Interaction between Alumina Inclusions in Molten Steel Due to Cavity Bridge Force

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The formation and extinction behavior of cavity bridges has been experimentally evaluated by allowing the cylindrical particles simulating inclusions to approach and separate off in mercury and in molten steel. An interaction model of spherical alumina particles close to the actual inclusion shape due to cavity bridge force has been developed on the basis of the experimental results. Using this interaction model, the processes of agglomeration and separation of alumina inclusions in molten steel have been analyzed, and the agglomeration force due to cavity bridge force has been discussed in comparison with different agglomeration forces that are derived from the van der Waals force in molten steel and the capillary force on the surface of molten steel. When two isospherical alumina inclusions are approaching each other in molten steel, a large agglomeration force of $1.54d\cdot\gamma_{Fe}$ (d: the diameter of alumina inclusions, $\gamma_{Fe}$: the surface tension of molten steel.) is generated by the cavity bridge formation from the interparticle surface distance of 0.07d, and then the agglomeration force also gradually increases to reach the maximum value of $1.88d\cdot\gamma_{Fe}$ in complete contact state. Conversely, when two isospherical alumina inclusions in molten steel are separated from the contact state, a large agglomeration force of $0.92d\cdot\gamma_{Fe}$ and above is maintained until the cavity bridge extinction in the interparticle surface distance of 0.12d, whereas the agglomeration force gradually decreases from $1.88d\cdot\gamma_{Fe}$. In addition, it is assumed that alumina inclusions in aluminum deoxidized molten steel principally agglomerate and coalesce on the basis of the agglomeration force derived from very strong cavity bridge force in comparison with the van der Waals force in molten steel and the capillary force on the surface of molten steel, and coarse alumina clusters are thus formed in molten steel.

KEY WORDS: continuous casting; agglomeration and coalescence; alumina cluster; agglomeration force; cavity bridge force; interfacial chemical interaction; Al deoxidized molten steel.

1. Introduction

To meet the recent demand for higher quality, alumina inclusions must be removed from molten steel as coarse inclusions that are favorable for floating separation. This requires a scientific understanding of the agglomeration mechanism of suspended alumina inclusions in molten steel for facilitating agglomeration and coalescence of the alumina inclusions.

In a previous report, the author established a new experimental method that extracts the agglomeration force exerted between alumina particles in molten steel separately from fluid dynamic action and thus directly determines the agglomeration force to elucidate the agglomeration mechanism of alumina inclusions in molten steel in view of the interfacial chemical interaction between molten steel and inclusions. Using the above-mentioned method, it was demonstrated that a large agglomeration force is exerted between the alumina particles in molten steel, and through the analysis of the actually measured agglomera-

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interaction model due to the cavity bridge force in which the interparticle surface distance is considered. This model was logically extended to that of spherical alumina particles whose form resembles that of actual inclusions; thus, the interparticle surface distance affecting the agglomeration force between the alumina inclusions in molten steel due to the cavity bridge force was quantitatively evaluated. In addition, the agglomeration force due to the cavity bridge force calculated by the present interaction model was compared with the other agglomeration forces derived from the van der Waals force in molten steel and the capillary force on the surface of molten steel. On the basis of this comparison, the agglomeration mechanism of alumina inclusions in molten steel was studied from the viewpoint of interfacial chemical interaction.

2. Experimental Methods

2.1. Experiments on the Cavity Bridge Formation of Glass and Copper Cylinders in Mercury

The behavior of cavity bridges formed between inclusions in molten steel was examined by experiments using silica glass cylinders in mercury as simulated inclusions. Figure 1 shows an experimental device for cavity bridge formation in mercury. The transparent acrylic vessel for mercury is 60 mm in width, 65 mm in height, and 30 mm in depth (interior dimensions). Two pieces of glass cylinder of 4–8 mm in diameter and 25 mm in length were attached in parallel to under parts of a fixed arm and a moving arm of iron so as to come in line contact with each other; the circular sections of the glass cylinders were closely contacted with the front of the transparent acrylic vessel so as to enable observation of cavity bridges in mercury. The fixed arm was mounted on the transparent acrylic vessel, whereas the moving arm was mounted on the moving stage to work with a dial having the same scale mode as a micrometer. Rotation of the dial allows the glass cylinder on the moving arm to move in the direction of approach or separation under the condition that it is parallel to that on the fixed arm, and the moving distance of glass cylinders can thus be read from the indicated value on the scale.

The transparent acrylic vessel was filled with mercury to a level 15 mm above the center of the glass cylinder. The glass cylinders were allowed to fully be in contact with each other in mercury such that a cavity bridge was formed. Starting at that position, the glass cylinder of the moving arm was moved in the direction of separation, and a change in the cavity bridge was observed. The space between the glass cylinders was allowed to further widen by several mms after extinction of the cavity bridge until the interaction between the glass cylinders was completely extinguished; thereafter, the glass cylinder on the moving arm was gradually allowed to approach the glass cylinder on the fixed arm. The state of cavity bridge formation was observed while the glass cylinders approached and until they finally contacted with each other. For the purpose of varying the wettability of the cylinders with the mercury, copper cylinders were used instead of the glass cylinders in some experiments.

