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The contribution of the nucleation process to grain formation in calculating solidification microstructure by CA–DFD

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Abstract

To predict solidification macrostructures, the direct finite difference (DFD) method for heat transfer calculation was coupled with cellular automaton (CA). In the CA–DFD, the nucleation process as the initial condition of calculations was determined according to Gaussian distribution or being instantaneous. In the case of Gaussian distribution, there are six nucleation-related parameters. In this work, the inspection of relationships of preset nuclei to grain, preset nuclei to born nuclei, and born nuclei to grain was carried out by comparison of each ratio. However, these relationships are only valid under the same solidification environments. In contrast to the preset method, the successive nuclei setting method (SNS) was carried out. This method manages the nucleus settings by every time step. The microstructures by SNS were compared with those by preset. © 2001 Published by Elsevier Science Ltd.

Keywords: Solidification microstructure; Cellular automaton; Equiaxed structure

1. Introduction

High quality and good mechanical properties of metal can be obtained generally from small and fine equiaxed microstructures of castings. To control the microstructures of cast alloys, processing control is so important that microstructure prediction by numerical analysis becomes necessary. Gandin and Rappaz [1] used the cellular automaton (CA) method in predicting the solidification microstructure and applied their model for Al–Si cast alloys, firstly under given cooling rate. This method is considered the preferred orientation of crystal growth, nucleation by Gaussian distribution, and dendritic growth following the stochastic method. The stochastic model was managed as the position, preferred orientation and temperature of nucleation. However, this model was not fully coupled with the heat transfer calculation, so that micro segregation was ignored and the latent heat was not fed back to the solidification calculation. The full coupling of micro–macro calculation was stated by Gandin and Rappaz [2]. They coupled CA with FEM (CA–FE) and also updated the CA capturing method to preserve the preferred orientation in the growth calculation under a non-uniform temperature field. In the CA–FE model, micro segregation was considered by calculating the solid fraction change using the CA temperature, fed back to the heat transfer calculation. Lee and Hong [3] coupled CA with Finite Volume (FV), and the microstructure of ribbon under high cooling rate was predicted. They used the LKT model for dendritic growth in contrast to the KGT (Kurz, Giovanelo, Trivedi) model in CA–FV. The CA–FV was found to be successful in predicting the ribbon structures through comparison with experiments. However, at the initial stage of calculation, the CA–FE and CA–FV methods set nucleation-related information by Gaussian distribution to random CA meshes.

Following Gaussian distribution in nucleation, six nucleation-related parameters were used, and by changing these parameters, the predicted microstructures showed various changes. Thus, the determination of nucleation parameters is very important in calculating the microstructure correctly compared to the real microstructures of cast alloys. Unfortunately, it is impossible to obtain the information of nucleation from the experiments directly and the relationships between the nucleation parameters are not yet clear. To clarify the relationships between the nucleation and grains of microstructures, the nucleation effectiveness was inspected.

Firstly, CA was coupled with the direct finite difference
(DFD) method, and then the effects of nucleation parameters on the microstructures were inspected using CA–DFD. The nucleus numbers and grain numbers were counted by repeated calculation experiments, and the successive nuclei setting method was carried out and compared with the conventional preset method. All calculations were carried on Al–7 wt% Si alloys and in 2D.

2. Calculation method

2.1. CA–DFD

The heterogeneous nucleation was assumed to follow the Gaussian distribution by Thevoz et al. [4] or was taken as instantaneous nucleation. The parameters used in the Gaussian distribution are the mean undercooling for nucleation (ΔT_{nuc}), the standard deviation (ΔT_{n}), which is treated as zero in the instantaneous nucleation, and the maximum nuclei density (n_{max}) for surface or bulk. In the Gaussian distribution the nucleation is characterized as a function of undercooling as in Eq. (1):

\[
\frac{dn}{d(\Delta T)} = \frac{n_{max}}{\sqrt{2\pi} \Delta T_{n,\sigma}} \exp \left[ -\frac{1}{2} \left( \frac{\Delta T - \Delta T_{nuc}}{\Delta T_{n,\sigma}} \right)^2 \right]
\]  

