Evaluation of Bubble Eye Area to Improve Gas/Liquid Reaction Rates at Bath Surfaces

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In ladle refining operations, a plume eye is the area of the bath surface that is not covered with slag, due to bubbling gas, and this area is the most important in slag–metal reactions. In contrast, in vacuum degassers, most of the surface area is not covered with slag, and in this case, the bubble rising area at the bath surface is important for gas–metal reactions. This area is referred to as the “bubble eye area” in this paper. To evaluate the bubble eye size, and its effect on the surface reaction rate, water model experiments were carried out. The plume eye size was measured by changing the thickness of a polystyrene particle layer, which was added to the surface as a slag layer. The bubble eye size was estimated by extrapolating the plume eye size to the value calculated for a polystyrene layer thickness of zero. The volumetric mass transfer coefficient of the surface reaction was measured at various size of plume eye. The volumetric mass transfer coefficient at the bubble eye area was evaluated by extrapolating the mass transfer coefficient at the plume eye area to the value calculated for a plume eye size equal to the bubble eye size. It was concluded that the size of the bubble eye is influenced by the gas flow rate through each nozzle, whereas the volumetric mass transfer coefficient at the bubble eye area is influenced by the bubble eye area and the bubble rising velocity.

KEY WORDS: water model experiment; secondary steelmaking; bubble eye; plume eye; reaction rate at bath surface.

1. Introduction

Extensive research has been carried out with the aim of improving the refining efficiency of vacuum decarburization. For example, Sumida et al.1) showed that the circulation rate in the high carbon-content region and the reaction rate in the low carbon-content region are rate-controlling steps. The decarburization reaction can be written as:

\[ [C] + [O] \rightarrow CO(g) \] (1)

The reaction mechanisms can be divided into the following three types: surface reactions, bubble reactions, and inner reactions. More specifically, decarburization will occur at a specific depth below the bath surface when the hydrostatic pressure at this depth becomes smaller than the equilibrium partial pressure of carbon monoxide. This decarburization reaction, also known as the “inner reaction”, is caused by a rapid decrease in pressure.2) In contrast, the decarburization reaction shown in Eq. (1) proceeds at sites of relatively low pressure, i.e. at the argon bubble surface and at the free surface of the bath. The former reaction is known as the “bubble reaction,” and the latter as the “surface reaction”. Kitamura et al. demonstrated that the surface reaction is more pronounced in the period of low carbon-content.3)

In an attempt to measure the reaction surface rate, water model experiments have been developed; these experiments monitor the changes in concentration of oxygen or carbon dioxide dissolved in the water.4–9) In these experiments, either oxygen or carbon dioxide is removed not only from the free surface (surface reaction) but also from the surface of the rising argon bubbles (bubble reaction). The observed reaction rate is the sum of these two reaction rates. If the concentration changes in the water are measured under different surface atmospheres, the reaction rate at the free surface can be obtained by canceling the bubble reaction rate.

Kitamura et al.4–9) have investigated the ruling factor that uniquely characterizes the surface reaction and have stressed the importance of the plume eye area. In a previous
study, Maruoka et al. used a water model experiment to quantify the surface reaction rate to estimate the influence of bottom bubbling conditions on the gas–liquid reaction rate.\(^7,8\) It was concluded that the plume eye area, as well as the rising velocity of the liquid, influence the surface reaction. Empirical equations were proposed for estimating the reaction rate at the free surface, in which the reaction rate at the plume eye area was evaluated separately from those at the other surface areas for various nozzle configurations.\(^8\)