2.2. Experiment on Cavity Bridge Formation between Al₂O₃ Cylinders in Molten Steel

An experiment in which the Al₂O₃ cylinders between which the agglomeration force was measured in the previous report¹ were immersed in molten steel was conducted to examine the influences of the interparticle surface distance on the cavity bridge formation between the Al₂O₃ inclusions in molten steel. Figure 2 shows an experimental device for cavity bridge formation in molten steel. A pair of Al₂O₃ cylinders for cavity bridge observation was prepared such that two isodiametric Al₂O₃ cylinders (a high purity of 99.6 mass%) of 8 mm in diameter and 50 mm in length were placed parallel to each other; a superposed iron foil (50 µm in thickness) with a thickness of 0.1–1.2 mm was sandwiched between the Al₂O₃ cylinders, for the purpose of adjusting the surface distance between the Al₂O₃ cylinders as well as filling the space between the same with molten steel, and both ends of the Al₂O₃ cylinders were fixed with alumina cement. One end of the alumina cylinders was attached to the tip of an alumina protective tube with an outer diameter of 8 mm and a length of 500 mm so that they could be immersed in molten steel. The melting furnace is a resistance heating furnace of which the heating element is a high-frequency induction heated graphite cylinder. Six hundred grams of electrolytic iron was charged into an alumina crucible with an inner diameter of 40 mm and a height of

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Footnote:

1. Previous report refers to a previous study or publication that is not detailed in this text.
150 mm and melted under an Ar gas atmosphere. The molten steel was constantly maintained at a temperature of 1600°C, and a pair of the fixed Al₂O₃ cylinders with the sandwiched iron foil was immersed in non-deoxidized molten steel (at a dissolved oxygen concentration of approximately 0.025 mass%). After Al deoxidation at a target Al concentration of 0.08 mass% (equilibrium oxygen concentration of 0.0004 mass%), the alumina cylinders were held in Al deoxidized molten steel for five min. Subsequently, the power source of the melting furnace was disconnected so that the molten steel was solidified as the Al₂O₃ cylinders were immersed. After the experiment, the Al₂O₃ cylinders together with the solidified steel ingot were cut in cross-section, and the state of cavity bridge formation was observed. In addition, when two isodiametric Al₂O₃ cylinders which sandwich the steel foil and whose ends are coupled to each other with alumina cement are immersed in molten steel, the surface distance between the Al₂O₃ cylinders increases due to thermal expansion of the connections that are made of the alumina cement. Conversely, this distance decreases due to thermal expansion in the radial direction of the Al₂O₃ cylinders themselves; so even if the surface distance between the cylinders in the molten steel is calculated (thermal expansion coefficient of Al₂O₃ of 8.1 × 10⁻⁶ K⁻¹), it becomes no more than 1.3% greater than the value defined at room temperature. Consequently, in this research, the effect of the thermal expansion on the surface distance between the cylinders can be ignored.

3. Experimental Results

3.1. Cavity Bridge Formation between Glass Cylinders and between Copper Cylinders in Mercury

Figure 3 shows the changes in cavity bridges in the case of separating glass cylinders or copper cylinders in mercury from the contact state with each other. Here, a is the surface distance between these cylinders and \( d_{\text{CY}} \) is the diameter of the cylinders. No cavity bridges were observed between the cylinders in a series of processes (Figs. 3(E)–3(H)) where the copper cylinders were gradually separated away from the state where they came in complete contact with each other in mercury. Meanwhile, as the glass cylinders were allowed to come in contact with each other in mercury, a cavity bridge was formed in the contact portion (Fig. 3(A)). As the glass cylinders were separated away from each other, the width of the neck of the cavity bridge gradually became smaller (Fig. 3(B)) and reached a minimum at a certain surface distance between them (Fig. 3(C)). As the glass cylinders were further separated away from each other, the width of the neck of the cavity bridge became somewhat enlarged. The mercury gradually filled the cavity bridge from the inside toward the surface; finally, the depression of the cavity bridge shape was extinguished (Fig. 3(D)). Conversely, when the glass cylinders were allowed to approach each other in mercury, a cavity bridge was formed while the change in the process of separation was reversely traced back, and further, as the neck of the cavity bridge slowly widened, the glass cylinders came in complete contact with each other.

On the basis of the above-mentioned results, in either case of the approach and separation of the glass cylinders in mercury, it was considered that when the neck of cavity bridges comes to have the smallest width observed from the front of the transparent acrylic vessel, a real cavity bridge is formed (in case of approach) or extinguished (in case of separation) between the cylinders lying within the mercury from the observation face; therefore, the surface distance between the cylinders at that time was defined as the \( d_{\text{CB},\text{Max}} \) of the maximum length of cavity bridges. The relation between the diameter of the glass cylinders and the maximum length of cavity bridges is shown in Fig. 4. The maximum length of cavity bridges is the mean value obtained by averaging the values measured by alternating the approach and separation of the above-mentioned glass cylinders several times for each case of separation and approach. While the maximum length of cavity bridges in the case of approach (\( D_{\text{CB},\text{Max}}^{\text{a}} \)) is smaller that in the case of separation (\( D_{\text{CB},\text{Max}}^{\text{s}} \)), it has a positive value (\( D_{\text{CB},\text{Max}}^{\text{a}} = 0.40–0.42 \) mm, \( D_{\text{CB},\text{Max}}^{\text{s}} = 0.23–0.24 \) mm) in either case. Thus, it is found that cavity bridges are formed even where the glass cylinders are not in complete contact with each other. Within this measurement range, the cylinder diameter had relatively small influences on the maximum length of the cavity bridges. In addition, the data spread on the maximum length of the cavity bridges in Fig. 4 seems to be caused by some errors in measurement of the surface distance between the cylinders when the neck of cavity bridges comes to have the smallest width and to
be no correlation with the cylinder diameter.

