Agglomeration Simulation of Chain-like Inclusions in Molten Steel
Based on Fractal Cluster-Cluster Agglomeration Model

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Based on the fractal cluster-cluster agglomeration (CCA) model, two-dimensional simulations of cluster aggregation process have been carried out by varying initial particle number, particle size and sticking probability between clusters. Particle size and amount of seed particles have small effects on the shape of clusters. Actual alumina clusters observed in tundish show the chain-like appearance and are very similar to the simulated agglomerated clusters with high sticking probability. The agglomeration process of alumina chain-like clusters around immersion nozzle in the tundish and RH process are plausibly controlled by the random collision process with high sticking probability values. [doi:10.2320/matertrans.MRA2007037]

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1. Introduction

Non-metallic inclusions in the steelmaking process have a very important and critical role in the production of clean steel.¹) Thus, many studies of the kinetics and thermodynamics of the deoxidation process have been carried out to understand the details of the inclusion formation mechanism.

Most theoretical studies of the inclusions assume that they are spherical particles, and that they agglomerate to form spherical particles with a larger diameter. However, in the case of the aluminum deoxidation process, small alumina particles assemble in relatively loose and irregular chain-like clusters,²–⁴) which can cause defects in cold-rolled sheets and other steel materials if some of these inclusions remain at the surface or the near-surface zone of the continuous cast slabs. The structure of the typical alumina inclusions is shown in Fig. 1. The floating behavior of cluster-shaped inclusions can be very different from that of the spherical inclusions as expressed by Stokes’ law. Thus, it is very critical to understand the shape formation mechanism of the inclusions in order to improve the qualities of steel. However, most previous investigations on inclusions have mainly focused on the macroscopic average size distribution and the methods used to decrease the inclusions. Therefore, the shape forming mechanism of chain-like cluster inclusions has not been established.

The growth of inclusions involves a fluctuation or randomness process, which plays an essential role in shaping the final morphology of the inclusions. The main origin of the randomness of inclusion growth is the fluctuation in the flow and the concentrations of elements in a vessel. Thus, the formation mechanism of inclusions should be treated on the basis of probability theory. The model based on probability theories such as the population balance model¹⁵) has been applied for analyzing inclusion agglomeration, and this model has made it possible to evaluate the macroscopic parameter of the agglomeration constant; however, it is not suitable for the evaluation of the shape forming mechanism.

It is noted that fractal theory⁶–⁸) is a very powerful tool to analyze random shapes and structures. During the last few decades, the field of fractal growth phenomena has continued to develop rapidly, and a large number of important new studies have been published. It has been found through these studies that the fractal theory is quite useful to study the geometrical aspects of the particle agglomeration process. Fractal agglomeration models such as the diffusion-limited agglomeration (DLA)⁹) and the cluster-cluster agglomeration (CCA) models¹²,¹³) have attracted much attention in the field of material science, and most studies have concentrated on their geometrical properties.

There have been many reports that alumina inclusions in steel have an open branching structure. Such objects can be described in terms of fractal geometry. In this study, the formation mechanism of alumina chain-like inclusions in steel has been investigated using fractal agglomeration simulation.
2. Fractal Model and Its Simulations

2.1 Cluster-cluster agglomeration

The agglomeration of microscopic particles diffusing in a fluid medium occurs frequently. If the density of the initially randomly distributed particles is greater than zero, the probability of the particles colliding and sticking together will be finite. In such systems, it is expected that the resulting two-particle aggregate may diffuse further and form larger clusters by joining other aggregates. As a result, the mean cluster size increases with time. In many cases, the force between two particles is of a short range, and it is strong enough to bind the particles irreversibly when they come into contact with each other. This process is called cluster-cluster agglomeration (CCA). In this process, all clusters are equivalent with regard to the conditions of their motion, i.e., there is no particular seed particle whose behavior differs from the case of the well-known diffusion-limited agglomeration (DLA) model. It is supposed that the CCA process corresponds more closely to the physical situation in a system of aggregating particles.

The rules of agglomeration in the CCA model are quite simple. A typical two-dimensional simulation begins by randomly occupying a small fraction of the sites on a square lattice to represent the particles. Each particle is allowed to move at random (diffuse) until it arrives at a site adjacent to another particle, whereupon it sticks to that particle, forming a two-particle cluster. At each time step, a particle or cluster is selected randomly and is moved by one lattice unit in a randomly chosen direction. If a particle or cluster exits or crosses over the boundary, it is allowed to enter a square lattice from the other side of the boundary. Two clusters or a particle and a cluster can stick when they touch each other. This process is then iterated repeatedly. In this way, large clusters can be generated. The agglomeration process is schematically shown in Fig. 2.

