The Effect of the Gas Jet on the Gas/Liquid Reactivity in the Metallurgical Vessel

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The effect of the turbulent gas jet on the reaction characteristics in gas blowing was investigated by the simultaneous measurements of gas/liquid interfacial area and gas holdup. Gas/liquid interfacial area was measured by KOH/CO₂ model system on the fast pseudo first-order reaction, and gas holdup was measured by using the electrical conductivity method. By estimating the mean diameter of bubbles from the measured data of gas/liquid interfacial area and gas holdup, in the gas/liquid two phase region, the jet effect on the improvement of the reactivity was investigated.

Both the specific interfacial area and the gas holdup were increased as the aspect ratio decreased. The variations were larger in a high gas flow rate and in a small nozzle diameter. The effect of nozzle diameter was larger in a low aspect ratio. The calculated mean diameter of bubbles was slightly increased with the increase of the nozzle diameter and the aspect ratio. The result shows that the increase in the reacting interfacial area according to the development of jet was mainly affected by the increase of gas holdup through being dense of bubble swarms in the jet.

KEY WORDS: turbulent buoyant jet; mean diameter of bubbles; gas holdup; gas/liquid interfacial area; electrical conductivity method; KOH/CO₂.

1. Introduction

There are many cases in metallurgical processes where the two phase gas jet is developed by the gas blowing such as the refining of steels.¹⁾ The gas blowing not only has effects on the degassing and the mixing but also improves the reactivity by increasing the gas/liquid interfacial area. In the aspect of the improvement of reactivity through the increase of the reacting interfacial area, the vertical column reactor in chemical engineering fields is much more advantageous.

The column reactor has a large aspect ratio (H/D), generally greater than 10, for the purpose of the increase in the total gas/liquid interfacial area, and the gas/liquid reaction occurs in the whole region of the reactor. However, the reaction in the vessels like in metallurgical processes with small aspect ratios, commonly less than 1, occurs only in the gas/liquid two phase region, the reacting interfacial area in a metallurgical vessel is much less than that of a column reactor.

When a fluid of density ρ_0 is passed with a velocity u_0 through a vertical bottom nozzle of diameter d into a environmental fluid of density ρ_E ($\rho_0 < \rho_E$), the modified Froude number Fr' is defined as a ratio of inertial force to buoyancy force and given as follows.²⁾

$$Fr' = \frac{u_0^2}{gd(\rho_E - \rho_0)/\rho_0} \dots\dots\dots(1)$$

The fluid passed the nozzle can be developed as a jet

owing to the high speed of the injection but then it entrains the environmental fluid with a turbulent mixing and undergoes a gradual transition from a jet to a plume as consuming its inertial force.³⁾ A pure jet is defined as a state that the inertial force is dominant over the buoyancy force ($Fr' = \infty$), and a pure plume state is defined to the contrary ($Fr' = 0$). The transition region is called a forced plume or a buoyant jet.

As a column reactor has a small Froude number with which difficulties arise in developing the gas jet, the reactivity is mainly determined by the gas holdup.^{4,5)} In a column reactor, the gas holdup is governed by the superficial gas velocity (Q_g/D) and the physical properties between gas and liquid phases because the portion of the jet in the whole swarms of bubbles is negligible, as shown in many research works.^{6,7)} Therefore, the results in a column reactor could not be applied to the metallurgical vessel where the gas jet is very much developed with high gas flow rates. In a reactor of a small aspect ratio, the injected gas is then developed as a jet to increase the reacting interfacial area, as a result, in a metallurgical vessel of a small aspect ratio, it is very important to comprehend the bubble structure by the development of the jet in order to study the reaction characteristics.