3.2. Cavity Bridge Formation between Al₂O₃ Cylinders in Molten Steel

The state of the cavity bridge formation in the section of the Al₂O₃ cylinders in the solidified steel ingot is shown in Fig. 5. Although the space between the Al₂O₃ cylinders is filled with molten steel while they are separated away from each other (Figs. 5(A) and 5(B)), cavity bridges are observed as the Al₂O₃ cylinders comparatively approach each other closer (Figs. 5(C), 5(D), and 5(E)). It can be seen in the molten steel that cavity bridges are formed under the condition that the Al₂O₃ cylinders are not yet in complete contact with each other. In addition, although the cavity bridges are concavely curved toward the steel according to the after-mentioned interaction model in Fig. 7 when the agglomeration force interacts between the Al₂O₃ cylinders in molten steel, they in Figs. 5(D) and 5(E) are slightly convex toward the steel. It is assumed that this is because the curvature of the cavity bridge in molten steel was not maintained due to the deformation caused by the solidification shrinkage until after the solidification.

The state of the cavity bridge formation between the two isodiametric Al₂O₃ cylinders in the molten steel experiment corresponding to the diameter of the cylinders and the surface distance between the cylinders is collectively shown in Fig. 6. Each case of the presence and absence of cavity bridges is marked with the symbols of ○ and ●, respectively. For the Al₂O₃ cylinders of 8 mm in diameter, cavity bridges are formed or extinguished on the border of the surface distance between the cylinders of 0.4–0.5 mm. In the present experiments, the space between the Al₂O₃ cylinders was filled once with the molten steel because the two isodiametric Al₂O₃ cylinders between which the iron foil had been sandwiched were immersed in non-deoxidized molten steel.

**Fig. 4.** Relation between the diameter of glass cylinders and the maximum length of cavity bridges.

![Graph showing relation between diameter and maximum length of cavity bridges](image)

**Fig. 5.** State of cavity bridge formation in the section of the Al₂O₃ cylinders (d_{CY} = 8 mm) in the solidified steel ingot. (A) a = 0.7 mm between Al₂O₃ cylinders, (B) a = 0.5 mm between Al₂O₃ cylinders, (C) a = 0.4 mm between Al₂O₃ cylinders, (D) a = 0.3 mm between Al₂O₃ cylinders, (E) a = 0.1 mm between Al₂O₃ cylinders.

![Images of cavity bridge formation with varying distances](image)

**Fig. 6.** Influence of cylindrical diameter and surface distance between cylinders on cavity bridge formation between Al₂O₃ cylinders in molten steel.

![Graph showing influence of diameter and surface distance](image)
steel, which contains oxygen at high concentration and tends to wet Al₂O₃. After that, the molten steel deoxidized with Al became difficult to wet Al₂O₃, and was thus discharged from the space between the Al₂O₃ cylinders so that a cavity bridge was formed, as long as the surface distance between cylinders was relatively short. For this reason, the surface distance between cylinders of 0.4–0.5 mm obtained at the time of the formation and extinction of cavity bridges from the molten steel experiment represents the maximum length of cavity bridges in the case of approach between cylinders where molten steel is discharged and a cavity bridge is formed. In comparison between the maximum length of cavity bridges in the case of approach, the measurement value of 0.4–0.5 mm in the molten steel-Al₂O₃ system is approximately two times larger than 0.23 mm as the measurement value in the mercury-glass system.

4. Considerations

4.1. Influences of Wettability on Cavity Bridge Formation

No cavity bridges were observed between the copper cylinders in mercury; however they were formed between the glass cylinders in mercury as well as between the Al₂O₃ cylinders in molten steel. Whether cavity bridges are formed or not depends on the wettability of particles with a liquid metal. Copper has good wettability with mercury because the mercury-copper system involves reactions of amalgam formation. The contact angle of copper with mercury that is calculated using the surface tension of mercury on the basis of the measurement results of the initial wettability according to the meniscograph method by Nakae et al. results in 73° on an average. On the contrary, glass has difficulty in wettability with mercury, and the contact angle between the two is approximately 140°. Moreover, the contact angle of Al deoxidized molten steel with Al₂O₃ is 152.6°, as was measured in the previous report. On the basis of the results mentioned above, it is assumed that cavity bridges are formed between particles in liquid metal when the particles have the contact angle with the liquid metal of 90° or more and difficulty in wettability with liquid metal.