The term “plume eye area” generally describes the surface that is not covered by slag due to the bubbling gas. More specifically, it is formed by the energy balance of the hydrostatic pressure of the slag and the kinetic energy of the bubbling gas. It therefore changes with the thickness of the slag layer. Furthermore, the plume eye area has an important role in slag–metal reactions, for example slag entrainment and gas–metal reactions. On the other hand, at a free surface with no slag, the bubble rising area is larger than that when slag is present. This area, formed by bubbling gas, is referred to as the “bubble eye area” in this paper. This area is formed only by the kinetic energy of the bubbling gas. For the vacuum degassing reactions in RH\(^9\) or REDA,\(^10\) this bubble eye area has a more important role than that of the plume eye area, because there is no slag in the vacuum vessels in these processes. In a previous study,\(^11\) the influences of bath height and gas flow rate on the reaction rate in the bubble eye area were investigated for a single nozzle configuration. In the present study, the bubble eye areas formed by multiple nozzles are investigated and their effects on the surface reaction are discussed.

2. Experimental Method

Figure 1 shows a schematic diagram of the experimental apparatus used for the water model experiment. The change in the oxygen content of the water was measured to investigate the reaction rate at the free surface of the bath. The experimental procedure has already been described in previous papers.\(^7,8,11\) The experimental apparatus comprised a transparent acrylic cylindrical vessel of height 0.6 m and inner diameter (\(r\)) 0.194 m; an outer vessel for preventing refraction; upper and bottom flanges as the gas outlet and inlet, respectively; a dissolved oxygen meter to measure the changes in oxygen concentration in the water; and a digital camera to measure the plume eye area. In this study, the height of the water (\(H\)) was 200, 300, and 400 mm, and the argon bubbling gas flow rate (\(Q\)) was 1.67, 5.0, and 10.0×10\(^{-3}\) N m\(^{-2}\) s. The three different nozzle configurations shown in Fig. 1 were employed.

Polystyrene particles were used to cover the outer area of the plume eye and the deoxidation rate at the plume eye was measured. Although a viscous oil has often been used to cover the surface in water models, emulsified oil particles in the water adhere to the oxygen sensor and interfere with the measurements under our conditions. In this experiment, therefore, polystyrene particles were used. These particles can prevent reaction at the outer area of the plume eye.\(^8\) The thickness of the polystyrene (\(h_{\text{pol}}\)) was set at 2.5, 4.3, and 6.5 mm.

The movement of rising bubbles was recorded using a high-speed camera (Casio EX-F1, Casio, Tokyo, Japan) at 1200 fps, and the bubble rising velocity was calculated from these images. When the surface was covered by polystyrene, a plume eye was formed by the rising bubbles and its area was recorded using a digital camera (Nikon D70, Nikon, Tokyo, Japan) and calculated using an image analysis program (Image-Pro Plus, Media Cybernetics, Bethesda, MD, USA).

In order to investigate the surface reaction rate, changes in the dissolved oxygen content of the water were measured using a dissolved oxygen meter (TOADKK DO-24P, DKK-TOA Corporation, Tokyo, Japan). The position of this meter was set at the center height of the bath, which was determined from the results of a previous analysis,\(^7\) in which the reaction rate was analyzed by dividing into the surface reaction zone and the mixing zone. It was concluded that the influence of bath mixing is negligible under the present experimental condition unless the dissolved oxygen meter was placed at the bottom side of the vessel, i.e., dead space.

Oxygen was removed not only from the bath surface but also from the rising argon bubbles. If the reaction is assumed to be due to mass transfer of oxygen in the water, the surface reaction rate and the bubble reaction rate can be expressed by Eqs. (2) and (3), respectively:

\[
\frac{d[O]}{dt}\bigg|_S = V (A k_L)_S ([O] - [O]_E^*)
\]

\[
\frac{d[O]}{dt}\bigg|_B = V (A k_L)_B ([O] - [O]_E^*)
\]

where \(t\) denotes time (s); \([O]\) is the content of oxygen in the water (mg/L); \(V\) is the volume of water (m\(^3\)); \(A\) is the reaction area (m\(^2\)); and \(k_L\) is the mass transfer coefficient between the gas and the water (m/s). The superscript * indicates the equilibrium value at the interface, and the subscripts S and B indicate the surface reaction and the bubble reaction, respectively.

