Particle-Size-Grouping Method of Inclusion Agglomeration and its Application to Water Model Experiments

Takehiro NAKAOKA, Shoji TANIGUCHI1), Katsutoshi MATSUMOTO1), and Stein Tore JOHANSEN2)

Kobe Steel Ltd., PhD Student of Graduate School of Engineering, Tohoku University, Aoba-yama 02, Aoba-ku, Sendai 980-8579 Japan. 1) Graduate School of Engineering, Tohoku University, Aoba-yama 02, Aoba-ku, Sendai 980-8579 Japan. 2) SINTEF Materials Technology, N-7465 Trondheim, Norway.

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Agglomeration of inclusions in liquid steel causes not only the enhancement of inclusion removal by flotation but also the increase in the number of large inclusions in final products. To clarify agglomeration behavior theoretically, a lot of studies have been made until now. However, the behavior is not clearly understood yet. In this study, a new particle-size-grouping (PSG) method has been established, which enables a simple calculation of the agglomeration by a small number of size groups with complete conservation in total particle volume. This method has been verified by the comparison with the exact solution of a revised population-balance equation. An experimental study of the agglomeration of polyvinyl-toluene latex (PVTL) in a stirred electrolyte solution has been made in an agitated vessel under a rapid agglomeration condition. An effective Hamaker constant of PVTL in water, $A_{131}$, has been obtained by adjusting the measured agglomeration curve with the curve calculated by the PSG method. Good agreement has been obtained between observed and calculated agglomeration curves for $A_{131} = 0.8 \times 10^{-20}$ J under a wide range of initial particle concentrations and agitation speeds. Numerical simulations of the fluid flow and particle transport in the vessel have been made to confirm the applicability of the PSG method. Computed agglomeration curves agree well with the theoretical curve if the energy dissipation rate averaged with the residence time of liquid in computational cells is used to calculate the dimensionless agglomeration time.

KEY WORDS: inclusion; turbulent agglomeration; clean steel; model experiment; agglomeration coefficient; Hamaker constant; numerical simulation; $K-\varepsilon$ model.

1. Introduction

In recent years, removal of inclusion is desired in steel industries to achieve higher cleanliness of steel products in order to improve workability, surface quality, and fatigue strength. The size of inclusions that should be removed from the products is required to be smaller year by year. In order to meet the demands for higher cleanliness, it is needed to clarify the mechanisms of inclusion removal such as flotation, agglomeration and adhesion to wall or bubble surfaces. Among them, the agglomeration of inclusion particles in liquid steel is essential because it results in the generation of enlarged inclusions harmful for final products. In order to meet the demands for higher cleanliness, it is needed to clarify the mechanisms of inclusion removal such as flotation, agglomeration and adhesion to wall or bubble surfaces. Among them, the agglomeration of inclusion particles in liquid steel is essential because it results in the generation of enlarged inclusions harmful for final products. On the other hand, the enlarged particles are easy to be removed due to large floating velocities. Therefore, it is quite important to control the agglomeration process to obtain clean steel products. In order to estimate the agglomeration process accurately, a number of numerical simulations have been made until now.\(^\text{2–6,10–16}\)