2.2 Fractal dimension

In the present study, two-dimensional simulation of the inclusion agglomeration has been carried out instead of three-dimensional simulation. The actual inclusion formation occurs in three-dimensional space. The large-scale simulation of the CCA model in two- and three-dimensional space proved that the values of the fractal dimension of the CCA cluster, \( D \), are 1.43 (two dimensions) and 1.75 (three dimensions). It is the projection or cross section of fractal structures that is of interest to us and can be experimentally studied. In general, the projection of a \( D (< d-m) \) dimensional fractal in \( d \) dimensional space onto a \( (d-m) \) dimensional surface \( (D_S) \) results in a structure with the same fractal dimension, namely, \( D_S = D \). On the other hand, the removal of a \( (d-m) \) dimensional slice (cross section) of a \( D \) dimensional fractal embedded into \( d \) dimensional space \( (D_S) \) usually leads to a \( (D-m) \) dimensional object, or \( D_S = D - m \). Thus, the expected fractal dimension of the projected three-dimensional CCA cluster onto a two-dimensional plane is 1.75 and that of the sliced cross section is 0.75. A fractal dimension of less than 2 means that the structure is very sparse such that many parts of the three-dimensional structure projected onto the two-dimensional plane do not overlap on the two-dimensional plane. Consequently, these parts will retain a qualitatively similar structure. Thus, as a first approximation, we can assume that the simulated structure obtained by two-dimensional CCA simulation reflects the cluster structure of the observed cross-sectional or projected cluster inclusions.

2.3 Simulation method

The actual simulations are performed on a periodic square lattice such that \( S = L^2 \) \((L = 10000)\) with initially \( N_0 \) randomly distributed particles. At the start of the simulation, a fraction \( \rho \) \((\rho = N_0/S)\) of the lattice sites is selected at random and occupied (avoiding multiple occupancy). The sites occupied at the nearest-neighbor positions are considered to belong to the same cluster. The clusters (including single-particle clusters) are rigid and randomly move one unit lattice spacing at a time, but they do not rotate. The clusters are selected at random and moved with a probability proportional to their “mobility” by a unit lattice spacing along one of four equally probable directions \((\pm x, \pm y)\) also picked at random). If a cluster comes into contact with other clusters (via the nearest-neighbor occupancy), the contacting clusters will be merged to form a single cluster. In this manner, the clusters grow larger and larger until only one large cluster remains. The calculation was continued until a particular number of clusters was formed. For a Brownian particle, its mobility is generally inversely proportional to the size of the cluster. However, in the present study, the cluster mobility was assumed to be the same for all the clusters since it is known that the mobility has a very small effect on the cluster structure in the agglomeration simulation. In this manner, the clusters grow larger and larger until only one large cluster remains. The calculation was continued until a particular number of clusters was formed. For a Brownian particle, its mobility is generally inversely proportional to the size of the cluster. However, in the present study, the cluster mobility was assumed to be the same for all the clusters since it is known that the mobility has a very small effect on the cluster structure in the agglomeration simulation. The simulations have been carried out by varying the sticking probability. For a sticking probability of 1, the contacting clusters always merge together to form a single cluster when they come into contact with each other. This means that the reaction (sticking) time \( t_R \) of a Brownian particle at the cluster surface is significantly short as compared to the diffusion time \( t_D \) for reaching the cluster surface \( (t_R/t_D \ll 1) \). This process can be considered as...
diffusion limited. When the sticking probability is very low or nearly zero, the Brownian particle moves around the surface for a considerably long time ($t_R / t_D \gg 1$). In this case, $t_R$ is much larger than $t_D$, and the process is reaction limited. This type of agglomeration can be typically simulated by the Eden model. The actual inclusion agglomeration can occur in between these extreme cases. Thus, the formed cluster structure can be possibly characterized by the ratio of $t_R / t_D$ in the system.

A "sticking probability" is introduced to evaluate the effect of $t_R / t_D$ on the structure of clusters. The sticking probability governs the possibility of the permanent agglomeration of clusters or particles or subsequent movement back into free space. The sticking probability of $p (p < 1)$ means that a cluster is allowed to stick to another cluster for only $p$ times even though the particle approaches the nearest-neighbor sites of other clusters during its movement. In other words, clusters can stick together only once after $1/p$ trials. The large sticking probability means a small $t_R / t_D$ value or the dominant influence of the diffusion process.

As additional parameters in the simulation, "initial particle size" and "initial particle number" were introduced. The seed particle is initially square shaped and its side length is $n$ ($n$ is any integer) lattice unit. In this study, the particle size was represented by the side length of the square particle. By introducing the sticking probability and particle size considerations, the arbitrary length in the simulation can be roughly correlated to the actual dimension when the detectable size of the inclusions is known since their dimensions are related to each other through the lattice unit length. The simulation was carried out by changing the number of initial seed particles, initial seed particle size, and sticking probability.

