Analytical Modeling of the Growth of Arrays of Needles during Unidirectional Solidification Process under High Temperature Gradient: Part II. Comparison with Experiments

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Predicted needle dimensions given by the model proposed in part I for the growth of arrayed needles under solidification with a high temperature gradient are compared with those given by solidification of alloys; Al-Cu, Al-Fe and SCN-Argon. Correspondences between predicted primary arm spacings and experimental ones are shown to be very high for arrayed solidification of needles under high temperature gradients.

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1. Introduction

An approximate analytical description of the growth of arrayed needles is proposed and a model of the growth under a high temperature gradient has been developed in the previous work.¹⁾

Before comparing predictions with experimental results, a way to solve linear equations between unknown coefficients in the expressions of fields is shown in a model (AB-model) expressed by the addition of the integral exponential functions. Then, the convergence of summations of contributions from neighboring needles is studied.

Predicted primary spacings of needles given by models are compared with those given by solidification of alloys; Al-Cu,²⁻⁴⁾ Al-Fe⁵⁾ and SCN-Argon.^{6,7)}

2. How to Predict Needle Dimensions

2.1 How to solve linear equations and how to predict dimensions

Temperature and solute concentration are expressed by the linear combinations of the integral exponential functions. These include twelve coefficients (including tip radius of curvature of needle). Unknown coefficients are determined by boundary conditions. Boundary conditions at the interface include (a) temperature continuity, (b) heat flux conservation, (c) solute redistribution, (d) solute flux conservation and (e) temperature at the interface (Gibbs-Thomson condition). They give ten restrictions among these coefficients. As well, the solidification condition gives two restrictions (temperature gradient at the needle tip and the average solute concentration). Altogether, these twelve restrictions determine twelve unknown coefficients. For a given primary spacing (or dimensionless distance, λ), therefore, one can determine coefficients and tip radius of curvature.

Primary spacing of needles may be determined when one assumes the growing needle to be one which has the smallest tip undercooling than others have.

An example of how to solve twelve linear equations be-

tween unknown coefficients of temperature and solute concentration is given in Part I of this work. How to solve linear equations and predict dimensions is shown for AB-model in Appendix A.

2.2 Convergence of summations of contributions given by neighboring needles

In doing these calculations one has to sum contributions given by neighboring needle. Close neighbors to a specific needle each give a large contribution to the summation, but ones which are farther away give only a little, because every term in the summation decreases exponentially. Therefore, we sum individual contributions of close neighbors, and approximate the contributions from farther away neighbors by using integrated value.

For example, we have to sum contributions from neighbors to define the temperature gradient at the needle tip,

$$\sum_{i,j} \frac{e^{-P_L(\sqrt{(i^2+j^2)\lambda^2+0.25}+0.5)}}{2\sqrt{(i^2+j^2)\lambda^2+0.25}}$$

For small Peclet numbers, we have to sum contributions from infinitesimal neighbors to achieve this convergence. This convergence is easily achieved if the contributions from the members far away from the specific needle are replaced by the integrated value. We sum individual contributions within close members inside the circle with radius i_M in (i, j) plane, and take into account the contribution of members outside of this circle by the integrated value. The summation shown above is replaced as follows;

$$\sum_{i,j} \frac{e^{-P_L(\sqrt{(i^2+j^2)\lambda^2+0.25}+0.5)}}{2\sqrt{(i^2+j^2)\lambda^2+0.25}} + \frac{\pi}{\lambda^2} \frac{e^{-P_L(r_0+0.5)}}{P_L}$$

where

$$i_M = \text{maximum of } \sqrt{i^2+j^2} \text{ and } r_0 = \sqrt{(i_M\lambda)^2+0.25}$$

In this way, we can easily take into account the contributions from more distant needles. In Table 1 a list of summations needed for the calculations is shown in both summation and integrated form. Functions needed in predictions of dimen-

Table 1 Definition of functions and their integrated forms.