4.2. Interaction between Cylinders in Liquid Metal Involving Cavity Bridge Formation

4.2.1. Interaction Model between Cylinders in Liquid Metal

When two isodiametric cylinders having poor wettability with a liquid metal approach each other with only a narrow space between them, a cavity bridge is formed between the cylinders as shown in Fig. 7. In this case, the agglomeration force \( F_{AS} \) (N·m⁻¹) per unit length that occurred between the two isodiametric cylinders is expressed by Eq. (1), as the sum of the resulting force from the pressure difference between a cavity bridge and a liquid metal \( \Delta P_{LM} \) (Pa), and the surface tension of the liquid metal \( \sigma_{LM} \) (N·m⁻¹).

\[
F_{AS} = 2X_4 \cdot \Delta P_{LM} + 2\sigma_{LM} \tag{1}
\]

\( X_4 \) is the half-width of the neck of a cavity bridge (m). The relation presented by Eq. (2) is obtained from the geometrical conditions given in Fig. 7.

\[
X_4^2 + 2r_{CY} \cdot X_4 + (a^2/4 + a \cdot r_{CY} - 2r_{CY} \cdot r_{LM} \cdot \cos \theta_{LM}) = 0 \tag{2}
\]

When Eqs. (2) and (3) from which \( r_{CY} \) is eliminated are rearranged, Eq. (4) is obtained regarding \( X_4 \).

\[
X_4 = -\sigma_{LM}/\Delta P_{LM} + \left\{ (\sigma_{LM}/\Delta P_{LM})^2 - a^2/4 - a \cdot r_{CY} \right\}^{1/2} \tag{4}
\]

Equation (5) is obtained by giving \( X_4 \) from Eq. (4).

\[
X_4 = -\sigma_{LM}/\Delta P_{LM} + \left\{ (\sigma_{LM}/\Delta P_{LM})^2 - a^2/4 - a \cdot r_{CY} \right\}^{1/2} \tag{5}
\]

The agglomeration force between the two isodiametric cylinders in liquid metal can be calculated by substituting \( X_4 \) that is sought from Eq. (5) for Eq. (1). Moreover, once a peaks at \( D_{CB,Max}^2 \) (the maximum length of cavity bridges at the time of separation between cylinders) as the two isodiametric cylinders are slowly separated away from each other, the \( X_4 \) turns to zero and the cavity bridge is thus extinguished. \( D_{CB,Max}^2 \) is sought as in Eq. (7) by solving Eq. (6) rearranged after applying this condition to Eq. (4).

\[
D_{CB,Max}^2 = 4r_{CY} + 8r_{CY} \cdot \sigma_{LM} \cdot r_{CY} \cdot \cos \theta_{LM} / \Delta P_{LM} = 0 \tag{6}
\]

\[
D_{CB,Max}^2 = 4r_{CY} + 8r_{CY} \cdot \sigma_{LM} \cdot r_{CY} \cdot \cos \theta_{LM} / \Delta P_{LM} \tag{7}
\]

4.2.2. Application of the Interaction Model to Glass Cylinders in Mercury

The pressure difference between a cavity bridge and mercury is calculated by applying the experimental data in mercury at the time of separation of the glass cylinders in Fig. 4 to Eq. (6) and thus results in 1 670 Pa on average, where the surface tension of mercury and the contact angle of mercury with glass were put at 0.465 N·m⁻¹ and 140°, respectively. The calculated curvature radius of cavity bridges from Eq. (3) corresponding to this \( \Delta P_{LM} \).
4.2.3 Application of Interaction Model to Al2O3 Cylinders in Molten Steel

Equation (7) estimating the maximum length of cavity bridges when cylinders are separated away is applied to the results of the molten steel experiment in Fig. 6. Here, according to the results of the previous report,1) since the molten steel used in the experiments was the Al deoxidized molten steel whose composition resembles that of pure iron, the surface tension of Al deoxidized molten steel, the contact angle of that with Al2O3, and pressure difference between cavity bridges and molten steel were putted at 1.884 N·m\(^{-1}\), 152.6\(^{\circ}\), and 3.86 \(\times 10^3\) Pa, respectively. The calculated results of \(D_{\text{B, max}}\) obtained through the experiments is shown in the solid line in Fig. 6. The values representing 49% and 61% of the calculated \(D_{\text{B, max}}\) are plotted with chain lines in Fig. 6. It is found that 0.4–0.5 mm of \(D_{\text{B, max}}\) is shorter than the calculated \(D_{\text{B, max}}\) by Eq. (7) and equivalent to 49–61% of the calculated values. The mean value of the actually measured \(D_{\text{B, max}}\) approximately corresponds to 58% of the calculated \(D_{\text{B, max}}\). Hence, it is assumed that the maximum length of cavity bridges when particles approach each other decreases to 58% of that when they are separated from each other. It is assumed that this is because in order to form a cavity bridge when particles approach each other, it is necessary for a nucleus to be formed in the liquid metal between the particles, and also because by allowing the separated particles to approach excessively so that the distance between them reaches 58% of the maximum cavity bridge length when they are separated from each other, the necessary driving force for generating a cavity nucleus is acquired.

