Attenuation of Side Branching in Locally Modulated Dendrite under Phase-Field Simulation

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Growing from supercooled melt crystal shows complicated shapes like trees which have periodic side branching. This solidification pattern called dendrite is one of self-organized phenomena. In this phenomenon, phase and temperature distributions interact with each other during the crystal growth, since the phase transition causes latent heat. Thus, the side branches are induced from instabilities at the side wall of dendrite, though the tip of the dendrite shape was stable. In this study we have simulated pure-metal solidification by adding periodic thermal perturbations at the vicinity of the dendrite tip to investigate the shape of side branching. As a result, two types of side branches were observed when local perturbations had appropriate frequency. One type shows small wave length induced directly from perturbations, and the other large wave length expressed in comparatively away from tip. Former thin branches were attenuated to be disappeared during the latter main arms were advanced in propagation. Especially when the perturbations had small periodicity, thin side branch had not been attenuated in certain distance from modulating point. Additionally we evaluated distance from modulating point to attenuation starting point which was expected that modulation had operated side branching directly.

Key words: solidification, dendrite, phase field model, instability

1. INTRODUCTION

Solidification pattern under the non-equilibrium state such as supercooled melt shows complex shape. Especially dendritic solidification has spatial periodicity between each side branches. The dendritic solidification is one of the self-organized phenomena resulted from the balance between two opposite driving forces, inverse-thermal gradient under supercooling and capillary effect of surface energy at the interface. In this phenomenon, phase and temperature distributions interact with each other during the crystal growth, since the phase transition causes latent heat. Thus, the side branches are induced from instabilities at the side wall of dendrite, though the tip of the dendrite shape was stable.

According to mathematical models by Jian-Jun Xu, dendritic crystal growth is caused from wave emissions and reflections from singularities at the tip and one at each side [3]. In contrast, experimental results indicate the thermal noise effect to the side-branch shapes were scarce, but the gravity and convection effects were dominant. Although observations of solidification are generally difficult, there were experimental results that investigated the relation between side-branching and thermal perturbation caused from heat pulses at the dendrite tip, by using transparent organic materials [2, 5]. However, the existence of singularity has not been reported.

The phase-field model is based on the local equilibrium of free energy by treating solid-liquid interface as a diffuse layer. By using this model, shape of side branchings were investigated about thermal noise [1, 7].

In this study we have simulated pure-metal solidification by adding periodic thermal perturbations at the vicinity of the dendrite tip [4] to investigate the shape of side branching.

As a result, two types of side branches were observed when local perturbations had appropriate frequency. One type shows small wave length induced directly from perturbations, and the other large wave length expressed in comparatively away from tip. Former thin branches were attenuated to be disappeared during the latter main arms were advanced in propagation. Moreover, far away from the tip dendrite shape was similar to the previous result with whole perturbations [6]. Especially when the perturbations had small periodicity, thin side branch had not been attenuated in certain distance from modulating point.

2. CALCULATION METHOD AND CONDITIONS

2.1 Initial and boundary conditions

Simulation proceeds by using Eq. (1) and (2) with the finite difference method. The size of the 2-dimensional system was 50 × 50 µm, and the whole area was divided into 2000 × 2000 grids as shown in Fig. 1. The time increment was put at 10^{-2} ns, in order to satisfy a\Delta t / (\Delta x)^2 < 1/4, since thermal diffusion is faster than phase propagation. As the initial condition, \phi = 0 (x = 0, y = 0), \phi = 1 (x \neq 0, y \neq 0) and uniform temperature 1641 K (dimensionless supercooling (T_s - T) c_p / L = 0.2) were given so that solidification starts at the origin of the Cartesian coordinate. Priority growth orientation <100> corresponds to x- and y-axis, the boundary condition kept eight-fold symmetry (whose axis x = 0, y = 0 and x = y) and the phase and
temperature were fixed at $\phi = 1$ and $T = 1641$ K at the edge of the calculation area. As long as the solid-state region is away enough from the edge of calculation area, we assume that these conditions represent the crystal growing from a melt with homogeneous nucleation.

