarburization reaction in the RH process can be divided into two categories.

One category analyzes the reactions through experiments on field operations or model systems. In particular, analyses of the carbon and oxygen concentration acquired during the RH operations have been made. These studies established the decarburization model based on an assumed circulation flow rate estimated by empirical equations. Kishimoto et al. carried out small scale experiments and evaluated the factors that influence the decarburization rate. These studies can qualitatively explain the phenomenological behavior of the carbon concentration. However, there are limitations in applying these results to general RH systems with various shapes and different operational conditions. This is because it is difficult to evaluate the actual effect of each parameter such as the shape and operational conditions.

The other reports have calculated the fluid flow and the carbon concentration during the RH process. However, the previous studies considered gas behavior only for decarburization reaction and the fluid flow was calculated by assuming the circulation flow rate. Therefore, the effects of parameters, which affect both the fluid flow and decarburization rate, could not be evaluated properly.

In this study, the gas behavior for the fluid flow and decarburization rate was modeled. First, the plume shape and bubble expansion were calculated. From these results, the buoyancy force, which acts as the driving force for melt circulation was calculated. In addition, decarburization models at each reaction site were established with some assumptions.

By combining the Ar gas behavior, fluid flow and decarburization model of the RH system, a numerical model, which can calculate the decarburization reaction considering the configurations of the system and operation conditions, were developed.

2. Calculation of Gas Behavior and Fluid Flow

In the RH process, Ar gas injected at the snorkel wall causes strong fluid flow. The melt circulates from the ladle to the vacuum vessel through the snorkels. The decarburization reaction occurs near the vacuum bath surface and at the rising Ar bubble surface.

The decarburization reaction rate is affected by mass transport and the concentration distribution of the dissolved carbon and oxygen in the melt. Therefore, fluid flow is important for calculating the decarburization reaction. In this study, fluid flow was simulated by calculating the Ar bubble behavior. The details for the calculation methods of the Ar gas behavior and fluid flow are represented in Ref. 9).

Briefly, the Ar buoyancy force was calculated from the drift flux model as shown in Eq. (1).

$$f = g \left( \rho_g - \rho_f \right) \cdot V_g \cdot \alpha (1 - \alpha) \quad \ldots \quad (1)$$
where \( g \) is the acceleration due to gravity (m/s\(^2\)), \( \rho_g \) and \( \rho_m \) are the densities of the Ar gas and the melt (kg/m\(^3\)), \( V_p \) is the volume of the plume (m\(^3\)) and \( \alpha \) is the average volume fraction of the gas. The plume volume was calculated from the plume shape, which is affected by the pressure of the vacuum vessel, nozzle number and position, gas flow rate etc. The average volume fraction is the total volume ratio of gas in the plume realm. In addition, gas bubble expansion due to the pressure drop and heat transfer from melt during rising was calculated and this effect was considered when calculating the total buoyancy force. In the Ref. 9), the bubble expansion due to only pressure drop was considered. In this study, the numerical model was modified to consider both the thermal expansion and pressure drop, Szekely’s model\(^{11}\) is modified as

\[
R_p^2 + \frac{3}{2} R_p^2 = \frac{1}{\rho_p R_p} \left[ P_0 \left( \frac{R_{p0}}{R_p} \right)^3 \left( \frac{T}{T_0} \right) - P_0 + \rho_g \rho_m \alpha \right]
\]

\[
P_p = P_v + \rho_g \rho_m \alpha (r = 0; \quad R_p = R_{p0}; \quad R_p^2 = 0)
\]

In Eq. (2), \( \rho_p \) is the density of the melt, \( R_p \) is the radius of the bubble, \( T \) is temperature in the bubble, \( R_{p0} \) and \( T_0 \) are radius and temperature at the nozzle respectively, \( P_0 \) is the pressure of the vacuum vessel and \( H \) is the height of the melt. To solve Eq. (2), the temperature should be known. The temperature of bubble was estimated by following equation.

