Experiments on Removal of Hydrophilic Fine Particles in Bubbly Flow

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It is commonly believed that the solid oxides, like aluminum inclusions, were easily removed by bubbles because of their poor wettability with molten steel. However the liquid oxides like ladle slags were hard to remove because of their good wettability. The purpose of this study was to reveal the removal rate coefficient of the hydrophilic particles, which simulated liquid oxides in molten steel, in bubbly flows. The water model experiments were carried out. The air was injected into water-particles mixtures in a rectangular tank and the amount of particles included in the mixture overflowed from the tank was measured. The experiments confirmed that bubbles have abilities to bring hydrophilic particles upward even though the particles do not adhere to the bubble interfaces. The hydrophilic particles are entrained into the bubble wake at a certain probability and dragged upward while riding on the liquid flow in the wake. The removal rate coefficient per a bubble increases with the bubble diameter whereas the increase in the number of bubbles deteriorates the efficiency due to the reduction of the wake volume which is effective for particle transportation.

KEY WORDS: hydrophilic inclusion; bubble; bubble wake; removal rate coefficient.

1. Introduction

Improving the purity of molten steel in the continuous casting is of great importance to realize high quality steel products. Therefore, there have been various engineering processes for removing inclusions in the tundish. Gas injection into molten steel, by which gas bubbles are expected to bring inclusions up to a slag layer at free surface, is one of the promising inclusion removal techniques.^{1–6)}

Aluminum inclusions, Al_2O_3 , have been considered to be attached to bubble interfaces due to their good wettability with molten steel. There are many studies on the mechanism and efficiency of removal of hydrophobic particles.^{7–15)} Reay and Ratcliff⁸⁾ theoretically investigated the removal efficiency of fine particles by rising bubbles. Their model showed that the decrease in bubble size increases the removal rate. Yoon and Mao¹²⁾ modeled the removal of fine particles in a liquid pool as a first-order ordinary differential equation, in which the time derivative of the total number of particles in the liquid pool is assumed to be proportional to the total number of particles itself, and therefore, the proportional coefficient is referred to as the particle removal rate coefficient. The removal rate coefficient is given in terms of the bubble surface area and the probability of particle collection consisting of the collision, adhesion and detachment probabilities. In their model, the

collision probability increases with decreasing bubble size and with increasing particle size, which is consistent with Reay's model and other reports.¹¹⁾ Flint and Howarth¹⁶⁾ deduced the efficiency of collision between a bubble and a particle in the Stokes and potential flow regimes. The collision efficiency depends on two dimensionless parameters: the particle settling velocity scaled by the bubble velocity and the particle Reynolds number. They pointed out based on the model that the collision efficiency is governed by the former and increases with decreasing bubble size when the particle size is small, whereas both parameters affect the collision efficiency, which increases with bubble size, when the particle size is large.

The Yoon-Mao model and single-bubble flotation experiments¹²⁾ implied that the hydrophobic force plays a dominant role in particle removal by bubbles. Zheng *et al.*¹³⁾ carried out particle removal experiments and also pointed out the importance of the hydrophobicity of particle in removal efficiency. Pan *et al.*⁹⁾ investigated the effects of particle properties on the removal efficiency. They divided the particle transportation process into the particle transport from bulk liquid to the bubble interface due to adhesion and that in the bulk liquid due to the liquid flow caused by bubbles. Their experiments revealed that, for hydrophobic inclusions like Al_2O_3 , the latter process governs the removal rate since the former proceeds much more rapidly.

