Monte Carlo Simulation of Solidification

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Abstract

The solidification process in metals and alloys is the competitive growth of solid phase between existing solid phase and new solid nuclei. Combined with classical growth kinetics and a new nucleation criterion, a two dimensional Monte Carlo simulation technique was applied to simulate this metallurgical phenomenon in present studies. The simulation results on isothermal solidification showed that an Avrami-type solidification kinetics was exhibited in this model. The simulation results on continuous cooling solidification revealed that a wide range of microstructures from complete columnar structure to fully equiaxed grains can be obtained by changing the nucleation criterion, and that the undercooling of the melts can also affect the solidified microstructures.

Introduction

Monte Carlo simulation technique has been used to simulate microstructural evolution in many metallurgical phenomena such as recrystalization, grain growth and solidification in metals and alloys. The present studies propose a two dimensional real time Monte Carlo simulation model to simulate the microstructural evolution in solidification of metals and alloys. A triangular lattice was chosen as the simulation matrix. A lattice point in the simulation matrix represented a hexagonal area of simulated material and is referred to as a "cell". A cell in the simulation matrix can be either liquid or solid. The orientation number of a solid cell is a random integer between 1 and 100,000 and that of a liquid cell -100. A cluster of solid cells with the same orientation number defines a grain. Grain boundaries exist between two solid cells with different orientation numbers.

The transformation of a liquid cell to a solid cell is determined by nucleation probability and growth probability. The heat flow in the simulation is calculated by a finite difference method. The latent heat of fusion is also included in the simulation.

Nucleation

From classical nucleation theory, the homogeneous nucleation rate (m⁻³sec⁻¹) can be expressed as^[9]

$$I(T) = 10^{39} \exp\left[-\frac{16\pi \gamma_{s/l}^3 T_m^2}{3\Delta h_l^2 (\Delta T)^2}\right]$$
 (1)

while the heterogeneous nucleation rate can be expressed as^[9]

$$I(T) = \frac{n_s'}{n} \cdot 10^{39} \exp\left[-\frac{16\pi \gamma_{sll}^3 T_m^2}{3\Delta h_f^2 (\Delta T)^2} \cdot f(\theta)\right]$$
 (2)

where $\gamma_{s/l}$ is the solid/liquid interface energy, T_m is the melting point, Δh_f is the latent heat of fusion per volume, ΔT is the local undercooling at cell (i,j), n_s ' is the number of surface atoms on the substrate per volume of liquid, n is the number of atoms in the liquid, and $f(\theta)$ is given by

$$f(\theta) = \frac{(2 + \cos\theta) \cdot (1 - \cos\theta)^2}{4}$$
 (3)

In the simulation, it is assumed that the nucleation in a cell (i,j) would not take place until the accumulation of nucleation (N_i) within the cell reaches a critical value N_c . The accumulation of nucleation is calculated by

$$N_i = \sum_{t=0}^{t} \left[I(T) \cdot \Delta t \right] \tag{4}$$

where the nucleation rate I(T) is a function of temperature, and Δt is the real time in one simulation step.

The critical accumulation of nucleation N_c is the nucleation barrier. The higher the N_c value, the more difficult the nucleation. In present studies, the N_c was assigned as a constant and could be validated from experiments in future work.

Crystal Growth

In crystal growth theory, the rate of continuous growth (R) of a solid phase into a liquid phase is given by^[9]

$$R = R_0 \cdot \Delta T \tag{5}$$

where R_0 is a constant and ΔT is the undercooling of the liquid.

The dendritic growth rate is also a function of undercooling. Walker^[10] made extensive measurements on the growth of dendrites in nickel and cobalt and got a parabolic relationship between the growth rate and the undercooling, i.e.:

$$R = R_0 \cdot (\Delta T)^2 \tag{6}$$

The growth rate can be expressed by either Eq.(5) or Eq.(6) depending on experiment results in the alloy system of interest.