Saffman and Turner\(^\text{1)}\) proposed a turbulent collision model of spherical particles to explain the generation of raindrops in a cloud. This model has often been applied to the studies on the inclusion agglomeration in liquid steel. Linder\(^\text{2)}\) developed a general model of the rate of deoxidation based on the Saffman-Turner model and applied the model to the prediction of deoxidation curves in an ASEA-SKF furnace. Nakanishi and Szekely\(^\text{3)}\) applied the Saffman-Turner model to estimate the rate of Al deoxidation of liquid steel in an ASEA-SKF furnace. They found that the deoxidation rates estimated from the values of the energy dissipation rate, $\varepsilon$, calculated by a $K-\varepsilon$ turbulence model were larger than observed rates. They introduced a correction factor of 0.3–0.6 to the collision frequency of the Saffman-Turner model. Shirabe and Szekely\(^\text{4)}\) solved a set of 2-D transport equations of ten-size groups of inclusion particles from 2 to 20 $\mu$m by using a $K-\varepsilon$ model combined with the Saffman-Turner model. In their simulation, the 20 $\mu$m particle was assumed to be instantaneously removed by flotation. Sinha and Sahai\(^\text{5)}\) simulated inclusion behaviors composed of turbulent agglomeration, flotation and adhesion to walls in a continuous casting tundish by a 3-D transport analysis. They divided particles into 12-size groups from 25 to 150 $\mu$m and calculated the removal efficiency of each particle size. In the above studies except Nakanishi and Szekely\(^\text{3)}\), the Saffman–Turner model was applied without any correction for the agglomeration efficiency.
Taniguchi et al. applied Higashitani’s model to the determination of the agglomeration coefficient. This coefficient, less than unity in general, was derived from the interparticle forces composed of a viscous resistance force to remove a liquid film between particles and an attractive force due to the London–van der Waals force. They derived Hamaker constants of SiO₂ and Al₂O₃ in liquid steel and estimated the agglomeration coefficients for several steelmaking vessels. They also applied the estimated Hamaker constant of SiO₂ to the simulation of Si-deoxidation rate of liquid metal in an induction furnace. 10) Miki et al. analyzed the removal of inclusion particles in an RH degasser considering the agglomeration coefficient and made a clear simulation of inclusion agglomeration and flotation in a continuous casting tundish. They proposed a fractal structure of Al₂O₃ clusters and calculated the rate of agglomeration and flotation for 29-cluster levels starting from a 1-μm-diameter primary particle. In their calculation, the agglomeration coefficient was taken into account. Recently, Zhang et al. made a 3-D simulation of fluid flow in continuous casting tundishes and discussed the inclusion behaviors like agglomeration, flotation and adhesion to wall. In the calculation, particles were divided into 1 to 70 μm in radius with 0.1 μm interval and the agglomeration was estimated from average energy-dissipation rates in two regions separated by a weir in the tundish. They found that the value of the agglomeration coefficient should be 0.18 to obtain a good agreement with the measured result. The other studies have not been clearly shown in the previous studies. Another difficulty comes from the coexisting removal processes with agglomeration such as flotation and adherence to walls and bubble surfaces, all of which are not well elucidated yet. Furthermore, break-up of clusters may sometimes locally dominate over agglomeration, as discussed by Johansen and Taniguchi. Break-up must therefore be expected to affect the inclusion behavior in highly turbulent flows, and this will need further attention in the future.

In summary, the difficulty of the analysis of inclusion agglomeration comes from the complexity of the calculation of the particle transport equation considering the population balance of a variety of inclusion sizes. The particle-size grouping (PSG) method is effective to avoid this complexity, so that many previous studies used this method. However, how to conserve the total mass of particles has not been clearly shown in the previous studies. Another difficulty comes from the coexisting removal processes with agglomeration such as flotation and adherence to walls and bubble surfaces, all of which are not well elucidated yet. Furthermore, break-up of clusters may sometimes locally dominate over agglomeration, as discussed by Johansen and Taniguchi. Break-up must therefore be expected to affect the inclusion behavior in highly turbulent flows, and this will need further attention in the future.

Considering these unsolved problems, the present study aims to establish a precise method for analyzing the particle population balance in turbulent agglomeration. To achieve this, a water-model experiment on the particle agglomeration in an agitated vessel is made under the condition without particle removal. A particle-size grouping method with an accurate mass balance of particles is developed. And furthermore, a numerical simulation is made of the fluid flow and particle transport in an agitated vessel by applying the K–ε model and the PSG method.

