Modeling of Growth and Impingement of Spherical Grains

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A model was developed to simulate the growth and impingement of spherical grains, which are frequently encountered in the solidification, recrystallization and other processes in association with phase transformations. The initial grain was assumed as spherically approximated 128 faces polyhedron. Various cases were investigated under different conditions of final number of grains, nucleation and spatial distribution. The validity of the Kolmogorov, Johnson-Mehl and Avrami (KJMA) equation was examined for various cases.

The results proved that KJMA equations are valid for the real volume and the effective surface area (effective for later growth) calculations under the conditions of site saturation/constant growth rate and the conditions of constant nucleation rate/constant growth rate. When the grain shape is not maintained same throughout the process, the results show that the effective surface area deviate largely from the KJMA equation even though the KJMA equation is approximately valid to calculate the real volume. Finally, when the spatial distribution of grains is non-random, clustered or ordered, the KJMA equation is not valid and the deviation from it is large especially at the end of transformation both for the real volume and for the effective surface. Even when the KJMA equation is not valid for the effective surface area, Rath's empirical equation is effectively applicable for it, though the physical meaning of the parameters used in it is not clear.

(Received April 1, 1996)

Keywords: grain growth, grain impingement, random distribution, spatial distribution, clustered structure, ordered structure, Kolmogorov/Johnson-Mehl/Avrami equation

I. Introduction

In the analysis or the computer modeling of the processes of solidification\(^{(19)}\), recrystallization\(^{(19)}\) or other processes in association with phase transformations, the following Kolmogorov\(^{(6)}\), Johnson-Mehl\(^{(5)}\) and Avrami\(^{(6)}\) (KJMA) equation\(^{(7)}\) is frequently used:

\[
V = 1 - \exp (-V_\infty)
\]

where \(V_\infty\) is the volume fraction of the transformed phase in the absence of impingement (the extended volume fraction) and \(V\) is the volume fraction of the transformed phase (real volume fraction). When the grains impinge on each other during growth, the impinged interface between the grains will not be effective for the hereafter growth process. The surface area which participates in the growth \(S\) (the effective surface area) can be derived from eq. (1) as follows\(^{(40-42)}\):

\[
S = (1 - V) S_\infty
\]

where \(S_\infty\) is the surface area of the grains in the absence of impingement of the grains (the extended surface area).

On the other hand, Rath\(^{(9)}\) suggested an empirical expression of \(S\) as a function of \(V\),

\[
S = k V^q (1 - V)^p
\]

where \(k\), \(q\) and \(p\) are constants. Rath\(^{(9)}\) proposed that the fading course of the matrix is not the same course of the growth of grains and hence \(q\) and \(p\) are not equal and their values are in the range between 0 and 1. Equation (3) is actually the general form of the empirical equation by Speich\(^{(10)}\) where \(q = 1\) and \(p = 1\) and the equation by Cahn\(^{(11)}\) where \(q = 2/3\) and \(p = 2/3\).

In the simulation of eutectic solidification of spheroidal graphite cast iron, the shape of the austenite shell which surrounds the spheroidal graphite is often presumed as a sphere\(^{(12)}\). The volumetric solidification rate is the result of the effective surface area \(S\) multiplied by the solidification rate. Therefore, it is very important to know how the effective surface area should be calculated and which equation can be used for this purpose.

In the present study, the impingement process of spherical grains is simulated by computer experiments and the relation between the eqs. (1), (2) and (3) and their validity are investigated.

II. Simulation Method

As shown in Fig. 1, the basic grain or nucleus is defined as a polyhedron with 128 triangular faces and 66 vertices which are the growth points. The numbers of faces and vertices are decided in consideration of two factors: computation time and surface evolution accuracy. Too many vertices elongate the computation time and few vertices make the surface of the polyhedron rough which brings large errors in the calculation of growth and impingement.

A number of \(10^5\) sites is uniformly positioned in a cube of length 1.04 unit (experimental domain, 1.04\(^3\)) It should be noticed that the length used here can have any unit, such as m or \(\mu\)m and the unit will be called as
"length unit" throughout this paper). The sites are numbered serially from 1 to $10^3$. After that, the nucleation sites are chosen among all sites using random function in consideration of two conditions: only one nucleus is allowed to form at one site and no intersection occurs between the nuclei at the initial position. After being positioned at their nucleation sites the nuclei are continuously grown. The growth rate is linear, radial and the unit of growth rate is not SI unit but it is simply length unit/ ts, where ts is a time step of the calculation cycle, taking no account of the ambient conditions which control the growth in practice such as concentration of elements and temperature. Figure 2(a) shows an example of 50 grains after being randomly located and then grown.