The observed reaction rate is the sum of these two reactions. A variation in the oxygen concentration in the atmosphere leads to a change in the equilibrium value at the surface \([O]_E^*\); as a result, the surface reaction rate changes. However, the bubble reaction rate is not influenced by changes in the surface atmosphere (the air or argon atmosphere). Hence, in this method, two sets of experiments were carried out by varying the surface atmosphere under the same bottom bubbling condition. By subtracting the reaction rate of one experiment from that of the other experi-

\[\text{Fig. 1. Schematic diagram of the experimental apparatus and nozzle configuration of bottom flange; } r \text{ represents the vessel radius.}\]
ment, the bubble reaction is canceled out and the surface reaction rate can be evaluated. The volumetric mass transfer coefficient at the surface, \( (Ak_L)_S \) (m³/s), was used for the evaluation and is expressed as

\[
(Ak_L)_S = \frac{(d(O)/dt)_S - (d(O)/dt)_S^*}{[O]_S^* - [O]_S^*} \times V \quad \cdots (4)
\]

where the superscripts “air” and “Ar” indicate the air and argon atmospheres, respectively.

3. Experimental Results

3.1. Evaluation of Bubble Eye Area

Figure 2 shows photographs of the plume eye area at various bath heights and polystyrene layer thicknesses. The size of the plume eye decreased with increasing thickness of the polystyrene layer covering the bath surface. The number of plume eyes was the same as the number of nozzles. In the case of two and three nozzles, no coalescence of the plume eyes formed by the bubbling gas injected through each nozzle was observed.

Figure 3 shows the dependence of plume eye size on gas flow rate, bath height, polystyrene layer thickness, and nozzle number. The size of the plume eye increases with gas flow rate under all conditions. The influence of bath height on the size of the plume eye was not very significant under our conditions, although it was expected that the size of the plume eye would generally increase with bath height. At almost all conditions of gas flow rate, thickness of polystyrene layer, and nozzle number, a bath height of 0.3 m produced the largest plume eye area. This is thought to be because the swirling motion of the plume is enhanced by increasing bath height, and at a bath height of over 0.3 m, the plume eye rotates widely and the gas stirring energy injected from bottom is consumed in rotation of the bath. Figure 3 also shows that the size of the plume eye decreases with increasing polystyrene layer thickness. If the cross section of the plume eye is assumed to be as shown in Fig. 4, the relationship between the radius of the plume eye and the thickness of the polystyrene layer can be described by the following equation:

\[
r_p = r_b - \alpha \times h_{\text{poly}}, \quad \alpha = \tan \theta^{-1} \quad \cdots (5)
\]

where \( r_p \) represents the radius of the plume eye area (m), \( h_{\text{poly}} \) is the thickness of the polystyrene layer (m), and \( \theta \) is the angle of the plume eye. In this equation, \( r_b \) is the plume eye radius extrapolated to \( h_{\text{poly}} = 0 \), and this value corresponds to the radius of the bubble eye area. By using the value of \( r_b \), the height of the bubble eye area, \( h_b \), can be calculated as follows:

\[
h_b = \frac{r_b}{\alpha} \quad \cdots (6)
\]

Figure 5 shows a typical experimental result. Such diagrams can be used to obtain the \( \alpha \) and \( r_b \) values for each set of experimental conditions. In the case of a multi-nozzle configuration, the observed plume eye area was divided by the number of nozzles, \( n \), and the bubble eye size for the bubbling gas through each nozzle was calculated. Figures 6, 7, and 8 show the dependences of the radius, \( r_b \), height, \( h_b \), and volume, \( V_b \), of each bubble eye on the gas flow rate through each nozzle, \( Q/n \); \( V_b \) is calculated using the following equation:

\[
V_b = \frac{\pi r_b^2 h_b}{3} \quad \cdots (7)
\]