4.3. Agglomeration and Separation Mechanism of Al2O3 Inclusions in Molten Steel Involving Cavity Bridge Formation

4.3.1 Logical Extension to the Interaction Model between Spherical Al2O3 Inclusions in Molten Steel

To identify the interaction between Al2O3 inclusions in molten steel, an interaction model for two isospherical Al2O3 inclusions in molten steel is derived by extending the inter-particle surface distance a (m) and form a cavity bridge is expressed by Eq. (8) as the sum of the forces resulting from the pressure difference between a cavity bridge and molten steel \(\Delta P_{Fe}\) (Pa), and the surface tension of molten steel \(\sigma_{Fe}\) (N·m\(^{-1}\)), as in Eq. (1). \[ F_{A,S} = \pi \cdot R_a^2 \cdot \Delta P_{Fe} + 2\pi \cdot R_a \cdot \sigma_{Fe} \] \(R_a\) is the radius of a cavity (m). The geometrical conditions in Eq. (9) hold true in the case of two isospherical Al2O3 inclusions as in the case of cylinders. \[ R_3 + 2R_3 \cdot R_4 + (a^2 \cdot a / 4 + a \cdot r + 2R_3 \cdot r \cdot \cos \theta_{Al2O3-Fe}) = 0 \] \(R_3, R_4, \theta_{Al2O3-Fe}\), and \(r\) are the contact angle of molten steel with Al2O3 \((^\circ)\) and the radius of Al2O3 inclusions (m), respectively. According to the Laplace relation, the pressure difference between a cavity bridge formed between two isospherical Al2O3 inclusions and molten steel is expressed as in Eq. (10). \[ \Delta P_{Fe} = \sigma_{Fe} \left(1 / R_3 - 1 / R_4\right) \] When Eqs. (9) and (10) from which \(R_3\) is eliminated is rearranged for \(R_4\), Eq. (11) is obtained. \[ R_4^3 + A_1 \cdot R_4^2 + A_2 \cdot R_4 + A_3 = 0 \] Here, \(A_1, A_2, \) and \(A_3\) are each given as in Eqs. (12) through (14). \[ A_1 = 3\sigma_{Fe} / \Delta P_{Fe} \] \[ A_2 = a^2 / 4 + a \cdot r + 2\sigma_{Fe} \cdot r \cdot \cos \theta_{Al2O3-Fe} / \Delta P_{Fe} \] \[ A_3 = (a / 4 + r) \cdot a \cdot \sigma_{Fe} / \Delta P_{Fe} \] Although three solutions exist for the cubic equation of Eq. (11), the physical condition is satisfied that the radius of the neck of a cavity bridge becomes a positive real number under the state that at least two isospherical Al2O3 inclusions are in complete contact with each other, as shown in the previous report.1) Hence, \(R_4\) between two isospherical Al2O3 inclusions is given by Eq. (15).
Accordingly, the agglomeration force exerted on two iso- 
spherical Al₂O₃ inclusions with cavity bridge formation 
that are separated a certain interparticle surface distance 
away can be calculated by substituting the R₄ obtained from Eq. 
(15) for Eq. (8). Moreover, the further the two isospherical 
inclusions are separated away from each other, the smaller 
the R₄ becomes; however, unlike the case of two isodiamet- 
ric cylinder particles, the R₄ also becomes smaller due to 
the limiting condition in Eq. (10) (the ΔPₑ has a constant 
value). Therefore, the geometrical condition of Eq. (9) fails 
before the R₄ turns to 0, and cavity bridges are extinguished. 
Accordingly, when two isospherical inclusions are slowly 
separated away from each other, the maximum length of 
cavity bridges, Dₑmax, indicates the maximum interpar- 
ticle surface distance that can satisfy both the geometrical 
conditions and the Laplace relation, and that is achieved 
when the R₄ has reached the minimum positive value.

4.3.2. Logical Discussion Regarding the Agglomeration 
and Separation of Al₂O₃ Inclusions in Molten Steel 
Involving Cavity Bridge Formation 

Dₑmax was sought by trial and error allowing a to 
gradually increase so that the R₄ in Eq. (15) could reach the 
minimum positive value, with 1.884 N·m⁻¹, 152.6°, and 
3.86×10⁴ Pa each given as the surface tension of Al deoxi- 
dized molten steel, the contact angle of Al deoxidized molten 
steel with Al₂O₃, and the pressure difference between a 
cavity bridge and molten steel as in the molten steel experi- 
ments using Al₂O₃ cylinders. Moreover, on the basis of the 
experimental results regarding the cavity bridge formation 
between the glass cylinders in mercury, and between Al₂O₃ 
cylinders in molten steel, 58% of the Dₑmax was given as the maximum length of the cavity bridges at the time of 
approaching Al₂O₃ inclusions. Consequently, the maximum length of the cavity bridges when inclusions are 
separated away and when they approach each other results 
in Eqs. (19) and (20), respectively. Here, d is the diameter of Al₂O₃ inclusions (m), in a range from 0.1 to 100 μm in 
this calculation.

\[
\begin{align*}
Dₑmax &= 0.12d \quad \text{(19)} \\
Dₑmax &= 0.07d \quad \text{(20)}
\end{align*}
\]