\[
\rho_g C V \frac{dT}{dt} = h (T_{melt} - T)
\]

where \( \rho_g \) is the density of gas (kg/m\(^3\)), \( V \) is the volume of bubble (m\(^3\)) and \( C \) is the heat capacity (J/kg·K). \( h \) is the heat transfer coefficient (J/m\(^2\)·s·K) and can be derived from the penetration theory with the assumption of gas phase control. Komarove et al. derived heat transfer coefficient from experiments.\(^{12}\) Because the analysis in the Ref. 12) was based on spherical and constant size bubble, there should be some difference in the value of \( h \) and further study should be necessary to derive more realistic \( h \) value.

The buoyancy force calculated by Eq. (1) was inserted as the external force for the velocity components \((u, v, w)\) in the three dimensional Navier-Stokes equation to calculate fluid flow. The numerical model can predict the effects of the operational conditions and shapes of the RH system on fluid flow\(^{13}\).

3. Calculation of Decarburization Reaction

In this study, the decarburization reaction was simulated by calculating the dissolved carbon concentration during the RH process. The numerical model included mass transport of dissolved carbon and oxygen due to convection and diffusion and the reaction rate at the reaction sites. The reaction rate was calculated respectively at each reaction site considering the reaction area, rate determining step and thermodynamics.

3.1. Governing Equation

The governing equation in a cylindrical coordinate for calculating the concentration distribution of carbon and oxygen is represented as

\[
\frac{\partial (\rho C)}{\partial t} + \frac{1}{r} \frac{\partial (\rho u C)}{\partial r} + \frac{1}{r} \frac{\partial (\rho v C)}{\partial \theta} + \frac{\partial (\rho w C)}{\partial z} = D \frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial C}{\partial r} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( \Gamma_c \frac{\partial C}{\partial \theta} \right) + \frac{\partial}{\partial z} \left( \Gamma_c \frac{\partial C}{\partial z} \right) + S_c
\]

In Eq. (4), \( C \) represents the concentration of carbon and oxygen, \((u, v, w)\) is the velocity components (m/s) and \( \rho \) is the density of the melt (kg/m\(^3\)). \( \Gamma_c \) is the source or sink of the mass (kg/m\(^3\)·s), and the transport constant \( \Gamma_c \) is expressed by following equation.

\[
\Gamma_c = \rho \cdot D + \frac{\mu_t}{S_{c_t}}
\]

where \( D \) is the diffusion coefficient of carbon and oxygen (m\(^2\)/s), \( \mu_t \) is the turbulent viscosity (N·s/m\(^2\)) and \( S_{c_t} \) is the turbulent Schmit number.\(^{14}\)

Equation (4) was solved using the finite difference method. The velocity components and turbulent properties were calculated in the previous step for the fluid calculation.

3.2. Decarburization Reaction Site

In the decarburization reaction during the RH process, three reaction sites can be considered, as shown in Fig. 1.

At the bath surface, dissolved carbon and oxygen transfer to the bath surface and the decarburization reaction occurs. The CO gas formed moves into gas phase. At the rising bubble surface, the same reaction occurs as at the bath surface. One difference is that the CO partial pressure of the bubble and reaction area changes as the bubble rises. The decarburization reaction can occur in the melt through the formation of CO bubbles.

In order to establish the reaction model at each site, the thermodynamics, reaction site area, reaction rate controlling step and the reaction rate were considered.