Slag inclusions may not attach themselves to bubble interfaces since their wettability with molten steel is poor. In contrast to the studies on removal of hydrophobic particles,

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only a few studies on the effects of bubbles on removal of hydrophilic particles have been carried out. Li *et al.*¹⁷⁾ proposed a numerical method, which is a combination of the volume-of-fluid (VOF) method for tracking gas-liquid interface¹⁸⁾ and the discrete particle model (DPM) for tracking discrete particles,¹⁹⁾ to simulate the bubble-particle interaction. In their two-dimensional simulations of single bubbles, glass particles of 0.8 mm in diameter and 2 500 kg/m³ in density were transported upward by the bubble wake and the wake entrainment process agreed with that observed in an experiment.²⁰⁾ Xu *et al.*²¹⁾ carried out three-dimensional VOF-DPM simulations of single bubbles rising through stagnant liquid including particles. They investigated the effects of bubble diameter and particle diameter on the efficiency of the upward transportation of particles by bubble wake. The numerical predictions showed that the transportation rate of particles per a bubble increases with bubble size. The wake entrainment effect on particle motion was experimentally investigated by Yang *et al.*²²⁾ Single bubbles were continuously injected from the bottom of a liquid pool and the rate of capture of particles by bubble wake was evaluated from video images. The particle motion in the particle-bubble wake interaction was classified into (1) approaching and passing into a wake boundary zone, (2) passing into a rising zone of wake, and (3) going on floating up or escaping from the rising zone. Their study focused on the identification of the wake entrainment process, so that the particle removal rate coefficient was not investigated.

Experiments on the removal rate coefficient in bubbly flows were carried out by Arai.²³⁾ He used various particles having different wettability with the contact angle ranging from 80 to 94 degree. The bubble and particle sizes were about 2 mm and 10 μm , respectively. Particles were dispersed in bubbly flows in a cylindrical column and were overflowed from the column top. He evaluated the removal rate coefficient by measuring the number concentration of particles in the column. The removal rate coefficient for hydrophilic particles was much smaller than that for hydrophobic particles, and therefore, he concluded that the inclusion removal by bubble injection does not efficiently work for hydrophilic slag inclusions. However in the actual bubbling system for molten steel bubbles and slag particles can be several times larger than those in his experiments, and our knowledge on the inclusion removal for hydrophilic particles in bubbly flows is still insufficient.

Particle removal experiments were carried out in this study for hydrophilic particles of 100 μm diameter in bubbly flows consisting of bubbles of about 5 mm in diameter. The removal rate coefficient was evaluated using a measurement method similar to Arai's experiment. The mean bubble diameter was varied from 4 to 6 mm to investigate the effect of bubble size on the removal rate.

2. Experimental

2.1. Experimental Setup and Conditions

Figure 1 shows the experimental setup. The test section was the water tank, whose dimensions were 240 $\times$ 210 $\times$ 72 mm in width, height and depth. Mixture of water and particles was filled in the tank. The water temperature was kept at 25 $\pm$ 1°C throughout the experiments. The air dif-

fuser plate was mounted at the bottom of the tank. Stainless tubes were flash mounted on the diffuser plate as shown in Fig. 2 and they were placed inside the air chamber. Air was supplied from the compressor (HITACHI, OILFREE SCROLL-2.2) and flowed into the water tank through the mass flow controller (KOFLOC, Model3660), the air chamber and the tubes. The tube length was 160 mm. The long tubes caused large pressure drops, resulting in uniform bubbling.^{24,25)}

Spherical particles (Fuji-Silysia Chemical, CARiACT Q-10, porous silica) were used for inclusion. The particles were hydrophilic and were well dispersed in water. Orvalho *et al.*²⁶⁾ investigated the particle properties in detail. The particle size distribution had a dominant peak around 100 μm , and the mean diameter and the standard deviation were 103 μm and 15 μm , respectively. Initially the tank was empty and air was continuously injected at a constant gas volume flow rate, Q_G . Particles of the total mass, M_0 , of 0.30 kg were pre-mixed with water and the water-particle mixture was poured into the tank while keeping the air injection. The mixture supply was stopped when the free surface

Fig. 1. Experimental setup.