2. Theories

2.1. Turbulent Collision and Agglomeration

Suppose two particles, i and j, in a liquid medium collide with each other. If the collision radius $a_{i} + a_{j}$ is far less than the smallest eddy size (Kolmogoroff’s micro scale: $\eta = (\nu / \epsilon)^{1/4}$), the collision frequency in a unit volume of liquid medium, $N_{ij}$, is given by Saffman and Turner as follows:

$$N_{ij} = 1.3(a_{i} + a_{j})^{3} (\nu / \epsilon)^{1/2} n_{ij}$$

where $a$ is particle radius, $\epsilon$ energy dissipation rate, $\nu$ kinematic viscosity, and $n$ particle number density. The population balance equation for the agglomerated particle composed of $k$-pieces of primary particles is indicated by the following equation derived by Smoluchowski:

$$\frac{dn_{k}}{dt} = \frac{1}{2} \sum_{i=1}^{k-1} N_{ij} - \sum_{i=1}^{\infty} N_{ik}$$

In this equation, the generation term is halved because collision pairs are doubly counted for $i$ and $j$. However, in the case where two particles having same size generates the $k$ particle, the generation term should not be halved as the collision combination is unique. Furthermore, the dissipation term should be doubled in the case where $i = k$. These inappropriate points have not been noticed until now and Eq. (2) has been used commonly in previous studies including Higashitani’s study and our previous study. A correct expression of the population balance equation is as follows:

$$\frac{dn_{k}}{dt} = \frac{1}{2} \sum_{i=1}^{k-1} (1 + \delta_{ij}) N_{ij} - \sum_{i=1}^{\infty} (1 + \delta_{ik}) N_{ik}$$

where $\delta_{ij}$ is the Kronecker’s delta function ($\delta_{ij} = 1$ for $i = j$, $\delta_{ij} = 0$ for $i \neq j$).

Higashitani et al. introduced the agglomeration coefficient, $\alpha$, to Eq. (1) to consider the effects of the viscous force to eliminate the liquid film between colliding particles and the London–van der Waals force.

$$N_{y} = \alpha [1.3(a_{i} + a_{j})^{3} (\nu / \epsilon)^{1/2} n_{ij}]$$

Under a rapid agglomeration condition free from the repulsive force due to electrical double layers, $\alpha$ is correlated by the following approximate equation.

$$\alpha = 0.727 \left[ \frac{\mu a^{4} (\nu / \epsilon)^{1/2}}{A_{131}} \right]^{-0.242}$$

where $A_{131}$ is effective Hamaker constant of particle 1 in medium 3, $\mu$ is viscosity of liquid and $a_{i}$ is radius of the primary particle. In a strict sense, $\alpha$ depends upon the radius of agglomerate, however $\alpha$ is given by $a_{i}$ because of the difficulty in the derivation of a suitable function. Practically, this approximation is generally accepted in previous works. It should be noted that the London–van der Waals force is the unique attractive force in liquid metals and the above equations can be applied to the inclusion agglomeration in liquid metals.
2.2. Particle-Size-Grouping (PSG) Method

Dimensionless form of Eq. (3) combined with Eq. (4) is expressed by the following equation.

\[
\frac{dn_k}{dt} = \frac{1}{2} \sum_{i=1}^{k-1} (i^{3} + j^{3}) (1 + \delta_k) n_i n_j - \sum_{i=1}^{k} (i^{3} + k^{3}) (1 + \delta_k) n_i n_k^* 
\]

where \( n_k^* = n_k / N_0 \), \( t* = 1.3 \alpha a^2 (v / w)^2 N_0 M \), \( N_0 \) is the initial number density of primary particles and \( N_M \) the number of primary particles composing the largest agglomerated particle. Equation (6) can be solved numerically considering every particle size from the primary particle to the largest agglomerate. However, the load of calculation becomes large as the size of the largest particle increases considerably. Especially, if the Eq. (6) is combined with the particle transport equation, tremendous efforts should be made to solve the equations. In such cases, the following particle size grouping method is superior to this rigorous procedure.