In order to investigate the above-mentioned three equations, the real volume $V$, the extended volume $V_{ex}$, the real surface area $S$ and the extended surface area $S_{ex}$ of the grains which are included within a cube of 1 length unit (observation domain, 1', OD) are calculated at each time step. The grains which intersect with the wall of the OD are treated as shown in Fig. 2(b), where the part which grows out of the OD is removed and only the part included within the OD is considered in calculating the areas and the volumes.

In the present model, it is necessary to stop the growth point which impinges with other grain. This process is explained in Fig. 3(a). After growing by a definite amount during one time step, the growth point 0 of grain A impinges at the position 0' on the face 1'-2'-3' (grown from 1-2-3) of grain B and the point 0 is stopped at the position 0'. In order that the growth points 1', 2' and 3' which make the face do not lose their freedom of growth, a new growth point having the same position of 0' is created but stopped in grain B and new faces 0'-1'-3', 0'-1'-2' and 0'-2'-3' are defined. For example, the result of 4 impinging grains are shown in Fig. 3(b), which were created by these procedures for impingement.

The volume of each grain is calculated as the sum of the volumes of the tetrahedrons which result from connecting the center of weight of the grain to each of the three vertexes of each triangular face on the grain surface. The effective surface area is calculated as the sum of the areas of the triangular faces which has at least one moving growth point, i.e. the triangular face which has all its vertexes stopped by impingement will not be included in the effective surface area calculation. As shown in Fig. 4(a), it was made clear that the near-spherical shape cannot be preserved until the next growth point stops after two grains A and B stop at the growth point 1 or 2, and hence an error occurs during the calculations of the real volume and the effective surface area. Therefore, the error has to be compensated specially for the calculation of the effective surface area. The areas of the faces which include a part of the grain boundary edge a-b of grains A and B are calculated as shown in Fig. 4(b) by the hatched areas, in which the altitude of the triangular face is assumed to be a half of real one.

III. Simulation Results and Discussions

In the present work many cases were treated with different calculation conditions, with various numbers of grain, nucleation process and spatial grain distribution. These cases and their conditions are listed in Table 1. Here the minimum number used is 250 grains. It was made clear that KJMA equation was not satisfactory when the number of grains was less than 200 although the calculation conditions satisfied the KJMA equation with
250 grains. One reason is that the randomness condition was not properly achieved when the number was less than 200 grains. Furthermore, the size of one grain was too big at high volume fractions comparing to the case when the number was more than 250 at the same volume fraction and hence the surface of each individual grain became rough which brought errors in calculation of volume and surface area.

1. Site saturation and constant growth rate

Figure 5 shows the results of the case where the nucleusation is instantaneously finished at the time 0 and the growth rate was constant ($G=0.01 \text{ length unit/}$.t). It is clear that the KJMA equation in the form of eq. (1) and eq. (2) is valid even with a comparatively low number of grains, case A, as shown in Fig. 5(a) and Fig. 5(b) respectively. The calculated real volumes are slightly smaller than the theoretical values (Fig. 5(a)) because no correction was carried out to compensate this error at the grain boundary.

Now let the relation between the extended surface area $S_{ex}$ and the extended volume fraction $V_{ex}$ be represented as

$$S_{ex} = c \cdot V_{ex}^q$$

where $c$ and $q$ are constants and $q=2/3$ for spherical grains. From the eqs. (1), (2) and (4) the following relation can be derived(2):
Fig. 5 The calculated results under the conditions of site saturation and constant growth rate. (a) Plot of eq. (1), (b) plot of eq. (2) and (c) the solid lines, calculated results and the dashed lines, eq. (5).

Fig. 6 The calculated results under the conditions of constant nucleation rate and constant growth rate. (a) Plot eq. (1), (b) plot of eq. (2) and (c) the solid lines, calculated results and the dashed lines, eq. (5).

\[ S = c \cdot (1 - V) \cdot \left( \ln \frac{1}{1 - V} \right)^q \]  
\[ S = c \cdot V^q \cdot (1 - V)^{1 - (q/2)} \]

This equation can be replaced by the following approximation^{12} which is similar to the relation suggested by Rath^9, eq. (3).

Figure 5(c) represents the relation between the effective surface area \( S \) and the real volume fraction \( V \). The dotted lines in Fig. 5(c) represents eq. (5), where the constants \( c \) and \( q \) are determined from the relation between \( S_{\text{ex}} \) and \( V_{\text{ex}} \). (Table 2: A, B, C, D). When the number of grains increased, the calculated values of \( S \) (solid lines) tended to be higher than those calculated from eq. (5) (dotted lines), as can be observed in Fig. 5(c). The reason of this may be that the correction of the effective area calculation at grain boundary edge was not perfect. However, it can be said that calculated results agreed well with eq. (5), apart from this slight disagreement. Now, if eq. (5) is differentiated with respect to \( V \), the maximum of \( S - V \) plot is given when

\[ V_{S=\text{max}} = 1 - \exp (-q) \]  
when \( q = 2/3 \).