Fig. 2. Diffuser plate with tubes (Plate L).

reached the top of the tank. In the actual molten steel in the tundish, inclusions transported upward are captured in a slag layer at the free surface. On the other hand, in the water tank in the present experiment, particles reached the free surface can go back to the bulk liquid while riding on the liquid circulation, and therefore, we need a way to collect particles in the vicinity of the free surface to evaluate the particle removal rate. Arai²³⁾ supplied water into a tank filled with a water-particle mixture in his particle removal experiments. The water supply caused an overflow of the mixture from the top of the tank. Collecting particles in the overflowed mixture enabled him to evaluate the particle removal rate. We also used the mixture overflow. Water for overflow was filled in the reservoir (Fig. 1) and was kept at a constant temperature by using a heater (Hakko-Denki, AWA1510, DGC1151). Water was supplied from the reservoir into the tank at a constant liquid volume flow rate, Q_L , using the pump (IWAKI, MD-6K) and the flow rate was measured using the flowmeters (Tokyo-Keiso, F06-408813; Nippon-Flowcell, NSP-1-RC1/4-L). The water inlet port of 12 mm in diameter was mounted at the center of the tank in the width direction and 50 mm above the plate. Particles overflowed with water were captured using conical flasks.

Three kinds of air diffuser plates were used to vary bubble size. The diffuser plate shown in Fig. 2 has 56 holes made of the tubes, which were evenly positioned at the pitch of 16 and 14.4 mm in the width and depth directions, respectively. The inner diameters of the tubes were 0.70 mm. The numbers of tubes of the other plates were 80 and 115. The configurations of the plates are summarized in **Table 1**. Since the gas flow rate from each hole decreases with increasing the total number of holes while keeping Q_G , the bubble size is expected to decrease by changing the number of holes from 56 to 115. Hereafter the diffuser plates with 56 (4×14), 80 (5×16) and 115 (5×23) holes are therefore referred to as Plate L, M and S, respectively.

The values of Q_G were 2.33×10^{-4} , 2.83×10^{-4} , and 3.33×10^{-4} m³/s, for which the gas superficial velocities, $J_G = Q_G/A_T$, were 0.0135, 0.0164 and 0.0193 m/s, respectively, where A_T is the horizontal cross-sectional area of the tank. These conditions are referred to as Low, Medium and High Q_G in the following. The Q_W was 3.0×10^{-5} m³/s and the liquid volume flux, $J_L = Q_L/A_T$, was 1.74 mm/s. The effects of Q_W will be discussed later.

The true density (solid density), ρ_P , and the apparent density of particles were 2 050 and 1 480 kg/m³, respectively. The former was evaluated by measuring the mass and volume of a water-particle mixture, and the latter was evaluated from the settling velocity of particles in stagnant water and Stokes' drag law. The true density of oxides is smaller than

that of molten steel, but we used particles heavier than water to make the evaluation of the removal rate easy. Terminal velocities of particles in stagnant water were preliminary measured from high-speed video images. Particles were put into stagnant water through the free surface. A high-speed video camera (PHOTRON, SA-X2, 60 fps) mounted on a microscope (NIKON, SMZ800) was used to observe particles falling through water. The measurement position was 50 mm below the free surface. The sampling number was 70 to obtain an accurate mean value. The mean terminal velocity was 2.64 mm/s. It should be noted that the terminal velocity is larger than J_L , and therefore, particles would fall down even with water supply from the sidewall of the tank if no bubbles were injected in the tank.

2.2. Evaluation of Particle Removal Rate Coefficient

The time evolution of the total number of particles, N_P , in the liquid phase is governed by the following first-order linear ordinary differential equation:¹²⁾

$$\frac{dN_P}{dt} = -KN_P \quad \dots\dots\dots (1)$$

where K is the reduction rate coefficient, and t the time. The validity of this form of the differential equation will be examined later. Arai²³⁾ used overflow of water to evaluate K . He therefore divided K into the following two parts:

$$K = k_B + k_L \quad \dots\dots\dots (2)$$

where k_B is the removal rate coefficient representing the contribution of bubbles, and k_L the contribution of water overflow, which can be evaluated as

$$k_L = \frac{Q_L}{V_T(1-\varepsilon_G)} \quad \dots\dots\dots (3)$$

where V_T is the tank volume, and ε_G the gas holdup. In Eq. (3), the distributions of the gas and liquid phase fractions are assumed to be uniform in the system. A particle heavier than the liquid falls down relative to the liquid phase, and therefore, the relative particle settling velocity, U_P , can have a negative contribution to K . This effect was not accounted for in the evaluation of K in Arai's experiments since it could be small because of small particle diameters (10 μ m) and of the relative density close to unity. In the present experiment, the particle diameter is however 100 μ m, so that U_P is not small and the settling motion of particles can affect K . Hence we take into account the particle settling effect on K as