In a gas/liquid two phase jet phenomena, it is difficult to develop a pure jet because of a large density difference between the fluids.⁸⁾ Even if a pure jet is produced, the injected gas spends its inertial force and goes into the transition region immediately. Therefore, it is reasonable to regard the gas/liquid two

Fig. 1. Physical models of the jet structure.

phase jet as a buoyant jet.⁶⁾ It needs to know the structure of the buoyant jet in order to examine how the buoyant jet affects the reactivity, but it is not clearly understood yet.

The structure of the buoyant jet can be deduced from the relationship among the interfacial parameters such as gas/liquid interfacial area a , gas holdup α , and mean diameter of bubbles d_M , assuming spherical bubbles,⁹⁾ which is

$$a = \frac{6\alpha}{d_M} \quad \dots\dots\dots(2)$$

From the above equation, the increase in the specific interfacial area due to a buoyant jet might be the results of an increase of α or a decrease of d_M , or their combined effect, as shown in Fig. 1.

In this study, the specific interfacial area and the gas holdup were measured independently, and the mean diameter of bubbles is estimated from Eq. (2) with the measured data to investigate the structure of the two phase jet.

2. Experimental Procedure

The schematic diagram of the experimental apparatus is shown in Fig. 2. A cylindrical reactor of inner diameter 18 cm was made of transparent acrylic resin. Some cylindrical tubes were piled up vertically in order to allow the variation of the aspect ratio from 1.5–6.55, and the gas was injected from the bottom nozzle located at the center with the flow rates of 5–120 l/min.

In case of measuring the reacting interfacial area, the gas mixture of CO₂ and air was injected and reacted with KOH in the solution. The variation of pH was continuously recorded in the chart recorder. For the measurement of gas holdup, only the compressed air was injected from the bottom nozzle. The surface elevation due to the air blowing was measured by the use of an electrical conductivity probe. The measured data of the continuous variation of the electrical conductivities were converted into the digital ones via A/D (Analog/Digital) converter so that they were analyzed statistically in a personal computer.

2.1. Measurement of Gas/Liquid Interfacial Area

The popular reaction used for the reactor design of the gas/liquid reaction system is the fast pseudo first-order reaction and KOH/CO₂ model system is widely used for that reaction because of its easiness of treatment and analysis.^{4,10–13)} The determination of the model system and the choice of experimental condi-

Fig. 2. Schematic diagram of experimental apparatus for measuring the specific interfacial area and the gas holdup.

tions have been investigated rigorously in the research work of Yoon *et al.*¹⁴⁾

The reaction regime for the fast pseudo first-order reaction can be classified by some dimensionless variables⁹⁾ which influence the rate of gas absorption, such as Hatta number Ha and enhancement factor E_i .¹⁵⁾

$$3 < Ha < \frac{E_i}{2} \quad \dots\dots\dots(3)$$

$$\text{where, } Ha = \frac{(D_A k_2 C_{B0})^{1/2}}{k_L}$$

$$E_i = 1 + \frac{D_B}{z D_A} \cdot \frac{C_{B0}}{C_A^*}$$

The rate expression for the fast pseudo first-order reaction is^{9,16)}

$$\frac{P_A}{\Phi} = \frac{1}{a} \cdot \left(\frac{1}{k_G} + \frac{H_e}{(D_A k_2 C_{B0})^{1/2}} \right) \quad \dots\dots\dots(4)$$

The mass transfer coefficient for the liquid phase k_L is not involved in the rate expression in Eq. (4), so it is not necessary to know the exact value of k_L in calculating the gas/liquid specific interfacial area a in this case. Generally the value of k_L is not changed very much according to the conditions and is in

the order of magnitude of 10^{-2} cm/s,^{11,13)} hence this value is used in the determination of the experimental conditions.

From the data of a rate constant k_2 ,¹⁷⁾ a gas solubility $1/He(=C_A^*/P_A)$,¹⁵⁾ diffusivities of the reactants D_{CO_2} , D_{OH^-} ,¹⁷⁾ it can be seen that the fast pseudo first-order reaction occurs in the condition of

$$0.05 < C_{OH^-} < 0.1 \quad \dots\dots\dots(5)$$

From these considerations, the experimental conditions are determined as follows.