3. Results and Discussion

3.1 Effect of sticking probability

In Fig. 3, the evolution of the clusters is shown for a system of size $L = 10000$ with initially 1000 particles and the initial size is 50 lattice units. The iteration numbers for the calculations are 50 (Fig. 3 (a)), 300 (Fig. 3 (b)), and 3000 (Fig. 3 (c)). The sticking probability is set to 1 lattice unit so that the simulation reflects a diffusion-limited process. It is clearly shown that the cluster grows for the ramified structure with each calculation step. The final stage of clusters has a chain-like appearance.

Figure 4 shows the final stage of simulations carried out using sticking probabilities of 1, 1/3, and 1/10 on a 10000x10000 lattice. The initial number of particles is 1000, and the particle size length is 50 lattice units. The calculation was carried out until 10 clusters were formed. Some clusters are too small to be identified in the figure. From the calculated results, it is apparent that the sticking probability strongly influences the shape of the cluster. That is, with an increase in the sticking probability, the formed structure shows a chain-like appearance or more open branching structures.

3.2 Effect of initial conditions of particle distribution

Besides the sticking probability, the other factors such as cluster size, spatial randomness of cluster distribution, mean distance between clusters and cluster fraction may affect the shape of the clusters. The spatial randomness of clusters, mean distance between clusters and cluster fraction are closely related to the initial number of particles and the initial
particle size. Thus, to evaluate the effect of these initial conditions of particle distribution on the shape of clusters, the simulation with varying the number of particles and the initial particle size has been carried out.

From the simulated results, it is found that the variation of the initial number of particles and the initial particle size did not show the dominant effects on the shape of clusters. Namely, the chain-like appearance is mainly controlled by the sticking probability. In other words, the most relevant factor affecting the geometrical properties of the clusters can be the sticking probability. The chain-like appearance is due to the shade effect of the cluster structure itself. That is, the probability of adding a particle to the cluster in the diffusion-limited growth is determined by the structure of the whole cluster. Consequently, these processes are truly nonlocal.

3.3 Alumina cluster agglomeration in the tundish

The comparison between the simulated clusters with a sticking probability of 1 (Fig. 3 (c) and Fig. 4 (a)) and the actual alumina cluster shown in Fig. 1 reveals that they are apparently very similar to each other. The main advantage of the fractal models is that they provide a possibility to determine the most relevant factors affecting the geometrical properties of objects developing in a given type of growth phenomena. The assumptions of the present simulation are quite simple: (1) the total number of particles is constant and (2) the particles move randomly.

Actual alumina chain-like clusters were mainly observed around the immersion nozzle area in the tundish and RH process. In these zones, the molten steel flow was severely agitated, and many eddies and wakes were produced. It is expected that the alumina cluster formed in the tundish was involved in the strong turbulent flow and could stay around the immersion nozzle in the tundish and RH vessel for a considerably long time. Thus, assumptions (1) and (2) are reasonably satisfied in the actual tundish and RH process.

For developing chain-like structures in the simulation, a high sticking probability is also required in the agglomeration process. Yin et al.\(^{15}\) observed the alumina agglomeration process at the molten steel surface by using a confocal laser microscope and confirmed that the alumina clusters stick together immediately when they come close together and do not separate again. Thus, a high sticking probability value in the alumina cluster agglomeration process is also expected in the tundish and RH vessel. That is, the actual agglomeration process of alumina clusters around the immersion nozzle area in the tundish and RH process satisfy all the conditions required to form chain-like structures in the fractal CCA model simulation. Therefore, the agglomeration mechanism in the CCA model should be applicable to the actual agglomeration process of alumina clusters around the immersion nozzle area in the tundish and RH process. Thus, it can be concluded that the agglomeration process of alumina chain-like inclusions is controlled by the random collision process with high sticking probability values.

4. Concluding Remarks

In the present work, the formation mechanism of chain-like alumina inclusions around the immersion nozzle area in the tundish and RH process is investigated by applying fractal cluster-cluster agglomeration (CCA) model. Two-dimensional simulations by varying various particle sizes, particle number and sticking probability between clusters have been carried out.

The most relevant factor affecting the shape of clusters is found to be the sticking probability. With an increase in the sticking probability, formed structure shows a chain-like appearance or more open branching structures. Namely, the formation of chain-like alumina clusters is easily explained if they have abilities to stick together easily when they come close together without introducing any complex mechanisms. The high sticking probability between alumina clusters, however, has not yet thoroughly explained. This warrants future work on the study of sticking mechanism between alumina inclusions.

REFERENCES