(1)

According to Eq. (1), the nucleation temperatures (T_{i} − ΔT) are set to CA meshes. After the nucleation temperature determination, the preferred orientations are also set to them. These positions, the nucleation temperature and the preferred orientation, were selected randomly before solidification. Hereafter, this is called the preset nuclei method. The crystal or grain that has the preferred orientation having the best alignment towards the heat flow direction, can defeat the others. Assuming that Al–Si alloy has a cubic structure, the preferred orientation is equal to (10) and has four symmetries in 2D.

The preferred orientation was divided into 45 cases between −45 and 45°, so that the misfit angle between contacting 2 preferred orientation cases is equal to 2°. Among the 45 cases, the randomly selected preferred orientation numbers were set to the CA meshes above.

In the calculation, many random numbers were used, so it is important to choose a good random number-generating algorithm to avoid periodicity in random numbers. The BMW method [5] has a wide periodic range and was used in CA–DFD to generate random values between 0 and 1. The generated random numbers ranging from 0 to 1 were converted to integer values to select the CA meshes randomly, and to double-precision values to produce the nucleation temperatures by Eq. (2). The symbols ξ_{1}, ξ_{2} are the random numbers ranging from 0 to 1:

\[
T_{nuc} = T_{i} - \sqrt{-2\log(\xi_{1})} \sin(2\pi \xi_{2}) \Delta T_{i,\sigma} - \Delta T_{i,nuc}
\]  

(2)

\[
T_{nuc} = T_{i} - \sqrt{-2\log(\xi_{1})} \cos(2\pi \xi_{2}) \Delta T_{i,\sigma} - \Delta T_{i,nuc}
\]  

The KGT model was applied for the dendritic growth calculation under local equilibrium at the solid–liquid interface and steady-state growth. Following the KGT model, the dendrite tip temperature can be calculated from Eq. (3), by means of which, the dendrite growth rate (ν) vs. tip temperature (T_{tip}) relationship was calculated for the Al–7 wt% Si in the case of temperature gradient G = 0 and 50 K/cm. Fig. 1 shows the relationship of: (a) ν vs. T_{tip}, and (b) ν vs. ΔT_{tip}. These relationships were simplified by interpolation:

\[
T_{tip} = T_{i} + mC_{p} \left( \frac{1}{1 - (1-k)\nu(P_{c})} \right) - \frac{2T}{R}
\]  

(3)

2.2. Grain boundary recognition

To obtain grain numbers, cases numbered (1–45) of preferred orientation were used. The grain boundary was determined between CA meshes having different preferred orientation number. The recursive method is useful to trace the closed boundary of grain. The procedure is as follows.

1. Set the grain number of all CA meshes to 0, and store the scanning start position of the CA mesh in (x,y).
2. Set the ID number to the preferred orientation number of the CA mesh positioned by (x,y) for which the grain number is 0.
3. Compare the preferred orientation number with 8 neighboring CA meshes with the ID number.
4. If the neighbor has the same preferred orientation number and its grain number is equal to 0, set the parent’s grain number to it and repeat steps (2)–(4) on it.
5. Increase the grain number.
6. Repeat steps (2)–(5) until all CA meshes have a larger grain number than 0.
2.3. Successive nuclei setting method

The nucleation-related parameters were preset at the start of the calculation in CA–FE and CA–FV. However, this method (the nuclei preset method) fails to reflect the change of solidification environment to predict the resultant microstructures because no tools are available to modify the nucleation parameters during solidification. In this work, a new method to manage the nucleation parameter named the Successive Nuclei Setting method (SNS) was carried out. The nucleation parameters were preset only at the surface when the calculation starts, and subsequently CA–DFD calculations were carried out. The difference between SNS and the previous preset nuclei setting method is that the nucleation parameters in bulk are set with every time step. The procedure is as follows.