$$K = k_B + k_L + k_F \quad \dots\dots\dots (4)$$

where

$$k_F = \frac{U_P A_T}{V_T} \quad \dots\dots\dots (5)$$

Here $U_P < 0$ for particles falling relative to the liquid phase. The k_B should therefore be regarded as the removal rate coefficient for neutrally-buoyant particles. Integrating Eq. (1) yields

$$N_P = N_{P0} e^{-K(t-t_0)} \quad \dots\dots\dots (6)$$

where N_{P0} is N_P at the initial time t_0 . The K is therefore

Table 1. Plate configuration.

	Plate S	M	L
Number of tubes	115	80	56
	(5 × 23)	(5 × 16)	(4 × 14)
Inner (hole) diameter [mm]	0.51	0.58	0.70
Pitch in width direction [mm]	10	14	16
Pitch in depth direction [mm]	12	12	14.4

given by

$$K = -\frac{\ln(N_P / N_{P0})}{t - t_0} \dots\dots\dots (7)$$

Substituting Eq. (4) into Eq. (7) yields the following expression for k_B :

$$k_B = -\frac{\ln\left(\frac{N_P}{N_{P0}}\right)}{t - t_0} - \frac{Q_L}{V_T(1 - \varepsilon_G)} - \frac{U_P A_T}{V_T} \dots\dots\dots (8)$$

Since $M_P = N_P \rho_P \frac{\pi d_P^3}{6}$ and $M_{P0} = N_{P0} \rho_P \frac{\pi d_P^3}{6}$, we obtain

$$k_B = -\frac{\ln\left(\frac{M_P}{M_{P0}}\right)}{t - t_0} - \frac{Q_L}{V_T(1 - \varepsilon_G)} - \frac{U_P A_T}{V_T} \dots\dots\dots (9)$$

This is the basis of k_B evaluation. It should be noted that the second term in the RHS for the water overflow is based on the assumption that particle removal uniformly takes place over the cross section. This did not hold in the experiments and Q_L still had some effects on k_B even after subtraction of the second term as described in the next section. We therefore introduced the extra Q_L effect, $\hat{k}_L$, based on the experimental data of k_B for several Q_L .

$$k_B = K - k_L - k_F - \hat{k}_L$$

$$= -\frac{\ln\left(\frac{M_P}{M_{P0}}\right)}{t - t_0} - \frac{Q_L}{V_T(1 - \varepsilon_G)} - \frac{U_P A_T}{V_T} - \hat{k}_L \dots\dots\dots (10)$$

3. Results and Discussion

3.1. Characteristics of Bubbly Flows

In experiments on removal of fine particles in bubbly flows, it is important to understand the characteristics of the bubbly flows, *i.e.* the bubble diameter, the gas holdup and so on. Experiments on bubbly flows without particles were first carried out to measure the above-mentioned quantities.

Bubble diameters were measured from images obtained by using a high-speed video camera (Photron, SA-X2, 60 fps). Recordings were carried out at 110 mm above the tank bottom and at three positions, *i.e.* 40, 120 and 200 mm from the sidewall of the tank. The sphere-volume-equivalent bubble diameter was calculated with the assumption of ellipsoidal bubble shape. 300 bubbles were randomly selected in each condition. The mean bubble diameter, d_B , was calculated by taking number average for the selected bubbles.