Fig. 1. Decarburization reaction site in RH process.
3.3. Thermodynamics of Decarburization Reaction

The decarburization reaction in the reaction site can be represented by the following equation.

$$[C] + [O] = CO(g) \quad \text{ equation from the equilibrium constant represented by Eq. (6).}$$

In this reaction, the equilibrium constant \( K \) is expressed as following\(^{13)\)

$$K = \frac{P_{CO}}{a_C a_O} = 10^{[160.7 + 2.003]}$$

where,

\[ a_C = f_C [wt\%C] \]
\[ a_O = f_O [wt\%O] \]
\[ \log f_C = e_C [wt\%C] + e_f [wt\%O] \]
\[ \log f_O = e_0 [wt\%C] + e_0 [wt\%O] \]

In Eq. (7), \( a \) is the activity, \( f \) is the activity coefficient and \( e \) is the interaction parameter. Many studies have evaluated the interaction parameter. In this study, the values are adopted from reference, which are \( e_C = 0.243, e_O = -0.32, e_C = -0.17 \) and \( e_O = -0.421.\(^{13)\}

3.4. Reaction Model at Each Site

3.4.1. Bath Surface

There are two assumptions in the decarburization model at the bath surface. The first assumption is that that reaction represented in Eq. (6) reaches equilibrium at the local reaction site. The second is that the rate-determining step is the transport of dissolved carbon and oxygen to the reaction surface.

Because the decarburization reaction occurs at a very high temperature, the chemical reaction rate must be very high. Furthermore, the CO gas formed at the reaction surface may also transfer at high speed into the gas phase because the pressure of the vacuum vessel is very low.

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In the real RH operation, the initial concentration of dissolved oxygen is generally higher than that of dissolved carbon. Therefore, it is possible to assume that the transport of dissolved carbon is the rate-determining step for the bath surface reaction and the reaction rate can be calculated using the following equation.

Reaction rate (kg/s) \( = \frac{A_r \cdot \rho_{melt} \cdot h_c}{100} \)

where, \( A_r \) is the reaction area (m\(^2\)), \( \rho_{melt} \) is density of the melt (kg/m\(^3\)) and \( h_c \) is the mass transfer coefficient of dissolved carbon (m/s). \([wt\%C]_n\) is the carbon concentration of the melt near the reaction surface and is assumed to be the concentration of the surface grid in the numerical model. \([wt\%C]_i\) is the equilibrium concentration of carbon at the reaction site and is calculated by the following equation from the equilibrium constant represented by Eq. (7).

\[ [wt\%C]_i = \frac{P_{CO}}{K \cdot f_C \cdot f_O \cdot [wt\%O]} \] (9)

In Eq. (9), \( P_{CO} \) is the partial pressure of CO gas at the reaction surface in the gas phase and \([wt\%O]\) is the oxygen concentration at the reaction surface. In the numerical calculation, the oxygen concentration at the reaction surface was assumed to be the same as that of the bath surface grid.

The partial pressure of CO gas can be expressed by the following equation with the assumption that the gas fraction is same all over the vessel surface:

\[ P_{CO} = \frac{n_{CO}}{n_{total \ gas} \cdot P_{vacuum}} \] (10)

where, \( n_{CO} \) is the number of moles of CO gas that has escaped per unit time, which was calculated by summing the reaction quantities of CO gas at three reaction sites. \( n_{total \ gas} \) is the sum of the number of moles of CO and Ar gas that escaped per unit time at the bath surface. \( P_{vacuum} \) is the pressure of the vacuum vessel (N/m\(^2\)).

In order to calculate the decarburization rate using Eq. (8), the reaction area and mass transport coefficient of the carbon must be evaluated. In this study, the reaction area was assumed to be same as the bath surface area. The mass transport coefficient was evaluated by comparing the calculation result with real RH operation data and was calculated to be 0.005 m/s. This value is larger than the value of 0.0015 m/s used by S. Kitamura,\(^{14)\} \) and T. Kitamura.\(^{15)\} \) The difference with this study may be due to the estimation of reaction area of vessel surface. S. Kitamura and T. Kitamura estimated the reaction area to be larger than bath surface area.