2. Constant nucleation rate and constant growth rate

The "constant nucleation rate" can be interpreted into two meanings. First, the nucleation rate within a unit volume is constant irrelevant to the amount of phase that have already transformed, case E. Second, the matrix has the character to make the nucleation rate constant but the nucleation rate is practically proportional to the non-transformed volume, case F. The nucleation rate of both cases are given in eqs. (8) and (9), respectively,

\[ \dot{N} = \dot{N}_c \]  
\[ \dot{N} = (1 - V) \cdot \dot{N}_c \]

where \( \dot{N}_c \) is the constant value of nucleation rate. In this study, simulation was carried out with \( \dot{N}_c = 30N/\text{ts} \) in eq. (8), \( \dot{N}_c = 20N/\text{ts} \) in eq. (9) and \( G = 0.01 \text{ length unit}/\text{ts} \) (Table 1: E, F). Practically, the nucleation occurs in the non-transformed volume as done in the present simulation. In spite of the fact that the KJMA equation was derived originally in consideration to the "phantom nuclei" which nucleate in the already transformed volume, eqs. (1) and (2) are still valid as shown in Fig. 6(a) and (b). On the other hand, the maximum of the plot \( S - V \) in Fig. 6(c) tends to move towards little higher values of \( V \) com-
paring to Fig. 5(c). For both cases E and F the values of $c$ and $q$ are used to plot the dotted lines in Fig. 6(c). The values of $q$ for both cases are almost equal as listed in Table 2 (the theoretical value of $q$ for spherical grains is 0.75) and both curves do not show much difference. The reason of this may be that the effect of the term $(1 - V)$ in eq. (9) on $N$ is small at the initial stage of transformation and that the effects of the surface area and volume of the grains that nucleated from the middle to the end of transformation on the whole process are also small. Analytically the maximum of the plot $S - V$ occurs at $V_S = 0.528$ for $q = 3/4$ from eq. (7), which agrees with the results of simulation.

3. The effect of grain shape change

It becomes clear from the above results that both eqs. (1) and (2) are valid, if the shape of the grains can be maintained unchanged during the growth process and $S$ can be calculated from either eq. (5) or eq. (6)$^{12}$, if the relation between $S_{\text{cr}}$ and $V_{\text{cr}}$ is known. Because the curvature near by the grain boundary edge is determined by the balance of the interfacial energy between the matrix and the transformed phase$^{13}$ and then the grain shape is changed near by the grain boundary during grain impingement, it is important to know how the relation between eqs. (1) and (2) or eqs. (5) and (6) changes by its effect. The calculated results without correction in calculation of $S$ of the segments which include the grain boundary edge (Fig. 4(a) I, 2) must show quantitatively the same behavior as the case when $S$ is calculated greater due to the interfacial energy balance at the grain boundary edge than that without change of grain shape.

Figures 7(a), (b) and (c) show the calculated results in the cases G, H, I and J under the conditions of site saturation and constant growth rate $G = 0.01$ length unit/$s$ and no correction is carried out in area calculation. Under these conditions, Fig. 7(a) shows that KJMA equation is still valid even though the real volume $V$ is expected to be smaller than eq. (1) due to shape change. On the other hand, it is also clear that the shape changes so that the surface area is calculated larger than when there is no shape change and hence $S$ is larger than eq. (2) (Fig. 7(b)). From this it can be suggested that especially the effective surface area is largely affected by the shape changes with keeping the balance of the interface energy. Like this, eq. (2) will not be valid as long as the shape change of the transformed phase is not reflected on the calculation of $S_{\text{cr}}$ in eq. (4).

From Fig. 7(c), the shift of the maximum of $S - V$ plot towards the right is also clear, which behavior is well approximated by eq. (3) with the parameters shown in Table 2: G, H, I, J. This behavior agrees well with the results predicted analytically by Gokhale et al.$^{14}$ on the effect of the transformed phase curvature near grain boundary edge.

Although the case is not simulated in this study, it is expected that the plots may skew to lower in Fig. 7(b) and to the left in Fig. 7(c), when the grain shape changes keeping interfacial energy balance so that effective surface area $S$ is calculated smaller than when there is no shape change (Fig. 4(a) 1° and 2°).