Figure 3 shows the bubble size distributions for (a) Plate S, (b) Plate M, and (c) Plate L at $J_G = 0.016$ m/s. The bubble size distribution shifts from small to large diameters from left (Plate S) to right (Plate L). **Table 2** shows the mean bubble diameter with each plate for three gas flow rate conditions. The mean bubble diameter changes even at the same gas flow rate by changing the diffuser plate. Most of the bubbles are within ± 1.5 mm from the mean diameters. Davidson and Amick²⁷⁾ proposed the following empirical correlation for the bubble frequency for bubbles generated from an orifice:

$$f_B = \frac{12.26 Q_O^{0.13}}{d_O^{0.43}} \dots\dots\dots (11)$$

where Q_O is the gas volume flow rate from an orifice and d_O the orifice diameter. The mean bubble diameter is given by

$$d_B = \sqrt[3]{\frac{6 Q_O}{\pi f_B}} \dots\dots\dots (12)$$

By substituting the gas flow rate from each hole and the hole diameter into Eq. (12), d_B were calculated and were compared with the present data as shown in **Fig. 4**. They are in good agreement, which means that the bubbling regime in the present experiments is inertia dominant and the surface tension does not play any role in bubble size.

Fig. 3. Bubble size distribution (Low Q_G ($J_G = 0.016$ m/s)). (Plate, Mean bubble diameter [mm]) (a) (S, 4.4) (b) (M, 5.0) (c) (L, 5.9).

Table 2. Mean bubble diameter [mm].

$Q_G \times 10^{-4}$ [m ³ /s]	Plate S	M	L
2.3	4.1	4.7	5.6
2.8	4.4	5.0	5.9
3.3	4.7	5.3	6.1

Fig. 4. Mean bubble diameter compared with Davidson-Amick correlation (Dashed lines: $\pm 5\%$ error).

The total bubble volume in the tank was obtained by measuring the amount of water for filling up the tank while aeration. The gas holdups, ϵ_G , are shown in **Table 3**. The values are several percent in all the cases. The total numbers, N_B , of bubbles were calculated from ϵ_G and d_B as shown in **Fig. 5**. The N_B slightly increases with Q_G . Comparing N_B of dif-

Table 3. Gas holdup.

$Q_G \times 10^{-4} [\text{m}^3/\text{s}]$	Plate S	M	L
2.3	0.052	0.051	0.052
2.8	0.069	0.067	0.071
3.3	0.086	0.085	0.090

Fig. 5. Total number of bubbles, N_B , in tank.

ferent plates shows that N_B approximately proportional to the number of holes.

3.2. Removal Process and Removal Rate Coefficient

Figure 6 shows front images of the bubbly flow at Low Q_G . Initially ($t = t_0 = 0$) bubbles in the tank are invisible due to the presence of fine particles. The uniform color implies that particles are well dispersed and the particle concentration is almost uniform. The dotted lines represent a gutter, which surrounds the tank to lead overflowed water to the conical flask as shown in the image at 240 s. The visibility of the flow increases with t as a consequence of particle removal. If there were no bubbles, the particles settling velocity could be $U_P + J_L \sim -0.9 \text{ mm/s}$, for which particle sedimentation at the bottom of the tank would take 270 s for the tank height of 240 mm. Particles are still uniformly dispersed even in the image at 240 s, and therefore, bubbles effectively transport particles upward. The liquid phase becomes transparent at 720 s. Hence the particle removal has been completed.

Figure 7 shows the ratio, $\ln(M_P/M_{P0})$, of the total mass of particles to the initial total mass. The $\ln(M_P/M_{P0})$ linearly decreases with increasing $t - t_0$. Hence $K = -\ln(M_P/M_{P0})/(t - t_0)$ is constant and the model, Eq. (1), is valid. The K was evaluated from a regression line to the data.

Figure 8 shows $K - k_L - k_F$ measured at various Q_W . This quantity depends on Q_W . Hence the effect of the water inflow cannot be entirely removed with k_L only although k_L was introduced in Eq. (10) to eliminate that effect. The decrease in $K - k_L - k_F$ with increasing Q_W implies that the motion of water horizontally injected from the tank side-wall disturbed the upward motion of particles even at small

Fig. 6. Particle removal process (Q_W, Q_G) = ($3.0 \times 10^{-5}, 2.3 \times 10^{-4}$) [m^3/s], Plate S).