In the present PSG method, particles are divided into \( M \) groups having characteristic particle volume from \( v_1 \) to \( v_M \). The particle volume of each group is determined so as to increase from one group to the next with a constant ratio, \( R_v = v_2 / v_1 = \text{const.} \)

**Table 1.** List of combination of collision between two particles divided into eight size groups for \( R_v = 2 \).

<table>
<thead>
<tr>
<th>Group</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume</td>
<td>1.0</td>
<td>2.0</td>
<td>4.0</td>
<td>8.0</td>
<td>16.0</td>
<td>32.0</td>
<td>64.0</td>
<td>128.0</td>
</tr>
<tr>
<td>Radius</td>
<td>1.00</td>
<td>1.26</td>
<td>1.59</td>
<td>2.00</td>
<td>2.52</td>
<td>3.17</td>
<td>4.00</td>
<td>5.04</td>
</tr>
<tr>
<td>Threshold</td>
<td>1.13</td>
<td>1.42</td>
<td>1.79</td>
<td>2.25</td>
<td>2.85</td>
<td>3.59</td>
<td>4.52</td>
<td>5.40</td>
</tr>
</tbody>
</table>

Fig. 1. Definition of particle size distribution in PSG method.
In the PSG method, it is easy to introduce the fractal theory proposed by Tozawa et al.\textsuperscript{12) by the use of the following relationship between $a_k^*$ and $v_k^*$.

$$a_k^* = (v_k^*)^{1/D}; \quad D = \text{fractal dimension} \quad \text{(11)}$$

In the present study, $D = 3$ is adopted. Tozawa et al. proposed $D = 1.8$ for Al$_2$O$_3$ cluster in liquid steel. The applicability of the fractal dimension will be investigated in the Appendix.

2.3. Evaluation of PSG Method

In order to evaluate the accuracy of the present PSG method, an exact solution of Eq. (6) was obtained first. Runge–Kutta–Gill method was applied to integrate Eq. (6) and the time step for integral $\Delta t^*$ was selected as 0.001. The initial number distribution was given by $n_k^* = 1$ for $k = 1$ and $n_k^* = 0$ for $k > 1$. The maximum size of agglomerate was set as $N_M = 10^4$. In this case, 0.1\% accuracy in the total volume of particles is guaranteed up to $t^* = 1$.

Figure 2 gives the computed results of the change in the total particle number density with time obtained by the PSG method compared with the exact solution. In the PSG method, the maximum number of the size group $M$ was 20. There is good agreement between the PSG method and the exact solution in both cases of $R_v = 2$ and 2.5. In the same figure, a comparison is made between the conventional calculation based on the population balance equation, Eq. (2), and the present calculation base on Eq. (3). There is a considerable difference between these results. Figure 3 shows the change in the number densities of each size group with time. There is also good agreement between the PSG method and exact solution. In the Appendix, the effect of $R_v$ on the accuracy of the PSG method is discussed in detail. From these results it is concluded that the PSG method is applicable in good accuracy. In the following chapter $R_v = 2$ is used for calculation. It is worth to note that the present PSG method is available to estimate the inclusion agglomeration with a wide size range by a small computational load.

3. Experimental

3.1. Experimental Method

Figure 4 shows the experimental apparatus used for the turbulent agglomeration of particles in water. The agitated vessel was composed of a cylindrical glass vessel (750 ml), a two-blade impeller and four baffle plates. Polyvinyl-
Toluene latex (PVTL) with 2.02 μm in mean diameter was used as a model particle and NaCl aqueous solution was used as a liquid medium. In the experiment, particles suspended in distilled water were put into the solution agitated in the vessel. At suitable time intervals, a small amount of solution was taken, and the particle concentration was measured by the electric sensing zone method (Coulter Counter II). By a preliminary experiment, a rapid agglomeration condition was found to be achieved at \[ C_{NaCl} = 3 \times 10^3 \text{ mol/m}^3 \]. Experimental conditions are shown in Table 2 and the initial size distribution of PVTL is shown in Fig. 5.

Turbulent energy dissipation rate was obtained from the measured torque, \( T_S \), by the following equation.

\[
\varepsilon = 0.15 \times \frac{2\pi n_S T_S}{W} \quad \text{................................(12)}
\]

where \( n_S \) is agitation speed, and \( W \) is mass of liquid. The coefficient 0.15 means that the input power density partly dissipates in liquid, which is suggested in the previous work.\(^{19}\) Furthermore, a numerical simulation of turbulent flows in an agitated vessel confirmed the evidence of this coefficient. The measured value of \( \varepsilon \) for the present experimental condition were (3.9–838)×10\(^{-4}\) m\(^3\)s\(^{-3}\) for \( n_S = 200–800\ \text{min}^{-1} \), which corresponded to the smallest eddy size ranging from 131 to 59 μm. As these eddy sizes are larger than the observed agglomerates, the present theory can be applied.