In the simulation, the growth probability of a liquid cell into a solid cell is governed by:

$$\begin{array}{ll}
0 & \text{if } T \ge T_m \\
P = P_k & \text{if } \Delta E \le 0 \text{ and } T < T_m \\
P_k \exp\left[-\Delta E/kT\right] & \text{if } \Delta E > 0 \text{ and } T < T_m
\end{array} \tag{7}$$

where
$$P_k$$
 is the kinetic growth probability, and ΔE is defined as:

$$\Delta E = \lambda \delta \left[\Delta n_{s/l} \gamma_{s/l} + \Delta n_{s/s} \gamma_{s/s} - \frac{3\sqrt{3}}{2} \lambda \frac{\Delta h_f \Delta T}{T_m} \right]$$
(8)

where ΔT is the undercooling, λ is the cell size, δ is the cell thickness, $\Delta n_{s/l}$ and $\Delta n_{s/s}$ is the change in the number of solid/liquid and solid/solid interfaces between cells if a liquid cell transforms into solid, $\gamma_{s/l}$ and $\gamma_{s/s}$ is the solid/liquid and solid/solid interface energy, T_m is the melting point, Δh_f is the latent heat of fusion per volume.

The liquid cell may transform to solid by either attaching to its solid neighbors or growing from solid nuclei in the cell. If a liquid cell is attached to its solid neighbors, then the kinetic growth probability is given by

$$P_{k} = \frac{R \cdot \Delta t}{\lambda} \tag{9}$$

If a liquid cell transforms to solid by growing from solid nuclei,

$$P_{k} = \frac{0}{2R \cdot \Delta t / \lambda} \qquad if \ N_{i} \ge N_{c}$$
 (10)

It should be noted that the simulation time step, Δt , in Eq.(9) and (10) should be small enough to ensure that the kinetic probability is less than 1. In fact, the kinetic probability is the ratio of the growing distance of the solid in a cell within one Monte Carlo step to the cell size.

Heat Flow Analysis

The two dimensional heat conduction equation is given by:

$$\frac{\partial T}{\partial t} = \frac{\partial^2(\alpha T)}{\partial x^2} + \frac{\partial^2(\alpha T)}{\partial y^2} + \frac{q}{c}$$
 (11)

where $\alpha = \kappa/c$, κ is the thermal conductivity, c is the volumetric specific heat ($c = c_p \rho$, where c_p is specific heat, ρ is the specific gravity), q is internal heat source (J/sec.m³).

X, J

Fig.1 Coordinate transition from XOY to JOI

For the Monte Carlo simulation of solidification, a two dimensional triangular lattice was selected. The relationship between the Cartesian coordinates and the triangular coordinates is

$$J = x - y \cos \beta / \sin \beta$$

$$I = y / \sin \beta$$
(12)

where x and y are Cartesian coordinates, J and I are triangular coordinates, and β =60°.

By converting to triangular coordinates, Eq.(11) can be rewritten as:

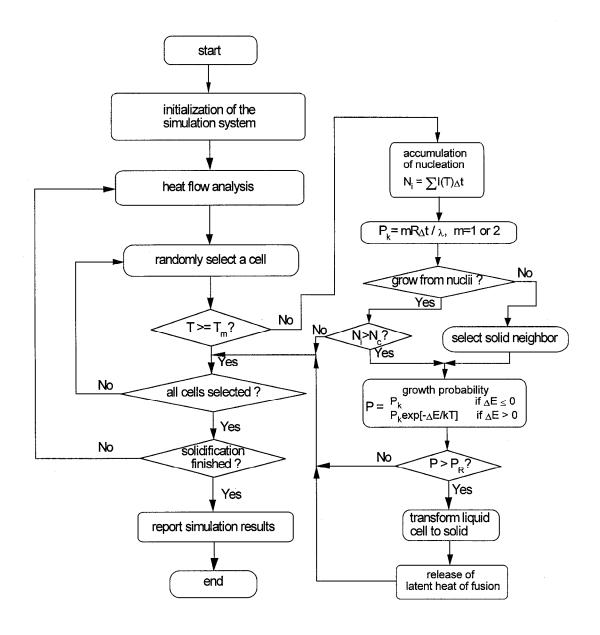


Fig. 2 Flow chart of the simulation process

$$\frac{\partial T}{\partial t} = \frac{4}{3} \frac{\partial^2(\alpha T)}{\partial I^2} - \frac{2}{3} \left[\frac{\partial^2(\alpha T)}{\partial I \partial J} + \frac{\partial^2(\alpha T)}{\partial J \partial I} \right] + \frac{4}{3} \frac{\partial^2(\alpha T)}{\partial J^2} + \frac{q}{c}$$
(13)