Fig. 7. Change in total particle weight, $\ln(M_P/M_{P0})$, at $(Q_W, Q_G) = (3.0 \times 10^{-5}, 2.3 \times 10^{-4})$ [m³/s] with Plate S.

Fig. 9. Removal rate coefficient k_B .

Fig. 8. Additional liquid inflow effect (Plate S).

Q_W . This water inlet effect can therefore be expressed as $k_L + \widehat{k}_L(Q_W)$ and $\widehat{k}_L(Q_W)$ was evaluated from the regression line shown in Fig. 8.

3.3. Effects of Gas Flow Rate and Bubble Diameter on k_B

Figure 9 shows k_B for all the conditions (three plates and three Q_G). The positive values of k_B mean that bubbles have abilities to bring hydrophilic fine particles upward even though the particles do not adhere to the bubble interfaces. The particle transportation can be attributed to the bubble wake as discussed for single bubbles in literature,^{17,21,22)} i.e. particles collide with bubble wake are entrained into the bubble wake at a certain probability and dragged upward while riding on the liquid flow in the wake. However as pointed out by Yang *et al.*²²⁾ particles may escape from the bubble wake after traveling some distances. In addition, bubble paths in the present range of d_B oscillate and bubble wakes are shed when a bubble changes its direction of motion,^{28–30)} resulting in many particle shedding from the near wake region. Such escaped particles however collide with wakes of trailing bubbles and entrained again. The k_B is almost independent of both plate types and Q_G , in other words neither d_B nor N_B affects k_B .

Fig. 10. k_B per a bubble, k_B/N_B .

Figure 10 shows k_B divided by N_B , i.e. the removal rate coefficient per a bubble. The coefficient increases with d_B at each Q_G . This might be due to the increase in the wake volume with increasing the bubble Reynolds number.³¹⁾ On the other hand, the coefficient decreases with increasing Q_G while keeping d_B . If all the bubbles in the tank are evenly positioned at a pitch of l in all the directions, l is given by

$$l = \sqrt[3]{\frac{V_T}{N_B}} \dots \dots \dots (13)$$

The upward transportation of particles by a bubble is to be effective in the volume, V_w , of the water-particle mixture between the bubble and its trailing bubble. Approximating V_w as the cylinder of the height, $l - d_B$, and the diameter of d_B yields

$$V_w = \left(\sqrt[3]{\frac{V_T}{N_B}} - d_B \right) \frac{\pi d_B^2}{4} \dots \dots \dots (14)$$

This volume decreases with increasing N_B at constant d_B . As shown in Table 2, d_B at Low Q_G of Plate M and that at High Q_G of Plate S are the same (4.7 mm), whereas N_B are 3 380 and 5 720, respectively, as shown in Fig. 5. $V_w = 9.6 \times 10^{-8}$ and 6.8×10^{-8} m³ for the former and the latter

cases, respectively. The smaller V_w in the latter could be one of the reasons of the small transportation efficiency by each bubble, k_B/N_B , at High Q_G .

In summary, the efficiency of particle removal by each bubble increases with bubble diameter, whereas the increase in the number of bubbles deteriorates the efficiency due to the reduction of the wake volume effective for particle transportation.

4. Conclusion

Experiments on removal of hydrophilic fine particles in bubbly flows were carried out. Air and water at room temperature and atmospheric pressure were used for the gas and liquid phases, respectively. Silica particles of 100 μm in diameter were used for inclusion. Bubbles were injected into water-particles mixtures in a rectangular tank and the amount of particles included in the mixture overflowed from the tank was measured to evaluate the removal rate coefficient. Three air diffuser plates and three gas volume flow rates were used to realize various conditions of bubble diameter and the number of bubbles. The data of the removal rate coefficients revealed that bubbly flows can transport fine particles upward. Because of the hydrophilic nature of the silica particles, the transportation of particles is caused not by attachment to the bubble interface but by the motion of bubble wake. Since the wake volume increases with the bubble Reynolds number (size and rise velocity), the removal efficiency for each bubble increases with bubble size. On the other hand, the increase in the number of bubbles decreases the efficiency for each bubble since the wake region effective for particle transportation reduces as the number of bubbles increases.

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