2.2 Material parameters

We used the following parameters by assuming pure Ni. Phase-field parameters $M = 1.73 \text{ m}^3/\text{Js}$, $\epsilon^2 = 3.13 \times 10^7 \text{ J/m}^2$ and $W = 7.85 \times 10^6 \text{ J/m}^3$ are decided to satisfy a sharp interface limit (surface thickness was assumed to be $2\lambda = 4\Delta x = 100$ nm). Material properties are listed in Table I. Parameters related to anisotropy of kinetics were decided to be $\gamma = 0.2$ and $k = 2$, since the crystal structure of Ni is fcc.

<table>
<thead>
<tr>
<th>Material parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface energy</td>
<td>0.37 J/m$^2$</td>
</tr>
<tr>
<td>Kinetic coefficient</td>
<td>2.0 m/Ks</td>
</tr>
<tr>
<td>Melting point</td>
<td>1728 K</td>
</tr>
<tr>
<td>Latent heat</td>
<td>$2.35 \times 10^7$ J/m$^2$</td>
</tr>
<tr>
<td>Specific heat</td>
<td>$5.42 \times 10^6$ J/Km$^3$</td>
</tr>
<tr>
<td>Thermal diffusion coefficient</td>
<td>$1.55 \times 10^{-15}$ m$^2$/s</td>
</tr>
</tbody>
</table>

2.3 Local thermal perturbation

To control dendrite side-branching occurrence we put local and temporally periodic perturbation to a particular point on dendrite surface near the tip. This perturbation was executed as thermal noise by substituting following equation for Eq. (2).

$$\Delta T(x, y, t) = \beta \sin \left( \frac{2\pi L}{\tau} \right) \delta(x + p_x - V t) \delta(y - s_z)$$

(5)

where $\beta$ is noise intensity which is decided for peak-to-peak value of the noise to $2 \times 10^{-3}$ of the dimensionless temperature, $\tau$ is cyclic time of perturbation, $p_x$ is distance on $x$-axis between the tip and noise-adding point, $V$ is dendrite tip velocity and $s_z$ is solid-liquid interface position found from $0.5 = \phi (V t - p_x, s_z)$. The width of delta function in Eq. (5) was considered to be $10\Delta x$. As shown in Fig. 2, noise-adding point as modulation was moved with tip propagation of dendrite.

3. RESULTS AND DISCUSSIONS

3.1 Dendrite shapes under periodic local perturbation

We used several perturbations with different frequency. For all the calculations of this paragraph, we used same location of surface modulation $p_x = 120\Delta x = 3\mu m$ and dimensionless supercooling $\Delta \mu = (T_s - T) c_p / L = 0.2$. Thus variables are only cyclic time of perturbations which had changed from 20 to 60 ns. Fig. 2 shows representative results of dendrite crystals growth after 1.6 $\mu$s. In all range of perturbation, side branches emerged and the tip velocity was constant $V = 29.6$ m/s after 0.1 $\mu$s. Dendrite shapes were able to classified into three types; side branches each had random thickness and length, shown as Fig.2 (a, b); side branching showed two periodicities as Fig.2 (c, d); side branches uniformly grew as Fig.2 (e).

![Fig.2 Dendritic solidification patterns applied periodic local perturbation with different cyclic times: (a) 20 ns, (b) 25 ns, (c) 30 ns, (d) 40 ns and (e) 50 ns. (f) is close-up of tip vicinity of (a). When the periodicity of perturbations was shorter than 20 ns (a, b), perturbations were not so effective for deciding on side-branching periodicity. When perturbations had long temporal periodicity (e), side-branches had been regularized by the perturbation directly. (c, d) show state between two side-branching modes.](image-url)

When the cyclic time $\tau$ was 20 - 25 ns, side branches had grown at random. Side branches induced directly by perturbations could not grow, besides, when $\tau = 20$ ns the interface showed...
distinctive shape shown in Fig.2 (f). We assumed that applied perturbation might be propagating without attenuation in specific region of the tip vicinity. By contrast, when cyclic time was longer than 45 ns, side branches grew uniformly. Moreover, dendrite secondary arms spacing (DAS) was directly proportional to the cyclic time of perturbation. In addition, this tendency was similar to results from periodic perturbations applying to whole calculation area[6].