A fairly good relationship between the bubble eye radius

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\[
h_b = \frac{r_b}{\alpha} \quad \cdots (6)
\]
and the gas flow rate from each nozzle can be observed in Fig. 6. This relationship leads to the following empirical equation, which can be used to estimate each bubble eye area, $A_b/n$.

$$A_b/n = 7.63 \left( \frac{Q}{n} \right)^{2/3}$$

In contrast, rather poor relationships are observed in Figs. 7 and 8. It can be assumed that the errors in the calculations of $b_b$ or of $V_b$ are significantly larger than that of $r_b$ because the experimental errors of the $r_b$ and $\alpha$ measurements are multiplied in the calculations. This is probably one of the reasons why rather poor relationships are observed in these two figures.

### 3.2. Evaluation of Bubble Rising Velocity

Figure 9 shows the relationship between the gas flow rate, $Q$, and the bubble rising velocity, $U_B$. $U_B$ increases with $Q$ for all bath heights. However, $U_B$ decreases with increasing nozzle number. This is because the gas flow rate from each nozzle decreases to $Q/n$ because of division of the gas by the multiple nozzles, although the total gas flow rate is constant. However, the influence of the bath height on the bubble rising velocity was not very significant. In general, $U_B$ was observed to increase in proportion to the gas flow rate to the power $0.2$.\textsuperscript{16–17} Iguchi et al. reported the following equation for the bubble rising velocity with a single nozzle\textsuperscript{17}:

$$U_B = k(Q/n)^{0.1}$$

where $k$ is a scaling factor. In this study, multiple nozzle configurations were used. The gas flow rate from each nozzle can be represented by $Q/n$, and the following relationship, shown in Fig. 10, is obtained:
Fig. 11. Correlation between the volumetric mass transfer coefficient and the gas flow rate at various nozzle configurations, bath heights (H), and polystyrene layer thicknesses (h_{pol}).

\[ U_B = 5.86 \times ((Q/n)^2/g)^{0.1} \] ........................(10)

where \( g \) is the acceleration due to gravity (m/s²). This equation indicates that the rising velocity has an insignificant effect on the bath height.

3.3. Surface Reaction Rate Evaluation

Figure 11 shows the relationship between the volumetric mass transfer coefficient at the bath surface, \( (Ak_L)_S \), and the gas flow rate for various nozzle configurations, bath heights, and polystyrene layer thicknesses. When a polystyrene layer exists, \( (Ak_L)_S \) indicates the volumetric mass transfer coefficient at the plume eye area, \( (Ak_L)_P \). \( (Ak_L)_S \) increases with increasing gas flow rate, and decreasing nozzle number, bath height, and polystyrene layer thickness.

4. Discussion

The plume eye plays an important role when slag is present, but the bubble eye area is more important under conditions when no slag is present, such as in RH⁹ or REDA¹⁰ processes. Previous studies indicated that the product of the plume eye size and the plume rising velocity depended strongly on the volumetric mass transfer coefficient. The mass transfer coefficient at the bubble eye was evaluated, based on this result. Under the same nozzle, bath height, and gas flow rate conditions, the volumetric mass transfer coefficient at the plume eye area is affected by only the plume eye area, which can be changed by changing the thickness of the polystyrene layer, because the plume rising velocity is constant unless the nozzle number, bath height, or gas flow rate change. Figure 12 shows the typical relationship between the size of the plume eye and \( (Ak_L)_P \), where the bath height is 0.2 m and the total gas flow rate is \( 1.0 \times 10^{-4} \text{ N m}^3/\text{s} \). A linear relationship was observed, and the volumetric mass transfer coefficient at the bubble eye area \( (Ak_L)_b \) can be calculated by extrapolating this relationship to the size of the bubble eye, which is obtained under the same gas bubbling conditions. Figure 13 shows the calculations of \( (Ak_L)_b \) under various conditions. In this figure, \( (Ak_L)_t \) represents the volumetric mass transfer coefficient at the total surface measured without the use of polystyrene, i.e. \( (Ak_L)_t \) at \( h=0 \) (mm), and \( (Ak_L)_o \) represents the volumetric mass transfer coefficient at the surface, except for the bubble eye area, calculated by the following equation:

\[ (Ak_L)_t = (Ak_L)_P - (Ak_L)_b \] ..........................(11)