3.4.2. Ar Bubble Surface

The decarburization reaction occurs at the rising Ar bubble surface. The reaction model of the Ar bubble surface reaction is similar to that of the bath surface reaction. The reaction rate at the Ar bubble surface is represented by the following equation.

\[ \text{Reaction rate (kg/s)} = \frac{A_r \cdot \rho_{melt} \cdot h_c}{100} \cdot ( [wt\%C]_n - [wt\%C]_b ) \] (11)

where, \( A_r \) is the surface area of the Ar bubble (m\(^2\)), \([wt\%C]_b\) is the carbon concentration near the bubble and \([wt\%C]_n\) is the equilibrium carbon concentration at the bubble surface.

The reaction surface area can be calculated from the bubble size. The bubble size was calculated in the previous step for calculating the gas behavior and the detailed method is reported elsewhere\(^{5)\}.

The equilibrium carbon concentration at the bubble surface was calculated by the following equation.

\[ [wt\%C]_b = \frac{P_{CO}}{K \cdot f_C \cdot f_O \cdot [wt\%O]_b} \] (12)

where, \([wt\%O]_b\) is the oxygen concentration at the bubble surface and is assumed to be the same concentration of oxygen near the bubble. \( P_{CO} \) is the partial pressure of CO gas in the bubble and was calculated using the following equation.

\[ P_{CO} = \frac{n_{CO}^b}{n_{total \ gas}^b \cdot P_{bubble}} \] (13)

where, \( n_{CO}^b \) is the number of moles of Ar and \( n_{total \ gas}^b \) is the number of moles of CO gas in the bubble. \( n_{CO}^b \) is calculated by sum-
The reaction quantity with the rising bubble to the position being calculated. \( P_{\text{bubble}} \) is the pressure of the bubble and is represented by its relationship with bubble size using the following equation.

\[
P_{\text{bubble}} = P_{\text{no}} \times \left( \frac{R_b}{R_{b,0}} \right)^{-3}
\]

In Eq. (14), \( P_{\text{no}} \) is the hydrostatic pressure at the nozzle position (Pa), \( R_{b,0} \) is the initial bubble radius at the nozzle exit and assumed to be 0.006 m. The initial bubble size may be dependent on gas flow rate. Also, the initial bubble size affects the buoyancy force of gas phase. In this study, the initial bubble size was estimated from comparing the calculated circulation flow rate with experimental result assuming constant for various conditions including gas flow rate. \( R_b \) is the calculated bubble radius at each position (m).

In this study, because of two reasons, \( P_{\text{bubble}} \) is calculated without considering CO gas as shown in Eq. (14). One reason is to prevent increasing calculation time. If the CO gas is considered for calculating the total pressure change of bubble, the fluid flow should be calculated again. If the fluid flow change, the decarburization reaction should be calculated again. Therefore, the calculation should be iterated for several times. The other reason is that the partial pressure of CO is not so large.

Figure 2 shows the CO partial pressure at the initial stage. As process time passes, the CO partial pressure decreases. Therefore, the error from the ignorance of the pressure and volume increase of bubbles by CO may be small.

The fraction of CO gas increases with increasing bubble height. Figures 2(a) and 2(b) show an example of the calculated CO gas pressure and the fraction of a bubble with the height from nozzle. The conditions are represented in Table 1.

As shown in Fig. 2(a), the bubble pressure decreases from the nozzle to the melt surface. However the CO partial pressure increases initially and then decreases. The reason for this is that the portion of CO gas in a bubble increases as the decarburization reaction proceeds but the bubble pressure decreases to the vacuum vessel pressure as the bubble rises.

CO gas fractions in the bubble increases as bubble ascends as shown in Fig. 2(b). The fraction reaches one-forth of total volume near the surface when 3 min passed after circulation starts. The CO gas fractions decrease as time passes. This CO fraction is much smaller than the value reported by M. Takahashi et al.\(^5\) In their report, the fraction is more than half near the surface at 5 min later after the process started. This difference may come from the difference of size expansion during the ascent of bubble.