### 3.2. Results and Discussion

An optical micrograph of agglomerated particles after experiment is given in Fig. 6. It is seen that particles agglomerate into a dense cluster. Although the structure of the cluster is not known, the cluster seems denser than \( \text{Al}_2\text{O}_3 \) clusters shown by Tozawa et al.

Figure 7 shows the change in total number densities with time for various initial concentration of PVTL at \( n_S = 400 \ \text{min}^{-1} \). To compare the measured agglomeration curves with calculated one, the Hamaker constant of PVTL in water, \( A_{131} \), should be known. By using Eq. (5) and adjusting \( A_{131} \) so as to fit the measured curve with the calculated one, \( A_{131} \) can be determined. Consequently, \( A_{131} \) was obtained as \( 0.8 \times 10^{-20} \text{ J} \) which by all measured curves fit well with the calculated curve as seen in Fig. 7. This value is very close to the theoretical value of polystyrene latex in water, \( A_{131} = 0.95 \times 10^{-20} \text{ J} \).\(^{20}\) The concentration change with time for each size group, 1, 3, 4 and 6 (see Table 1), is indicated in Fig. 8 for various initial concentrations. Although the observed curves are a little larger than the calculated ones at large \( t^* \), reasonable agreement can be seen in the range \( t^*<1 \). This discrepancy may be attributed to the breakup of larger agglomerates than the 6th group.

The agglomeration curves at various agitation speeds are given in Fig. 9. In this case also the measured curves fit well with the calculated curves. Figure 10 shows the comparison between observed and calculated agglomeration curves for groups 1, 3, 4 and 6. The observed curves fit well with the calculated curves except for the large \( t^* \) region.
4. Numerical Simulation of Fluid Flow and Particle Agglomeration

4.1. Fundamental Equation

It is well known that the value of $e$ changes widely with the position in an agitated vessel. In order to make clear the effect of $e$ distribution in the vessel on agglomeration rates, numerical simulations on fluid flow and particle transport are carried out. Simulations were made for the case of Fig. 9, in which the volume fraction of particle in liquid is $0.87 \pm 10^{-2}$. Therefore the flow of liquid may not be affected by particles. In the present simulation the fluid flow in the agitated vessel used in this study was computed first, and particle transport equations were then solved using the computed flow field. FLUENT 4.521) and MixSim1.5 were used. The standard $K$–$e$ model22) was adopted for the simulation. For the boundary conditions, non-slip condition and the wall function were applied at walls and free slip condition was applied at the free surface of liquid. The calculation was performed in a half region of the vessel because of the two-blade impeller. The density and viscosity of liquid used for the calculation was the same as Table 2.

The transport equation of particles in the liquid coupled with the PSG method is given by the following equation.

$$\frac{\partial n_k}{\partial t} + \frac{\partial u_i n_k}{\partial x_i} = \frac{\partial}{\partial x_i} \left( D_i \frac{\partial n_k}{\partial x_i} \right) - \sum_{j=1}^{M-1} \frac{\delta_{ijkl}}{(1+\delta_{ijkl})} \frac{\partial^2 n_k}{\partial x_i \partial x_j} + \sum_{j=1}^{M-1} \frac{\delta_{ijkl}}{(1+\delta_{ijkl})} \frac{\partial^2 n_k}{\partial x_i \partial x_j}$$

In the calculation, PSG method was added in a user-defined subroutine and $M=10$ was used to save computational time. The turbulent diffusivity of the particles, $D_t$, is obtained from the turbulent viscosity $\mu_t$ by the following equation:

$$D_t = \mu_t / \rho = 0.09K^2 / \epsilon$$

where $\rho$ is liquid density and $K$ is turbulent energy.