By applying the finite difference method, Eq.(13) becomes

$$\frac{\Delta T}{\Delta t} = \frac{2}{3\lambda^2} \sum_{n=1}^{6} \left[(\alpha T)_n - (\alpha T)_o \right] + \frac{q}{c}$$
 (14)

where $\Delta T = T(t + \Delta t) - T(t)$ is the temperature increase in Δt time interval, λ is the cell size, $(\alpha T)_0$ is (αT) at cell (i,j), $(\alpha T)_n$ are (αT) at its six nearest neighbors, q is the internal heat source at cell (i,j), and c is the volumetric specific heat. It can be seen from the above equation that the heat flow at a cell (i,j) is actually the thermal interaction between the cell and its neighbors.

In order for the calculation to converge, the cell size λ and time step Δt in Eq.(14) should be selected so that the following condition is met

$$\frac{4\alpha \cdot \Delta t}{\lambda^2} \le 1 \tag{15}$$

where α is the thermal diffusivity.

The internal heat source q during solidification is the release of the latent heat of fusion and can be expressed as

$$q = \Delta h_f \cdot P_f \tag{16}$$

where Δh_f is the latent heat of fusion per unit volume and P_f is the latent heat probability. $P_f = 1$ if a liquid cell at (i,j) transforms to solid and $P_f = 0$ if the liquid cell does not transform to solid.

Simulation Procedures

The flow chart in Fig.2 describes the simulation procedures. In the simulation, the simulation matrix size is 200×200. Due to the lack of data concerning nucleation rate and growth rate in the solidification of nickel-base superalloys, the simulations were performed in pure nickel, and the following parameters were used: $\gamma_{l/s}$ =0.255 J·m⁻², $\gamma_{s/s}$ =0.75 J·m⁻², a_s =1.548×10⁻⁵ m²·sec⁻¹, a_l =1.548×10⁻⁶ m²·sec⁻¹, c=5.184×10⁶ J·m⁻³·K⁻¹, ρ =7.2 g·cm⁻³, Tm=1728 K, Q=210,000 J·mol⁻¹, and Δh_e =1.08×10⁹ J·m³.

Simulation results and discussion

Isothermal Solidification Kinetics

The temperature in the simulation matrix was kept constant throughout the simulation. The lattice point spacing in the matrix was 5.0×10^{-4} m.

An Avrami type of solidification kinetics was observed in the simulations. The kinetics of solidification depends on nucleation rate and growth rate of the solid. An increase in the critical accumulation of nucleation, N_c , results in reduced solidification kinetics, as shown in Fig.3. The critical accumulation of nucleation N_c is actually a measurement of the nucleation barrier. Therefore, an increase in nucleation barrier must result in delaying of nucleation and

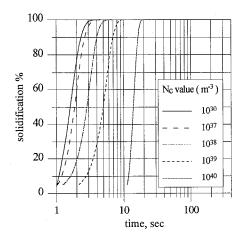


Fig.3 The effect of N_c on solidification kinetics

solidification. The crystal growth rate also influences the solidification kinetics. Fig.4 shows that an increase in growth rate accelerates the solidification kinetics.

An increase in undercooling of the melt can increase both growth rate and nucleation rate as long as the cooling rate is not extremely high. Fig.5 shows the effect of undercooling on isothermal solidification kinetics. As predicted from solidification theory, the simulated solidification rate increased with increasing undercooling.

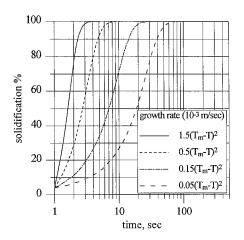


Fig. 4 The effect of crystal growth rate on solidification kinetics

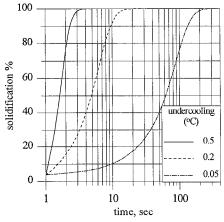


Fig. 5 The effect of undercooling on solidification kinetics

Continuous Cooling Solidification and Microstructural Simulations

In the continuous cooling solidification simulation, the cooling conditions were so defined that the mold walls (the boundaries of the simulation matrix) were kept at a constant temperature. The finite difference method was used to calculate the heat flow in the simulation matrix.