- Reaction temperature: 20°C
- Initial concentration of KOH solution: 0.1 mol/l
- Partial pressure of CO₂: 0.05 atm

The rate of CO₂ absorption ϕ is expressed as follows.

$$\phi = -\frac{dC_{CO_2}}{dt} = -\frac{1}{2} \frac{dC_{OH^-}}{dt} \quad \dots\dots\dots(6)$$

$$C_{OH^-} = k_w/10^{-pH}$$

Therefore, the specific interfacial area a can be obtained from the slope of the plot of P_A/ϕ vs. $He/(D_A k_2 C_{B0})^{1/2}$ in Eq. (4) after ϕ is obtained by the measurement of pH variation according to the absorption of CO₂.

2.2. Measurement of Gas Holdup

The gas holdup α is a volume fraction of gas phase in the overall aerated liquid. Some techniques to determine the gas holdup have been used in many works.¹⁸⁻²²⁾ In this study, the direct measurement of the height of an aerated liquid by the use of an electrical conductivity probe was conducted. Fig. 3(a) shows the schematic diagram of experimental apparatus for measurement of the gas holdup and Fig. 3(b) a representative signal from the electrical conductivity probe. The electrical conductivity probes were made of a copper wire in a thin stainless steel tube. One of the two probes is submerged deeply and fixed in the KOH solution, the other is movable and located at certain selected points near the surface level according to the elevation of the surface due to the aeration.

The surface of the aerated liquid is elevated and fluctuated by the rising bubbles according to the gas blowing. Then the probe tip near the elevated surface can be in contact with the surface and construct the electrical circuit, so that the variation of the electrical conductivity is measured in the out channel. In case that the probe tip is apart from the liquid surface, the circuit is open and no variation is found in the channel. A sample signal of the electrical conductivity is shown in Fig. 3(b).

For certain selected vertical positions of the probe tip including the position over the elevated surface and that under the surface, the variation of the electrical conductivity is obtained and the contact portion (integrated time of contact over the total measurement time) is calculated. The portions of contact time at each height of the probe tip are interpolated to get the height at the fraction of 0.5. This is considered as the height of aerated liquid (H_a). The

Fig. 3. Experimental apparatus and method for measurement of the gas holdup (α).

overall gas holdup is calculated from the total surface elevation and the height of the clear liquid without aeration (H) ($\alpha = (H_a - H)/H_a$).⁹⁾ The accuracy of the measured value of the total surface elevation is reported as ± 0.2 mm.²²⁾

This electrical conductivity probe method for measuring the gas holdup is very simple but gives much accurate results. In addition, it is effective and suitable for this study such as to investigate the structure of the bubble swarms by comparing the results of the gas holdup with those of the specific interfacial area.

3. Results and Discussion

The model system and the experimental conditions were carefully selected to simulate the pseudo first-order reaction to determine the specific interfacial area. As using the dilute CO₂ gas, it could be possible to obtain the stable experimental results up to the high gas flow rate, that could produce the high speed jet such as in the real steelmaking processes. Variations of the gas/liquid interfacial area per unit volume of the reactor (specific interfacial area, a) according to the changes of the gas flow rate and the nozzle diameter are shown in Figs. 4 and 5 with different aspect ratios.

The specific interfacial area was increased with the increased flow rate and the decreased nozzle diameter. The variations were larger in a high flow rate than

Fig. 4. Comparison of the variation of the specific interfacial area, the gas holdup and the mean diameter of bubbles with respect to the gas flow rate. ($H/D = 2.00$)

Fig. 5. Comparison of the variation of the specific interfacial area, the gas holdup and the mean diameter of bubbles with respect to the gas flow rate. ($H/D = 6.55$)

in a low one. This effect of nozzle diameter was different from the results of Inada and Watanabe,²³⁾ and Fruehan and Martonik,²⁴⁾ where the changes of the nozzle diameter reported to have no effect on the $k_L \cdot a$. This means that the specific interfacial area a is not changed with the nozzle diameter, since k_L is not changed very much as mentioned before. This discrepancy might be attributed to their inadequate choices of experimental conditions such as the use of pure CO_2 and too low flow rates to develop the gas jet. The results of the gas holdup α had the same tendency as those of the specific interfacial area a , in

which the gas holdup was increased as the flow rate increased and as the nozzle diameter decreased.