The configuration of the vessel and a mesh system are indicated in Fig. 11. The calculation was made in the half re-
region of the vessel. The number of mesh was (65 × 21 × 32).

4.2. Results and Discussion

Figure 12 shows the velocity distribution in the vertical plane including the axis. There is a strong down-flow just below the impeller due to the inclined blades (45°), and two vortices are formed in the outside of the impeller. The distribution of $\varepsilon$ is given in Fig. 13. The value of $\varepsilon$ is quite large near the impeller and small in the upper region of the vessel. There is almost a 1 000-fold difference in $\varepsilon$ with the position in the vessel.

To compare the computed result with the observed one, the average value of $\varepsilon$ in the vessel is needed. Two different averages are taken, one is a volume average described by Eq. (15), and the other is a residence-time average given by Eq. (16).

\[
\overline{\varepsilon}_v = \sum \varepsilon_i V_i / \sum V_i \quad \text{.................(15)}
\]

\[
\overline{\varepsilon}_r = \sum \varepsilon_i T_i / \sum T_i \quad \text{.................(16)}
\]

where $V_i$ is the volume of $i$-cell and $T_i$ is the residence time of liquid in $i$-cell. $T_i$ is obtained by Eq. (17).

\[
T_i = V_i^{1/3} |u| \quad \text{.................(17)}
\]

where $u_i$ is the velocity in the $i$-cell.

Table 3 shows a comparison between measured and calculated values (by Eq. (12)) of averaged $\varepsilon$. Although the residence-time average is a little smaller than the observed values of $\varepsilon$, the differences between observed result and calculated results are not large. Figure 14 indicates the distribution of the number density of the first size group in the vessel after 3 600 s. There can be seen a negligible difference in particle concentration in the vessel because of a strong mixing. Complete mixing assumption is reasonable in this case. Figure 15 shows the normalized plots of the computed total particle-concentration change with time obtained from Eq. (13). The normalization is made using two averages of $\varepsilon$, and these results are compared with the exact agglomeration curve for uniform $\varepsilon$. It is found from the figure that the computed agglomeration curves agree well with
the exact solution in the case where the residence-time average is taken for \( \varepsilon \). For the case of volume average, computed curves shift a little larger time. Although the difference is not large, it seems better to take the residence-time average than the volume average because the agglomeration depends on not only \( \varepsilon \) but also residence time.

5. Conclusions

In order to analyze accurately the inclusion-size distribution and its change with time in liquid steel, a new particle-size-grouping (PSG) method has been developed, and applied to the analysis of turbulent agglomeration of PVTL particles in agitated water. Numerical simulation on fluid flow and particle transport has been made to confirm the applicability of the PSG method. The following results have been obtained in this study.

(1) A correction is made in the population balance equation proposed by Smoluchowski\(^{(13)}\), which has been widely used in the previous studies.

(2) PSG method is confirmed by comparing with the exact solution of the corrected population balance equation. For the cases of \( R_i = 2 \) and 2.5, satisfactory agreement is verified between the agglomeration curves by the exact method and PSG method.

(3) Experiments on the agglomeration of polyvinyltoluene latex (PVTL) in NaCl aqueous solution are made in an agitated vessel under a rapid agglomeration condition. The agglomeration curves for various particle concentrations and agitation speeds agree well with the calculated results by the PSG method. The Hamaker constant of PVTL in water is found to be \( 0.8 \times 10^{-20} \) J.

(4) Numerical simulations are made of the fluid flow and particle transport accompanied by turbulent agglomeration. Computed volume-averaged and residence-time-averaged energy-dissipation rates at various agitation speeds are consistent with the measured values obtained by the torque of the impeller. The PSG method is successfully applied to the agglomeration analysis.