The relation between the interparticle surface distance 
a·d⁻¹ and the agglomeration force between two isospherical 
Al₂O₃ inclusions due to cavity bridge force Fₑ(d·σfinancial)⁻¹ 
which is calculated according to Eqs. (8) and (15) is shown 
in Fig. 8 and, the states of the cavity bridge formation in the 
processes of approach and separation of Al₂O₃ inclusions 
in molten steel from (a) to (e) in Fig. 8 are schematically 
illustrated in Fig. 9. As can be seen from Fig. 8, only a slight

\[
R₄ = \left\{ -B₂/2 + \left( \frac{-B₁/108}{} \right)^{1/2} \right\}^{1/3} \]

\[
B₁ = -A₁²/3 + A₂ \quad \text{(16)}
\]

\[
B₂ = 2/27A₁² - A₁²/3 + A₁ \quad \text{(17)}
\]

\[
B₃ = -108\left( B₂²/4 + B₃²/27 \right) \quad \text{(18)}
\]
difference is observed between the agglomeration force due to the cavity bridge force of the inclusions of 0.1 μm (chain line) and 100 μm (solid line) in diameter in the interparticle surface distance of 0.08d or more; therefore, assuming that the diameter of Al2O3 inclusions lies within a range of 0.1 to 100 μm, the calculation result of the agglomeration force due to the cavity bridge force can be substantially organized into one relation represented by a solid line in Fig. 8 regardless of the diameter. This result enables the explanation associated with the formation and extinction of cavity bridges for the processes that Al2O3 inclusions in molten steel approach each other and agglomerate, and the agglomerated Al2O3 inclusions separate out, as described below. A cavity bridge is formed when the interparticle surface distance of Al2O3 inclusions in molten steel reaches 0.07d, as they approach each other (Figs. 8(a) and 9(a)), and an agglomeration force of 1.54d·σe occurs between the Al2O3 inclusions (Figs. 8(b) and 9(b)). The agglomeration force gradually becomes larger as the interparticle surface distance of Al2O3 inclusions becomes narrower, the Al2O3 inclusions come in complete contact with each other at the final stage, and the contact state is strongly maintained under an agglomeration force of 1.88d·σe (Figs. 8(c) and 9(c)). Conversely, once the Al2O3 inclusions being in contact with each other start separating out due to some external force, the agglomeration force slowly decreases as far as 0.12d surpassing an interparticle surface distance of 0.07d at which a sharp change in the agglomeration force occurred in an approaching process, and the agglomeration force reaches 0.92d·σe (Figs. 8(d) and 9(d)). If the interparticle surface distance further widens, the agglomeration force dramatically declines with extinction of cavity bridges (Figs. 8(e) and 9(e)). Thus, it is revealed that the agglomeration force due to the cavity bridge force reaches the maximum under the condition that the Al2O3 inclusions come in complete contact with each other, and even if the interparticle surface distances reach the maximums of 0.07d (when inclusions approach each other) and 0.12d (when inclusions are separated away) at which the cavity exists, the large agglomeration force of 1.54d·σe and 0.92d·σe due to a cavity bridge force interacts between the Al2O3 inclusions, respectively.

4.4. Dominant Agglomeration Force during Al2O3 Cluster Formation in Molten Steel

4.4.1. Agglomeration Force between Al2O3 Inclusions in Molten Steel due to van der Waals Force

The interaction when solid particles approach each other in liquid principally includes the repulsion force due to overlapping diffuse electric double layers and the dispersion force (van der Waals force). As there is no need to take the diffuse electric double layers in Al2O3 inclusions in molten steel principally includes the repulsion force due to diffuse double layers in Al2O3 inclusions in molten steel, the repulsion force due to the cavity bridge force is estimated by comparison with the capillary force to the cavity bridge force for the experiments on turbulent agglomeration in aqueous solutions. Here, H is the effective Hamaker constant (J) between Al2O3 particles in molten steel in such a way that the Hamaker constant of Al2O3 particles obtained through the experiments on turbulent agglomeration of the Al2O3 particles in aqueous solutions and that of iron at room temperature were converted into these values at 1 600°C, to which the association relation of the Hamaker constant was applied. Hence, using Eq. (21) allows us to estimate the agglomeration force due to the van der Waals force acting between two isospherical Al2O3 inclusions according to the surface distance between the Al2O3 inclusions.

4.4.2. Agglomeration Force between Al2O3 Inclusions on the Surface of Molten Steel due to Capillary Force

In previous section, it was indicated that when the solid particles having poor wettability with a liquid metal approach each other with a narrow space between them in the liquid metal, an agglomeration force results from the pressure difference between a cavity bridge and a liquid metal, and the surface tension of the liquid metal for the cavity bridge formation by the discharge of molten steel from the space between the solid particles as shown in Fig. 7. Meanwhile, when the solid particles which have poor wettability with the liquid metal approach each other at the surface of the liquid metal, a capillary force is induced in the horizontal direction from the surface tension of the liquid through the deformation of the liquid metal surface due to the wettability of the particles as shown in Fig. 10. As a result, an agglomeration force (attractive force) acts between the solid particles. In what follows, the agglomeration force due to the capillary force is evaluated for comparison with the agglomeration force due to the cavity bridge force.

Agglomeration force $F_{AC}$ (N) due to the capillary force between two isospherical Al2O3 inclusions on the surface of molten steel is approximately given in Eq. (22) from the analysis by Paunov et al.

$$F_{AC} = 2\pi \cdot \sigma_{Fe} \cdot Q^2 / (a + 2r), \quad r_c \ll a + 2r \ll q^{-1} \quad (22)$$

$$r_c = 1 / [r \cdot \sin \theta_{Al2O3, Fe} + 2Q \cdot \cos \theta_{Al2O3, Fe} \cdot g] \quad (23)$$