The contribution of CO gas to the size of bubble was ignored in the present study, because several times of fluid flow calculation at each time should necessary to consider this effect.

In order to calculate the decarburization rate in the bubble surface using Eq. (11), the mass transport coefficient of carbon \((h)\) near the bubble should be known. In this study, the property was evaluated from Higbie’s penetration model.\(^6\)

In the Higbie’s model, the mass transport coefficient for a rising bubble in a liquid can be expressed using the following equation.
where, $D_C$ is the diffusion coefficient of dissolved carbon in the melt ($3.0 \times 10^{-4} \text{ m}^2/\text{s}$) and $t_e$ is the contact time (s). In the case of a rising bubble in a liquid, the contact time between the bubble and a fluid, $t_e$ (s) is estimated using the following equation.

\[ t_e = \frac{d_{\text{slip}}}{u_{\text{slip}}} \]  

(16)

where $d_{\text{slip}}$ is the bubble diameter (m) and relative velocity of bubble (m/s), respectively. If the bubble diameter is greater than 0.002 m, the rising velocity of bubble is not a function of bubble diameter due to the shape deformation from sphere to disk. In considering the bubble expansion, there may be difference of surface area between disk and sphere. However, in this model, the bubble shape was assumed to be sphere for simplicity but the rising velocity of bubble $u_{\text{slip}}$ was estimated from the following equation in the Levich’s discussion considering shape change of bubble.

\[ u_{\text{slip}} = \left( \frac{4 \sigma^2 g (\rho_m - \rho_{\text{Ar}})}{\alpha \rho_m \mu} \right)^{1/5} \]  

(17)

where $\sigma$ is the surface tension (1.8 N/m), $\mu$ is the viscosity of melt (0.0057 kg m$^{-1}$ s$^{-1}$), $\rho_m$ and $\rho_{\text{Ar}}$ are the density of melt and Ar gas (kg/m$^3$) and $\alpha$ is the constant (=30).

Figure 3 represents the mass transport coefficient of dissolved carbon in the melt calculated from Eq. (15) at the same conditions as Fig. 2.

In the Fig. 3, the mass transport coefficient decreases as the bubble rises because the contact time increases due to bubble expansion.

### 3.4.3 Inner Site

The decarburization reaction in the melt can occur near the vacuum vessel surface when the equilibrium CO pressure exceeds the hydrostatic pressure in the melt. However, at the inner site, there exist no initial reaction site such as the bath surface and the Ar bubble. Instead, the reaction occurs through CO bubble formation. This means the reaction at these sites needs additional driving force to overcome the surface tension effect between the melt and the CO bubble. Considering this effect, it can be assumed that a reaction can occur and a CO bubble can nucleate when the equilibrium CO pressure is greater than the sum of the hydrostatic pressure and a critical pressure, as shown in Eq. (17).

\[ P_{\text{CO}}^\text{eq} \geq P_{\text{hydrostatic}} + P_{\text{critical}} \]  

(18)

where, $P_{\text{CO}}^\text{eq}$ is the equilibrium CO partial pressure determined by the dissolved carbon and oxygen concentration (Pa), $P_{\text{hydrostatic}}$ is the hydrostatic pressure of the melt and $P_{\text{critical}}$ is the critical pressure difference for the stable nucleation of a CO bubble (Pa). In this study, the critical pressure difference is assumed to be 0.1 atm.

The reaction rate of decarburization reaction represented in Eq. (6) can be expressed as following equation.

\[ \frac{dC}{dt} = K_C \cdot \text{[wt%C]} \cdot \text{[wt%O]} \]  

(19)

where, $K_C$ is the reaction rate constant. We assumed that the reaction rate is very high and $K_C$ has a large value, 1,000 (wt% s)$^{-1}$. The value represents that the reaction rate is very fast and reaches equilibrium if the conditions of Eq. (18) are satisfied. This value has no quantitative meanings. If 1,000 or 10,000 is used for the reaction constant, the results are very similar with each other.