The effect of nozzle diameter was larger in a low aspect ratio in both cases of the gas holdup and the specific interfacial area. The reason for the large effect of nozzle diameter in a low aspect ratio is considered as the increased volume fraction of the gas jet over the total bubble swarms. The results show that the development of jet has large influence on the reaction efficiency in a reactor of the metallurgical processes of a small aspect ratio.

If the volume of a reactor is identical, it is favorable to improve the reaction efficiency through the increase of the residence time of bubbles and the increase of the gas holdup *via* elevating the aspect ratio, like in a column reactor.^{5,7)} However, in a reactor in metallurgical processes, the development of the buoyant jet gives rise to a problem of abrasion of refractories due to the strong stirring of the high temperature molten steel, so the vessels in metallurgical processes require low aspect ratios to reduce the abrasion.

With the intention of observing this jet phenomena clearly, the experiments including low flow rates of 5, 10, 20 l/min of a low aspect ratio of 1.5 were conducted. As shown in Figs. 6 and 7, the effect of nozzle diameter was not seen at low gas flow rates even in low aspect ratios. This is because the jet was not developed at such a low flow rate. Figs. 8 and 9 show the decrease of the specific interfacial area with the increase of the aspect ratio. This effect of the nozzle diameter is emerged because the injected gas has been developed as a jet at the nozzle tip according to the increase of the flow rate and the decrease of the nozzle diameter, and the portion of jet was decreased with the increase of the aspect ratio.

As mentioned before, the improvement of the reactivity through the increase of the specific interfacial area can be explained as the development of the buoyant jet. The cause of the increase of the specific interfacial area according to the development of the buoyant jet may be the increase of the gas holdup or the decrease of the mean diameter of bubbles from the Eq. (2). The mean diameter of bubbles can be calculated from the experimental results of the specific interfacial area and the gas holdup. The calculated results (Figs. 4 and 5) show that the gas bubble size was increased with the increase of the gas flow rate, but slightly increased as the nozzle diameter was increased. And the bubble size was increased with the increase of the aspect ratio, as shown in Figs. 10 and 11. But the variations of the bubble size with the nozzle diameter were negligible as the aspect ratio was increased.

Therefore, as comparing the variation of the gas holdup with that of the mean diameter of bubbles, the main cause of the improvement of the reactivity owing to the development of the buoyant jet is considered as being dense of the bubble swarms in the jet, and the effect of the bubble size is relatively small. The reason why the bubbles are the larger in the high aspect ratio is that the rising bubbles out of the jet region may undergo the coalescence and/or the growth.

Fig. 6. Effect of the nozzle diameter on the specific area. ($H/D=1.50$)

Fig. 7. Effect of the nozzle diameter on the gas holdup. ($H/D=1.50$)

The hydrodynamics on the gas/liquid two phase turbulent jet, which has an organized structure superimposed on the chaotic or random motion, should be fully understood if they want to know the cause of denseness of the bubbles due to the jet quantitatively.⁸⁾ However, at present, the theory has not confirmed yet, so only the qualitative inference is possible for the denseness of the bubbles. In the gas/liquid two phase turbulent jet, the bubble plume is developed in an early stage in the case that the initial inertial moment is relatively small compared with the buoyancy force. Therefore, the injected gas must be a compressed fluid with a high nominal Mach number Ma' so that the buoyant jet can be developed.⁸⁾ Hence the compressed gas, passed through the nozzle, experiences the immediate expansion by a pressure drop at the nozzle tip and an accumulation of the momentum flux arises along the jet direction in a