Here, $r_c$ is the radius of the contact line of an Al2O3 inclusion and the surface of molten steel (m) expressed by Eq. (23), Q is the capillary charge (m), q is the capillary constant (m$^{-1}$) defined by $\rho_{Fe} g / \sigma_{Fe}$, $\rho_{Fe}$ and $g$ are the density of molten steel being equal to 7 000 kg·m$^{-3}$, and the acceleration of gravity (m·s$^{-2}$), respectively. The ultimate value of the capillary charge $Q_c$ (m) where the interparticle surface distance of Al2O3 inclusions is infinite is given from the
analysis by Chan et al.\textsuperscript{9} in Eq. (24).

\[
Q_e = 1/6q^2r^3, \\
\left(2-4\rho_{Al_2O_3}/\rho_{Fe}+3\cos\theta_{Al_2O_3-Fe}-\cos^3\theta_{Al_2O_3-Fe}\right)...	ag{24}
\]

Here, \(\rho_{Al_2O_3}\) is the density of Al\(_2\)O\(_3\) inclusions and equal to 3 970 kg m\(^{-3}\). As the capillary charge of fine particles has a feeble dependency on the interparticle surface distance,\textsuperscript{9} \(Q\) can be approximately regarded to be equal to \(Q_e\). Accordingly, the agglomeration force due to capillary force can be calculated by substituting the value obtained as \(Q\) from Eq. (24) for Eq. (22). Furthermore, in this study, the application requirement of \(r_C\) < \(a+2r\) \(< q^{-1}\) for Eq. (22) is virtually satisfied, because the \(q^{-1}\) of Al deoxidized molten steel is 5 240 \(\mu\)m, and in addition, the \(r_C\) of the Al\(_2\)O\(_3\) inclusions with a radius of 500 \(\mu\)m or less can be approximated by \(\sin\theta_{Al_2O_3-Fe}/r = 0.46r\).

4.4.3. Comparison Study of the Agglomeration Force Acting between Al\(_2\)O\(_3\) Inclusions in Molten Steel

The effects of the interparticle surface distance on the various agglomeration forces acting between two isospherical Al\(_2\)O\(_3\) inclusions of 10 \(\mu\)m in diameter are shown in Fig. 11. Van der Waals force and cavity bridge force that is being studied are potential sources of the agglomeration force acting between the Al\(_2\)O\(_3\) inclusions in the molten steel in the present study. The agglomeration force due to the van der Waals force is a weak short-range force sharply decreasing with increasing the interparticle surface distance, as it weakens from 9.58 \(\times\) \(10^{-11}\) N to 9.58 \(\times\) \(10^{-15}\) N, as the interparticle surface distance widens from 10\(^{-2}\) \(\mu\)m to 1 \(\mu\)m. In contrast, the agglomeration force due to the cavity bridge force weakens only slightly from 3.50 \(\times\) \(10^{-5}\) N to 2.53 \(\times\) \(10^{-5}\) N, even if the interparticle surface distance is extended in a similar range, and it remains to be 1.73 \(\times\) \(10^{-5}\) N or larger unless cavity bridges are extinguished. Therefore, the agglomeration force due to the cavity bridge force can be said to be a far stronger long-range force compared with that due to the van der Waals force. On the other hand, an agglomeration force of 3.40 \(\times\) \(10^{-18}\) to 3.09 \(\times\) \(10^{-18}\) N derived from the capillary force at a similar interparticle surface distance acts between the Al\(_2\)O\(_3\) inclusions on the surface of molten steel. While the decrease in the agglomeration force due to the capillary force is comparatively small with the extension in the interparticle surface distance and presents a long-range force, the absolute value of that is smaller than the agglomeration force based on van der Waals force. The agglomeration force due to the capillary force obtained from the acceleration measurement regarding Al\(_2\)O\(_3\) inclusions of approximately 10 \(\mu\)m in diameter on the surface of Al deoxidized molten steel by Mizoguchi \textit{et al.}\textsuperscript{10} is marked with a small hatched area in Fig. 11. Their measurement values lie in 2.64 \(\times\) \(10^{-16}\) to 1.27 \(\times\) \(10^{-16}\) N in a range of 27 to 49 \(\mu\)m of the interparticle surface distance, and it represents 180 to 350 times the calculation value. This is because the Al\(_2\)O\(_3\) inclusions as the measuring object by Mizoguchi \textit{et al.} have a non-spherical complicate shape,\textsuperscript{10} and their actual measurement values resulted in the estimation by some two orders of magnitude larger than the agglomeration force acting between spherical Al\(_2\)O\(_3\) inclusions on the surface of molten steel due to the capillary force. Nevertheless, the agglomeration force due to the capillary force measured by them is only an order of magnitude larger (18 to 49 times) than that due to the van der Waals force; thus, it can be said to be a far smaller attractive force compared to that due to the cavity bridge force.