1. Preset the nucleation parameters on the surface of the castings, and determine the nucleation rate ($A_{v,nuc}$) to set every time step.
2. Perform the CA–DFD calculation.
3. Count the liquid state CA mesh ($N_{CA}$) of which the temperature is below the predefined-nucleation temperature.
4. Select the $N_{CA} \times A_{v,nuc}$ meshes from among the CA meshes above.
5. Set the random preferred orientation and the nucleation temperature to the selected values.

Repeat steps (2)–(5). In this paper, a nuclei distribution is followed by the Gaussian or the instantaneous nucleation.

3. Verification

The CA–DFD model was verified in three ways. First, the microstructure of Al–7 wt% Si was calculated under a uniform temperature field and given cooling rate. The results were coincident with those of Gandin and Rappaz [1]. The second verification was carried through comparison of the predicted microstructures with the real casting structure of Al–6 wt% Si reported by Kim [6]. In the calculation, the same thermal properties as in Ref. [6] were used. Through the comparison, the predicted microstructure was found to be equivalent to the real microstructure of the castings. Finally, to compare the microstructure of Al–7 wt% Si alloy with the predicted one, a casting experiment was carried out. The dimensions of the castings and the steel mold are $2 \times 6 \times 6 \text{ cm}^3$ and $8 \times 12 \times 6 \text{ cm}^3$, respectively. The cooling curves were recorded at 0.2 and 0.5 cm from the mold surface, and the center point of the castings. In the mold part, the temperatures of three points, 0.5, 1.0, 1.5 cm from the mold–castings interface were recorded. The molten Al–7 wt% Si alloy was poured at 973 K into a 573 K preheated mold. From the cooling curves, the heat transfer coefficient between castings and mold was calculated as a function of elapsed time. Using this heat transfer coefficient, the microstructures and a cooling curve were calculated and compared with the results of the experiment. The calculated result had a good coincidence with those of the experiment.

4. Calculation and results

4.1. Preset nuclei–born nuclei–grain number relations in equiaxed structure

Through the CA–DFD calculations, the importance of nucleation in bulk was understood, comparing to that of the surface. To inspect the nucleation process in bulk, the nucleation effectiveness should be evaluated. The mean undercooling to nucleate in bulk was fixed to be 6 K which makes the solidification structures equiaxed ones, and then the standard deviation of undercooling and maximum nuclei density were changed mutually. Every calculation stored the positions, temperatures and preferred
orientation of really nucleated CA meshes. After the calculations, the grain boundaries were determined and grains were counted by the recursive method explained in Section 2.2. At this time, to eliminate the effect of nuclei at the surface, the grains were counted only at the quarter-center part of the solidification structure. The relationships among preset and real-born nuclei, and grains are shown in Fig. 2.

(a) Grain number ratio \( \left( r_{\text{grain}} \right) \). The grain number ratio is defined as the number of grains divided by the preset-nuclei density in the bulk. In Fig. 2(a), the grain number ratio \( r_{\text{grain}} \) is very sensitive to \( \Delta T_{v,\sigma} \). Especially, \( r_{\text{grain}} \) has a value ranging from 80 to 90% when \( \Delta T_{v,\sigma} \) is small, this value continuing to decrease with increase of \( \Delta T_{v,\sigma} \), and then converging at about 40%. In the case of small \( \Delta T_{v,\sigma} \), \( r_{\text{grain}} \) shows the constant values even though the preset-nuclei density increases. In contrast to this result, the increase of \( \Delta T_{v,\sigma} \) results in the decrease of \( r_{\text{grain}} \) but it is not proportional to \( \Delta T_{v,\sigma} \). This means that as \( \Delta T_{v,\sigma} \) becomes larger, more nuclei become lost. \( r_{\text{grain}} \) can be expressed as Eq. (4):

\[
\frac{n_{\text{grain}}}{n_{v,\text{max}}} = \left( \frac{n_{\text{eff,mc}}}{n_{v,\text{max}}} \right) \left( \frac{n_{\text{grain}}}{n_{\text{eff,mc}}} \right)
\]