4. The effect of spatial distribution of nucleation site

Until the last section, the nucleation sites are randomly chosen within the experimental domain. Equation (2) can

<table>
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<tr>
<th>Case</th>
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<th>$q$</th>
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</table>

![Fig. 7](image-url) The calculated results under the conditions of site saturation and constant growth rate and no correction of the effective surface area. (a) Plot of eq. (1), (b) plot of eq. (2) and (c) the solid lines, calculated results and the dashed lines, eq. (3).
be derived on the assumption of a random distribution. For the non-random distribution, the following empirical
equation is suggested\(^{(10)}\):
\[
S = (1 - V)^i \cdot S_0
\]
(10)
where \(i\) is a constant whose value depends on the deviation of the spatial distribution of nuclei from randomness. When the grains tend to gather locally (clustering), \(i > 1\) and when the grains tend to arrange regularly (ordering), \(i < 1\).

To investigate the effect of spatial distribution of the nucleation site, the following four cases were studied.

(a) Strong clustering (case K): The observation domain was divided into 8 equal cubic units (2 \(\times\) 2 \(\times\) 2), then 400 grains were randomly distributed in 4 of these cubes, so that 100 grains in each unit cube and the rest 4 cubes were left empty.

(b) Weak clustering (case L): The same procedures as in the case of K but the only the difference is that 20 grains were randomly distributed in each of the 4 empty unit cubes.

(c) Strong ordering (case M): The observation domain was divided into 512 equal cubic units (8 \(\times\) 8 \(\times\) 8), then one grain was allowed to nucleate at the center of each unit cube.

(d) Weak ordering (case N): The same procedures as case M, but the grain was allowed to nucleate randomly in each unit cube.

From Fig. 8(a) it is understood that, eq. (1) is still valid at low volume fractions (in the present conditions \(V \leq 0.2\)) even though the spatial distribution of grains is far from random. However, the deviation from KJMA equation becomes large at higher volume fractions. Because of this behavior, it can be predicted that the volumetric transformation rate becomes slower in the clustering case and faster in the ordering case compared with the full random case especially in the range from the middle to the end of the transformation. The same can be also understood from Fig. 8(b). In Fig. 8(b), the clustering cases result in the smaller value of the effective surface area and the ordering cases resulted in the larger value of \(S\) compared with the full random cases (eq. (2)). These results qualitatively agree with those which can be predicted from eq. (10) but do not quantitatively agree with those from eq. (10) especially when \(i\) is smaller than 1.

The strong dependency of \(S\) on spatial distribution can be noticed also from Fig. 8(c). The maximum of \(S - V\) plot in clustering case tends to shift towards smaller values of \(V\) while it shifts towards higher values of \(V\) in ordering case. This is because the impingement occurs earlier in clustering case and the impingement occurs later in ordering case than in case of random distribution.

As shown in Table 2: K, L, M, and N, the values of \(p\) and \(q\) which were used to fit eq. (3) do not show the relation \(p = 1 - (q/2)\). The spatial distribution of grains has strong influence specially on the value of \(p\). Comparing with fully random case where \(p = 2/3\), \(p\) takes larger values in clustering case and smaller values in ordering case. Here \(p\) is not necessarily in the range suggested by Rath\(^{(9)}\) but \(p\) can be larger than unity in some cases.

In the case of previous Section III.3 where the shape of the grains change during growth process and in the case of the present section where the nucleation site distribution is non-random, eq. (2) is not valid but the value of \(S\) can be approximated by eq. (3). However, the effect of the shape change and the spatial distribution on the quantity of the parameters \(p\) and \(q\) and the physical meaning of them in eq. (3) is still ambiguous.

## IV. Conclusions

A computer model using a spherically approximated 128 polyhedron was developed to simulate the grain impingement process. The accomplished results can be summarized as follows.

1. The calculated results proved that the relation between the real volume fraction \(V\) and the extended volume fraction \(V_a\) can be presented by the KJMA eq. (1) under the conditions of site saturation/constant growth rate and the conditions of constant nucleation...
rate/constant growth rate.

(2) Also under the same conditions of (1), it was confirmed that eq. (2) can be used to represent the relation between the effective surface area $S$ and the extended surface area $S_{eq}$. Likewise, it was confirmed that eqs. (5) and (6) as special cases of eq. (3) are valid.

(3) In the cases when the grain shape changes due to grain impingement, it was expected that eq. (2) is not valid to calculate $S$ even though eq. (1) was approximately valid to calculate $V$. However, eq. (3) can be used as a good approximation for $S$.

(4) In the cases where the spatial distribution is non-random, clustered or ordered, eq. (1) is not valid, since the deviation from eq. (1) is large especially at the end of the phase transformation. Also the calculated results of $S$ show a large deviation from eq. (2) but eq. (3) can be used instead of it.

(5) In both cases of the shape change due to the impingement during the growth process and the non-random distribution, the value of $S$ can be approximated by eq. (3). However, the effect of them on the parameters $p$ and $q$ is only qualitatively understood and the quantitative physical meaning of these parameters is still unclear.

Acknowledgment

The authors wish to thank Mr. K. Kozasa for his assistance in performing the calculations and Japan Society for Promotion of Science for the financial support.

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