Fig. 8. Variation of the specific area with respect to the aspect ratio. (Nozzle diameter=0.16 cm ϕ)

Fig. 9. Variation of the specific area with respect to the aspect ratio. (Nozzle diameter=0.50 cm ϕ)

buoyant jet region. So the denseness of bubbles takes place in a jet region, whereas the bubble rises with a certain momentum in a plume region. In addition, the coalescence of the bubbles is suppressed by an eddy motion in a turbulent jet.

Consequently, it is thought that the bubbles of a small size are gathered in swarms high-densely in a jet region and have a complicated turbulent motions.

4. Conclusion

The variations of the gas/liquid specific interfacial area a and the gas holdup α was measured in order to investigate the effect of the jet phenomena on the gas/liquid reaction. The conclusions are as follows.

(1) The specific interfacial area was increased as the gas flow rate was increased. The increase of the gas/liquid interfacial area was larger in a small nozzle

Fig. 10. Variation of the mean bubble diameter with respect to the aspect ratio. (Nozzle diameter=0.16 cm ϕ)

Fig. 11. Variation of the mean bubble diameter with respect to the aspect ratio. (Nozzle diameter=0.50 cm ϕ)

diameter than in a large one at the same flow rate, but at low flow rates, it was negligible according to the changes of the nozzle diameter.

(2) The specific interfacial area was increased as the aspect ratio decreased. This is considered as the volume fraction of the jet over the total swarms of bubbles was increased.

(3) The influences of the gas flow rate, the nozzle diameter, and the aspect ratio on the gas holdup had the same tendency as the case in the specific interfacial area.

(4) The mean diameter of bubbles was calculated with the measured data of the specific interfacial area and the gas holdup. The variations of the bubble size with the nozzle diameter were negligible as the aspect ratio was increased. Therefore, the increase of the specific interfacial area due to the development

of the jet was mainly influenced by the increase of the gas holdup through being dense of the bubble swarms in the jet.

Nomenclature

- a : Specific interfacial area per unit volume of the system (l/cm)
- C_A^* : Concentration of gas A at the interface (gmol/cm³)
- C_{B0} : Bulk concentration of reactant B in the solution (gmol/cm³)
- C_{CO_2} : Concentration of CO₂ in the solution (gmol/cm³)
- C_{OH^-} : Concentration of OH⁻ ion in the solution (gmol/cm³)
- D : Vessel diameter (cm)
- D_i : Diffusion coefficient of i species (cm²/s)
- d : Nozzle diameter (cm)
- d_M : Volume surface mean diameter (cm)
- E_i : Enhancement factor corresponding to an instantaneous reaction
- Fr' : Modified Froude number
- g : Gravitational acceleration (cm/s²)
- H : Liquid height (cm)
- H_a : Height of the aerated liquid (cm)
- H/D : Aspect ratio (liquid height/vessel diameter)
- Ha : Hatta number
- He : Henrian coefficient (gmol/cm³atm)
- k_2 : Second-order reaction rate constant (cm³/gmol)
- k_G : Mass transfer coefficient for the gas phase (gmol/s)
- k_L : Mass transfer coefficient for the liquid phase (cm/s)
- k_W : Solubility product of water
- Ma' : Nominal Mach number
- P_A : Partial pressure of the soluble gas A (atm)
- Q_g : Gas flow rate (l/min)
- t : Time (s)
- u_0 : Mean velocity at the nozzle (cm/s)
- z : Stoichiometric coefficient in the chemical reaction
- α : Overall gas holdup in the vessel
- ρ : Fluid density (g/cm³)
- ρ_0 : Density of the injected fluid (g/cm³)
- ρ_E : Reference density (g/cm³)
- ϕ : Rate of absorption of gas (gmol/s)

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