Under all conditions, \( (Ak_L)_t \) and \( (Ak_L)_b \) increase with the gas flow rate. In contrast, \( (Ak_L)_t \) decreases with increasing gas flow rate under some conditions. At \( H=0.2 \text{ m} \), \( (Ak_L)_t \) does not change much for different nozzle numbers; with increasing bath height, the effect of the nozzle number on \( (Ak_L)_t \) becomes significant. On the other hand, \( (Ak_L)_b \) does not change much. From these results, it is clear that increasing the bath height and the nozzle number suppresses \( (Ak_L)_b \). It is assumed that the plume eye area is too large and that it is difficult for the water close to the surface area.
to move freely, thus water has to circulate below the bath surface. As a result, the mass transfer coefficient at the outer area of the plume eye decreases with increasing bath height. To confirm this hypothesis, velocity measurements such as Particle Image Velocimetry (PIV) will be necessary.

It is believed that the volumetric mass transfer coefficient at the bubble eye area is influenced by not only the area of the bubble eye but also the bubble rising velocity as an energy supplier for rising water in bubble eye. Figure 14 shows the relationship between \( (Ak)_b \) and \( A_b U_B \). A linear relationship was observed. By regression analysis, the following empirical equation for the volumetric mass transfer coefficient at the bubble eye area was obtained:

\[
(Ak)_b = 1.92 \times 10^{-4} A_b U_B + 2.30 \times 10^{-7} \quad \ldots (12)
\]

where the intercept term indicates that the reaction occurs when the gas flow rate is zero.

In this equation, \( A_b \) and \( U_B \) can be calculated using Eqs. (8) and (10). By substituting Eqs. (8) and (10) into Eq. (12), the volumetric mass transfer coefficient at the bubble eye area can be expressed using only operating parameters:

\[
(Ak)_b = 8.64 \times 10^{-3} \frac{g^{0.87} \theta^{0.13}}{\theta^2} + 2.30 \times 10^{-7} \quad \ldots (13)
\]

5. Conclusions

To evaluate bubble eye size and its effect on the surface reaction rate, water model experiments were carried out. The size of the plume eye was measured by changing the thickness of the polystyrene layer added to the surface. The size of the bubble eye area was calculated by extrapolating the size of the plume eye to its value at a polystyrene layer thickness of zero. The volumetric mass transfer coefficient of the surface reaction was measured as the size of the plume eye changed with varying polystyrene layer thickness. The volumetric mass transfer coefficient at the bubble eye area was then evaluated by extrapolating the mass transfer coefficient at the plume eye to the value at which the size of the plume eye area was equal to the size of the bubble eye area. The following conclusions were drawn.

1. The size of the bubble eye is influenced by the bubbling gas flow rate through each nozzle.
2. A comparison of the volumetric mass transfer coefficient at the bubble eye area \( (Ak)_b \) and that at the remaining surface area \( (Ak)_s \), revealed that \( (Ak)_b \) becomes larger than \( (Ak)_s \) as the gas flow rate increases.
3. The dependences of the volumetric mass transfer coefficient at the total surface area \( (Ak) \), and that at the bubble eye area \( (Ak)_b \), on bubbling gas flow rate are similar. \( (Ak)_b \) was influenced not only by the size of the bubble eye but also by the bubble rising velocity.
4. Empirical equations for the size of the bubble eye, the bubble rising velocity, and \( (Ak)_b \) were obtained.

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