Thus in the condition of the cyclic time between these two regions \(30 \leq \tau \leq 40\), cyclic branches had been induced from perturbation and from instability of the interface caused by capillary effect of surface energy and driving force of supercooling i.e. side branches had obtained two periodicity (fine branches and coarse branches). In these conditions length of fine side branches were shortened gradually by decreasing cyclic time[4].

Fig.3 shows the relation between DAS and cyclic time of local perturbation. DAS was measured from the mean distance between each root of side branch of simulated dendrite. When \(\tau > 30\) ns, DAS corresponds to theoretical equation, \(l = V\tau\), which indicates the side branching had been made by the thermal perturbation.

\[
\begin{align*}
\text{Fig.3 Relation between dendrite arms spacing and cyclic time of local perturbation. The solid line indicates DAS affected by the perturbation.}
\end{align*}
\]

3.2 Attenuation of side branching

We have changed the distance between modulating point and dendrite tip into 5 from 2 \(\mu m\) so as to investigate relation between finely side-branching and coarsely side-branching. We used same calculation conditions as above paragraph (supercooling rate \(\Delta u = 0.2\)). Applied cyclic times of perturbations were 30 and 40 ns that were selected for necessity of two periodicitys appearance in side branches. Fig.4 and 5 show dendrite growth after 1.6 \(\mu s\) to which local thermal perturbations was applied on different positions. When cyclic time was 30 ns (Fig.4) dendrites had side branches of two types; thin side branches had been appearing from the modulating point and thick side branches had been formed with thin side-branches dissolution. When distance between modulating point and the tip was less than 3.5 \(\mu m\), thin side branches also could exist at the root of the main arm.

\[
\begin{align*}
\text{Fig.4 Changes of dendrite patterns induced by different modulating positions that had been increased at every 1 \(\mu m\) from (a) 2 to (d) 5 \(\mu m\). Other conditions that had been used in all results were same (\(\tau = 30\) ns).}
\end{align*}
\]

Thin branches had been growing longer as shown in Fig.5, when cyclic time was 40 ns, nevertheless tendency to side-branches distribution was same, i.e. decaying of thin branches occurred when modulating point was located from the tip within 3.5 \(\mu m\). We expect that thin side branches and thick side branches propagate independently when cyclic time is less than 40 ns.

\[
\begin{align*}
\text{Fig.5 Changes of dendrite patterns induced by different modulating positions that had been increased at every 1 \(\mu m\) from (a) 2 to (d) 5 \(\mu m\). Other conditions that had been used in all results were same (\(\tau = 40\) ns).}
\end{align*}
\]

We had investigated attenuation of directly in-
duced side branching by perturbations. We had examined particularly tip vicinity region of the dendrite shape to investigate. In this method, we considered attenuation starting point as changing point to non-stationary growth from stationary as shown in Fig.6. Fitting the tangent line to the dendrite which includes most peaks of branches on the dendrite, attenuation starting point could be found on the last side branch propagating without attenuation because the envelope line of each side-branch top is straight while side branch has been growing stationary.

![Fig.6 Schematic figure of tangent line including a lot of tips of side branches. The distance from attenuation starting point and modulating point shown as δ in schematic means that side branches generated directly by modulation had grown without attenuation in this region.](image)

We evaluated distance from modulating point to attenuation starting point by using the above-described method. As shown in Fig.7 the value was constant as to same cyclic time on the whole, in spite of modulated position. In addition, the length of the wave-like-shape small branches induced by perturbations shown in Fig.2 (f) that had not attenuated and had been not amplified was about 4.6 µm. Additionally mean values of the length δ were 5.7 and 9.0 µm respectively, when cyclic time τ was 30 and 40 ns.

![Fig.7 Relation between distance from modulated point to dissoluation starting point and modulating point.](image)

4. CONCLUSIONS
We simulated dendritic solidification of pure metal which was applied local thermal perturbation at tip vicinity. Three types of pattern formation had been shown by difference of temporal period of the perturbation. Especially, fine pattern and coarse pattern of side branches had been existing together when periodicity of perturbation was 30 to 40 ns. In those conditions, attenuation of directly induced side branching from modulating point had been investigated by changing distance from the tip to modulating position for 2 to 5 µm. It was suggested that side-branches generated by the perturbation directly grow without attenuation.

5. REFERENCES

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