(b) Effective nucleation ratio \( \left( r_{\text{eff,mc}} \right) \). The ratio \( n_{\text{eff,mc}}/n_{v,\text{max}} \) in Eq. (4) is defined as the effective nucleation ratio. According to Fig. 2(b) \( r_{\text{eff,mc}} \) is constant at about 90% when \( \Delta T_{v,\sigma} \) is very small, but this value is also sensitive to \( \Delta T_{v,\sigma} \) \( n_{v,\text{max}} \). The \( r_{\text{eff,mc}} \) curve is same to that of \( r_{\text{grain}} \). This means that 90% of preset nuclei works efficiently at small \( \Delta T_{v,\sigma} \) but at large \( \Delta T_{v,\sigma} \), only 40% of them work actively and another 60% cannot work as nuclei to form grains. Of course the nucleation ratio increases with \( n_{v,\text{max}} \), but it is not directly proportional and \( r_{\text{eff,mc}} \) continues to decrease.

(c) Effective grain number \( \left( r_{\text{grain,eff}} \right) \). The effective grain number is the last term of Eq. (4) for which results are shown in Fig. 2(c). In this figure, \( r_{\text{grain,eff}} \) is almost constant at about 90% in all ranges, therefore the effective nuclei grow to grains. As a result, almost all of effectively nucleated CA meshes would develop to grains. These results show that the nucleation in equiaxed structures does not match proportionally with the preset nuclei, so that more nuclei than obtained grain numbers in experiments should be set, depending on cases when the Gaussian distribution is chosen as a nucleation-parameter-determining method.

4.2. Microstructures by SNS

The solidification microstructures of Al–7 wt%Si alloy were calculated by SNS and the preset method with a Gaussian distribution (Fig. 3). The increase of the mean undercooling to effect nucleation causes the extension of the columnar zone and it is the same with each nucleation method. SNS can also predict columnar growth, and columnar-equiaxed transition. However the sizes of the equiaxed grains are much different in SNS and the preset method: the grains are smaller and finer in Fig. 3(a) than in Fig. 3(b). In the preset method, the nuclei born in bulk are strongly controlled by the preset-nuclei density, so that the nucleation cannot override the preset or is much less, depending on cases. In SNS, the nucleation saturation point is controlled not by the, setting of parameters but by the competition of the nucleation and growth of grains. Then nuclei would continue to be born in the undercooled zone at a given nucleation rate until the zone is filled by non-liquid state CA meshes (nucleation, growth). During the solidification, the nuclei would be born and accumulated continuously, so that the nucleation can become a main process to fill (solidify) the undercooled zone. This makes the grain finer and as a result, the solidification microstructures become fine.

To evaluate SNS, the microstructures were calculated by changing the heat transfer coefficient between the castings and the mold, and were compared with those by the preset method. In the calculation, the mean nucleation undercooling was fixed at 2 K, which makes equiaxed structures form easily. Fig. 4 shows the effective nuclei numbers. Following the results, two lines \( n_{v,\text{max}} = 12, 13\% \) reach the saturation point of the nuclei densities that were preset, so that no more nuclei are available to reflect the effect of
thermal-environment change. However, another SNS result shows that the effective nuclei numbers increase with heat transfer coefficient. This indicates that the nuclei density should be modified depending on the thermal environments in the case of the preset method. As a result, one more parameter in addition to $\Delta T_{\text{rec}}$, $\Delta T_{\text{cr}}$, $n_{\text{max}}$ must be considered in calculating the microstructures by this preset method and this is not a simple task. However, by SNS, the effect of thermal field change is reflected by the microstructures automatically, the reason for which is that no nuclei saturation point was preset artificially.

5. Conclusion

CA was coupled with DFD, and the nucleation effectiveness was inspected in the case of Gaussian distribution. In this case, preset nuclei are not perfectly active as real nuclei, so some preset nuclei are lost, depending on the deviation of nucleation undercooling and nuclei density. In contrast to the preset method, a different method, SNS, was carried out. Through the comparison of microstructures by SNS and the preset method, SNS was found to be useful in reflecting the changes of the solidification environment on the microstructures.

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