### 3.5 Oxygen Concentration

The oxygen concentration has an effect on the decarburization reaction. If the oxygen content is high, the decarburization reaction rate is high. In order to calculate the decarburization reaction accurately, the variation of the oxygen concentration should be calculated. In this study, the oxygen concentration was calculated transiently with the carbon concentration. The reaction rate of oxygen at each site was calculated by multiplying the mass ratio between the oxygen and carbon to carbon reaction rate.

### 4. Result and Discussion

#### 4.1 Calculation Procedure

The calculation procedure is shown schematically in Fig. 4. In the first step, the buoyancy force for melt convection was calculated from a plume calculation considering the various conditions. Subsequently, the fluid flow of the steady state was calculated by inserting the buoyancy force. The details of step 1, 2 are explained elsewhere.

In the decarburization calculation, the carbon and oxygen concentration were calculated transiently using Eq. (4). The reaction rates at each site were inserted into Eq. (4) as the
The change in the vacuum vessel pressure should be considered. In a real RH operation, the vacuum vessel pressure varies with time as shown in Fig. 5. In order to consider the pressure variation in numerical calculation, the fluid flow was calculated at each pressure of the vacuum vessel in the step 2. In addition, the pressure of the vacuum vessel was maintained until the next measured pressure. Figure 6 shows the comparison of the pressure variation between a real RH operation and the numerical calculation.

4.2. Comparison with Operational Data

The decarburization reaction was calculated for the one of the RH process using this numerical model. The dimensions and the calculation results of the RH system are shown in Table 1 and Fig. 7, respectively. The pressure variation of the vacuum vessel for Fig. 7 was shown as case A in Fig. 5. The solid line represents the calculated carbon concentration and dashed line represents the oxygen concentration. The solid squares are the operational data, which are measured in the process. From the calculated results, the carbon concentration decreases with time and shows good agreement with the experimental data. It also can be seen that as carbon concentration decreases, the decarburization reaction rate also decreases. Figure 8 shows another result for comparison. The pressure history is case B in Fig. 5.

4.3. Contributions of Each Reaction Site

Using this numerical model, it is possible to determine the contributions of each reaction site. The contributions
can vary with the operation conditions (vacuum pressure, gas flow rate). However, a similar pattern with the carbon concentration exists.

Figure 9 shows the contributions of each reaction site for case A in Fig. 5. The reaction ratio fluctuates at the initial stage. This means that because the reaction occurs vigorously in each reaction site, the concentration difference is large in the vacuum vessel. Figure 10 shows a vertical concentration profile through up snorkel with process time. In the initial stage, there exists a kind of stiff concentration profile near vacuum vessel surface. This causes the fluctuations at the initial stage shown in Fig. 9.

In overall, the contribution of the inner site dominates at the initial stage. However, as the carbon concentration reduces, the contributions of the Ar bubble and the surface increases and the contribution of the inner site decreases drastically. This result is consistent with previous work. S. Kitamura proposed that the inner site is dominant in the initial stage, and bath surface and Ar bubble surface reaction dominate the decarburization in low carbon region.\(^5\)

This result suggests an increase in the decarburization rate in the RH process. Because the decarburization rate has a tendency to be low as the carbon concentration becomes low, the reaction rates of the surface and the Ar bubble sites are a dominant factor in determining the treatment time of the RH process.

5. Conclusion

In this study, a numerical model for the decarburization reaction in RH process was developed by combining the gas behavior, fluid flow and reaction models. The reaction model is incorporated of the models at each reaction site (bath surface, Ar bubble surface and inner site), which considered the thermodynamics equilibrium and reaction kinetics, were established. The calculated results of the decarburization reaction were in good agreement with the operation data.

The quantitative contributions of each reaction sites are estimated. At the initial stage, the contribution of inner site dominates. On the other hand, the Ar bubble and bath surface become dominant in the low carbon region.

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