Assuming that the interparticle surface distance of Al\(_2\)O\(_3\) inclusions is equivalent to 5% of the inclusion diameter, the effects of the inclusion diameter on the various agglomeration forces between the two isospherical Al\(_2\)O\(_3\) inclusions were calculated, and these results are shown in Fig. 12. As the diameter of the two isospherical Al\(_2\)O\(_3\) inclusions in molten steel increases from \(10^{-1}\) \(\mu\)m to \(10^{3}\) \(\mu\)m, the agglomeration force due to the van der Waals force weakens from 3.83 \(\times\) \(10^{-12}\) N to 3.83 \(\times\) \(10^{-16}\) N; however, that due to the cavity bridge force is intensified from 3.12 \(\times\) \(10^{-7}\) N to

![Fig. 11. Effects of interparticle surface distance that affects various agglomeration forces acting between two isospherical Al\(_2\)O\(_3\) inclusions.](image1)

![Fig. 12. Effects of inclusion diameter that affects the various agglomeration forces between two isospherical Al\(_2\)O\(_3\) inclusions.](image2)
2.98 × 10^{-3} \text{ N}, and thus becomes stronger than that derived from the van der Waals force. Meanwhile, the agglomeration force acting between two isospherical Al_{2}O_{3} inclusions on the surface of the molten steel due to the capillary force become sharply intensified from 3.24 \times 10^{-28} \text{ N} to 3.24 \times 10^{-8} \text{ N} with the similar extension of the inclusion diameter, and is thus stronger than the agglomeration force due to the van der Waals force in an inclusion diameter equal to or over 50 \text{ μm}. Nevertheless, even if the inclusion diameter is extended to 10^{3} \text{ μm}, the agglomeration force due to the capillary force still remains to be approximately 10^{5} times smaller than that due to the cavity bridge force and extremely feeble.

As mentioned above, it is assumed that when the Al_{2}O_{3} inclusions formed by Al deoxidation approach each other with the molten steel flow, the agglomeration force due to the cavity bridge is mainly exerted, the Al_{2}O_{3} inclusions agglomerate and coalesce, and coarse Al_{2}O_{3} clusters are formed in molten steel because the agglomeration force due to the cavity bridge force acting in molten steel is far stronger than that derived from the van der Waals force in molten steel or the capillary force on the surface of molten steel despite the variation in the interparticle surface distance and the diameter of Al_{2}O_{3} inclusions.

5. Conclusions

The formation and extinction behavior of cavity bridges was experimentally evaluated through the approach and separation of the cylinder particles simulating inclusions in mercury as well as in molten steel. An interaction model of cylindrical particles due to cavity bridge force was developed on the basis of the experimental results, and logically expanded to that of spherical Al_{2}O_{3} particles close to the shape of actual inclusions. Using this interaction model, the processes of agglomeration and separation of Al_{2}O_{3} inclusions in molten steel were analyzed, and the agglomeration force due to cavity bridge force was discussed in comparison with different agglomeration forces derived from the van der Waals force in molten steel and the capillary force on the surface of molten steel. Consequently, the following conclusions concerning the agglomeration mechanism of Al_{2}O_{3} inclusions were drawn:

1. No cavity bridges are formed between copper cylinders in mercury; however, they are formed between glass cylinders in mercury as well as Al_{2}O_{3} cylinders in molten steel. Therefore, it is considered that cavity bridges are formed between particles in liquid metal when the particles have the contact angle of the particles with the liquid metal of 90° or more and difficulty in wettability with the liquid metal.

2. When two isospherical Al_{2}O_{3} inclusions are approaching each other in molten steel, a large agglomeration force of 1.54d \cdot \sigma_{Fe} (d: the diameter of Al_{2}O_{3} inclusions, and \sigma_{Fe}: surface tension of molten steel) is generated by the cavity bridge formation from the interparticle surface distance of 0.07d, and then the agglomeration force also gradually increases to reach the maximum value of 1.88d \cdot \sigma_{Fe} in complete contact state.

3. On the contrary, when two isospherical Al_{2}O_{3} inclusions in molten steel are separated from the contact state, the large agglomeration force of 0.92d \cdot \sigma_{Fe} and above is maintained until the cavity bridge extinction in the interparticle surface distance of 0.12d while the agglomeration force due to cavity bridge force gradually decrease from 1.88d \cdot \sigma_{Fe}.

4. It is assumed that Al_{2}O_{3} inclusions generated in molten steel by Al deoxidation agglomerate and coalesce because of the agglomeration force derived from the cavity bridge force, which is very strong compared with the van der Waals force in molten steel and the capillary force on the surface of molten steel, whereby coarse Al_{2}O_{3} clusters in molten steel are formed.

REFERENCES

1) K. Sasai: ISIJ Int., 54 (2014), 2780.
2) H. Nakas, H. Yamaura, T. Shinozaki, K. Yamamoto and Y. Osawa: J. Jpn. Inst. Met., 52 (1988), 428.
3) Mijika-na Gennso-no Kagaku, ed. by The Chemical Society of Japan, Baifukan, Tokyo, (1993), 147.
4) Kinzoku Dehta Bukku, ed. by J. Japan Inst. Met., Maruzen, Tokyo, (1974), 58.
5) S. Taniguchi and A. Kikuchi: Tetsu-to-Hagané, 78 (1992), 527.
6) S. Taniguchi, A. Kikuchi, T. Ise and N. Shoji: ISIJ Int., 36 (1996), S117.
7) H. Yin, H. Shibata, T. Emi and M. Suzuki: ISIJ Int., 37 (1997), 936.
8) V. N. Paunov, P. A. Kralchevsky, N. D. Denkov and K. Nagayama: J. Colloid Interface Sci., 157 (1993), 100.
9) D. Y. C. Chan, J. D. Henry, Jr. and L. R. White: J. Colloid Interface Sci., 79 (1981), 410.
10) S. Mizoguchi, K. Nakajima and Y. Nabeshima: J. Jpn. Weld. Soc., 70 (2001), 631.