As shown in Fig.6, the critical value of N_c had a great influence on the solidified microstructures. As N_c decreased, the width of equiaxed grain zone in the center increased. This is because the decrease in N_c implies a decrease of the nucleation barrier for homogeneous nucleation in the center of the melt. Thus it would be easier to form nuclei in the center before the columnar grains grow into the center.

Undercooling of the melt before pouring into the mold has a significant effect on the microstructures. It was found that a small amount of undercooling could change the solidified microstructure dramatically, as shown in Fig.7. When the undercooling of the melt was small the nucleation probability in the center of the melt was small at beginning of the solidification. As the solidification proceeded, the temperature in the center dropped due to the heat flow towards the mold wall. This caused an increase in the nucleation probability at the mold center.



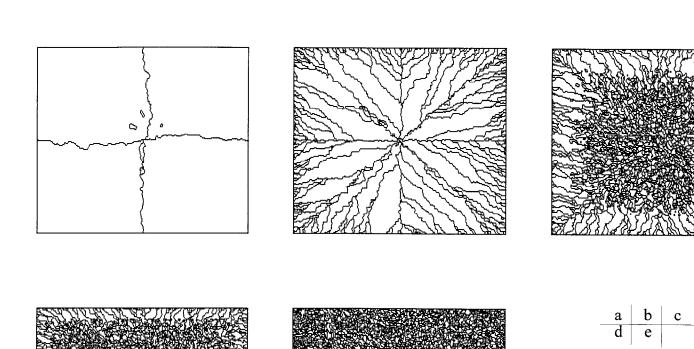


Fig. 6 The effect of N_c of microstructures, I gradually from (a (e) 1×10³⁶

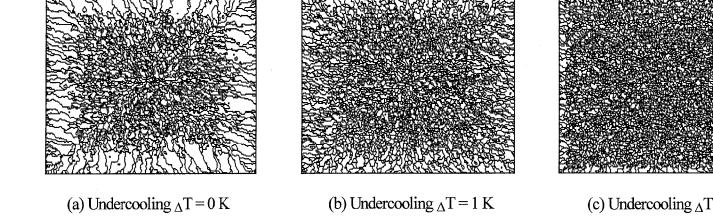


Fig. 7 The effect of undercooling on solidified microstructures

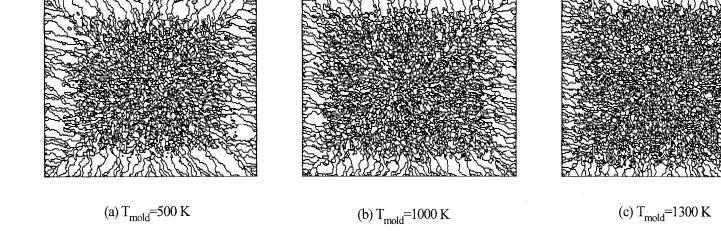


Fig. 8 The effect of mold temperature on solidified microstructures

Meanwhile, the nucleation probability in the melt layer near the mold wall is high since the mold wall temperature is far below the melting point. This produced a "quenched" layer at the mold wall. As a result of competitive growth between nuclei and existing solid, the higher the undercooling of the melt, the larger the equiaxed grain zone.

The mold wall temperature also influenced the microstructures. When the mold wall temperature is high, the columnar zone is short. When the mold wall temperature decreases, the length of the columnar zone increases, as shown in Fig.8. More heat flow is expected when the mold wall temperature is lower. And more heat flow results in a steeper thermo-gradient in front of the liquid/solid interface. This is a situation where the columnar grain growth is preferred.

Conclusions

The Monte Carlo simulation technique can be used to simulate the solidification process in the melt. In the simulation of isothermal solidification, an Avrami-type solidification kinetics was obtained. Increase of growth rate and/or decrease of critical value of accumulation of nucleation accelerate the solidification kinetics. In the simulations of continuous cooling solidification, a wide range of microstructures from complete columnar structure to fully equiaxed grains were generated by changing the nucleation criterion. The undercooling of the melt and the cooling condition of the mold affect the solidified microstructures, too.

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