Nomenclatures

\[ D_i : \text{Turbulent diffusivity (m}^2/\text{s}) \]
\[ \bar{v}_i : \text{Critical size number of agglomerated particle} \]
\[ K : \text{Turbulent energy (m}^2/\text{s}^2) \]
\[ M : \text{Maximum number of particle-size group} \]
\[ N_{ij} : \text{Collision frequency of particles} \ i \text{ and} \ j \text{ in a unit volume of liquid (m}^{-3} \text{s}^{-1}) \]
\[ N_0 : \text{Initial total number density of particle (m}^{-3}) \]
\[ N_M : \text{Number of elementary particle in largest agglomerated particle} \]
\[ n : \text{Particle number density (m}^{-3}) \]
\[ n_s : \text{Agitation speed (min}^{-1}) \]
\[ n_t : \text{Total number density of particle (m}^{-3}) \]
\[ R_i : \text{Volume ratio between neighboring size groups of particle} (v_i/v_{i-1}) \]
\[ T_0 : \text{Threshold size ((a_i+a_{i+1})/2) (m)} \]
\[ T_r : \text{Residence time of liquid in i-cell (s)} \]
\[ T_s : \text{Time step for integral (s)} \]
\[ \alpha : \text{Agglomeration coefficient} \]
\[ \beta : \text{Kronecker’s delta function} \]
\[ \varepsilon : \text{Turbulent energy dissipation rate (m}^2/\text{s}^3) \]
\[ \bar{\varepsilon}_r : \text{Volume-average of turbulent energy dissipation rate (m}^2/\text{s}^3) \]
\[ \bar{\varepsilon}_p : \text{Residence-time-average of turbulent energy dissipation rate (m}^2/\text{s}^3) \]
\[ \eta : \text{Smallest eddy size ((v_0/\varepsilon)^{-1/4}) (m)} \]
\[ \mu : \text{Viscosity (Pa s)} \]
\[ \mu_t : \text{Turbulent viscosity (0.09kT/\varepsilon) (Pa s)} \]
\[ v : \text{Kinematic viscosity (m}^2/\text{s}) \]
\[ \rho : \text{Density of liquid (kg/m}^3 \]
\[ \zeta : \text{Correction factor for mass balance of particle} \]
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Superscript

* : Dimensionless value

— : Averaged value

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Appendix

1. Effect of $R_v$ on the Accuracy of the PSG Method

Difference between the PSG method and the exact solution of the total number density of particles is investigated for the wide range of $R_v$. Figure A1 indicates the relation between the value of $(n_{PSG} - n_{exact})/n_{exact}$ and $R_v$ at two agglomeration times, $t^*=0.5$ and 1.0. In the case that $R_v$ is larger than 2.08, the value of $d_n$ is zero around $R_v=2.4$ for both agglomeration times. It increases with decreasing $R_v$ and reaches 15% at $R_v=2.08$ for $t^*=0.5$. In the case that $R_v$ is smaller than 2.08, the value of $d_n$ is zero at about $R_v=1.9$. The critical change observed at $R_v=2.08$ results from the change in $i_c, k$ as described in Eq. (10). From these results, it is concluded that the value of $R_v$ should be chosen adequately to obtain a good accuracy for the PSG method even though the total particle mass is conserved at any values of $R_v$. The value of $R_v$ used in the present study ($R_v=2.0$) is found to give an accuracy of less than 5%.

2. Fractal Dimension of Agglomerate

Tozawa et al. introduced a new concept of fractal dimension shown in Eq. (11) to inclusion flotation and agglomeration. They found that the fractal dimension was 1.8 for Al$_2$O$_3$ cluster of its constituent particle diameter was 3 μm. In the present study, the agglomerate of PVTL also formed a cluster-like shape as seen in Fig. 6. For examination, the fractal dimension of 1.8 was adopted to the agglomeration of PVTL particle. Figure A2 indicates the results of calculation for $D=1.8$ compared with $D=3$. As the collision radius increases rapidly with the increase in the number of the size group of agglomerate for the case $D=1.8$, the agglomeration proceeds very quickly around $t^*<0.1$, which seems unrealistic. Considering reasonable agreement between observed and calculated agglomeration curves seen in the present study with $D=3$, the unrealistically rapid agglomeration for $D=1.8$ may be attributed to that the fractal dimension of the PVTL agglomerate was closer to 3 than 1.8. Further studies are needed to make clear the agglomeration behavior of cluster-shaped inclusion.