(integrated $i_M \leq \sqrt{i^2 + j^2} < \infty$)	
Definition by summation	Integrated form
$\sum_{ij} \frac{e^{-Pr_L}}{2r_S}$	FF1 = $\frac{\pi}{\lambda^2} \frac{1}{P} e^{-P(r_0 + \frac{1}{2})}$
$\sum_{ij} \frac{e^{-Pr_L}}{2r_S^2}$	FF2 = $\frac{\pi}{\lambda^2} e^{-\frac{P}{2}} E_1(Pr_0)$
$\sum_{ij} \frac{e^{-Pr_L}}{2r_S^3}$	FF3 = $\frac{\pi}{\lambda^2} e^{-\frac{P}{2}} \frac{E_2(Pr_0)}{r_0}$
$\sum_{ij} \frac{e^{-Pr_L}}{2r_S^4}$	FF4 = $\frac{\pi}{\lambda^2} e^{-\frac{P}{2}} \frac{E_3(Pr_0)}{r_0^2}$
$\sum_{ij} \frac{e^{-Pr_L}}{2r_S^5}$	FF5 = $\frac{\pi}{\lambda^2} e^{-\frac{P}{2}} \frac{E_4(Pr_0)}{r_0^3}$
$\sum_{ij} \frac{e^{-Pr_L}}{r_S r_L}$	GF1 = $\frac{2\pi}{\lambda^2} E_1\left(Pr_0 + \frac{P}{2}\right)$
$\sum_{ij} \frac{e^{-Pr_L}}{r_S^2 r_L}$	GF2 = $\frac{4\pi}{\lambda^2} \left\{ e^{-\frac{P}{2}} E_1(Pr_0) - E_1\left(Pr_0 + \frac{P}{2}\right) \right\}$
$\sum_{ij} \frac{e^{-Pr_L}}{r_S^3 r_L}$	GF3 = $\frac{2\pi}{\lambda^2} \left\{ 4E_1\left(Pr_0 + \frac{P}{2}\right) - 4e^{-\frac{P}{2}} E_1(Pr_0) + 2e^{-\frac{P}{2}} \frac{E_2(Pr_0)}{r_0} \right\}$
$\sum_{ij} \frac{e^{-Pr_L}}{r_L}$	GG1 = $\frac{2\pi}{\lambda^2} \left\{ \frac{1}{P} e^{-P(r_0 + \frac{1}{2})} - \frac{1}{2} E_1\left(Pr_0 + \frac{P}{2}\right) \right\}$
$\sum_{ij} \frac{e^{-Pr_L}}{r_L^2}$	GG2 = $\frac{2\pi}{\lambda^2} \left\{ E_1\left(Pr_0 + \frac{P}{2}\right) - \frac{1}{2} \frac{E_2\left(Pr_0 + \frac{P}{2}\right)}{r_0 + \frac{1}{2}} \right\}$
$\sum_{ij} \frac{e^{-Pr_L}}{r_S^2 r_L^2}$	GGF = $\frac{4\pi}{\lambda^2} \left\{ -\frac{E_2\left(Pr_0 + \frac{P}{2}\right)}{r_0 + \frac{1}{2}} - 2E_1\left(Pr_0 + \frac{P}{2}\right) + 2e^{-\frac{P}{2}} E_1(Pr_0) \right\}$

where r_S , r_L and r_0 are defined by

$$\lambda_{ij} = \lambda \sqrt{i^2 + j^2}, \quad r_S = \sqrt{\lambda_{ij}^2 + \frac{1}{4}}, \quad r_L = r_S + \frac{1}{2} \quad \text{and} \quad r_0 = \sqrt{(i_M \lambda)^2 + \frac{1}{4}}$$

and i_M is the lower bound of the integration. The integral exponential functions are defined as

$$E_2(x) = e^{-x} - xE_1(x), \quad E_3(x) = \frac{1}{2}\{e^{-x} - xE_2(x)\} \quad \text{and} \quad E_4(x) = \frac{1}{3}\{e^{-x} - xE_3(x)\}.$$

Functions for thermal field are given if P is replaced by P_L .

sions are shown in Table 2 by summation forms of the individual contributions and by integrated ones.

The convergence of functions is studied in order to show how many individual members are required for the summation. The state of convergence is shown in Table 3 and Fig. 1 as a function of the Peclet number. Table 3 shows the contribution from inside of the circle with radius i_M , the integrated contribution from outside of the circle and those sums.

From Table 3 and Fig. 1 it can be easily seen that the sum can be easily converged from the contributions of small number of neighbors for large Peclet number. But for small Peclet number, we need to sum many members to reach convergence. Though convergence is not easily reached from the summation of the contributions of individual neighbors alone, the summation of individual contributions as well as the integrated value can easily reach the convergence.

3. Comparison of Predicted Primary Spacing with Those Obtained from Experiments

Predictions from BB-model and AB-model are compared with experimental results. The C-solution in AB-model is identified with the solution in BB-model in this comparison. Since predictions given by these two solutions differ in low growth rate, the prediction given by BB-model will be compared with the experimental results. In low growth rate the morphology would be cellular, and the distribution of temperature and solute near the needle tip will be well described by the exponential functions. This is the reason why we adopt the solution given by BB-model in low growth rate.

Experiments are performed in various solidification conditions: the species of alloy, the concentration of solute, the growth rate and temperature gradient. Predictions are com-

Table 2 Representation of $\alpha_\lambda(P)$, $\beta_\lambda(P)$, $\gamma_\lambda(P)$, $\delta_\lambda(P)$ and $\varepsilon_\lambda(P)$.

Contribution from neighbors close to the very needle would be significant but ones from neighbors very far from the needle would be weak individually. Then we sum up contributions from close neighbors individually (shown by suffix 1) and take into account contributions from far neighbors by integrated from (shown by suffix 2). Every function is given by the sum of these functions, for example

$$\alpha_\lambda(P) = \alpha_\lambda^{(1)}(P) + \alpha_\lambda^{(2)}(P).$$

A2-(1) Contribution from very close neighbors to

Denote $\lambda_{ij} = \sqrt{(i^2 + j^2)\lambda^2}$, $r_S = \sqrt{\lambda_{ij}^2 + \frac{1}{4}}$, $r_L = r_S + \frac{1}{2}$ then $\alpha_\lambda(P)$, $\beta_\lambda(P)$, $\gamma_\lambda(P)$, $\delta_\lambda(P)$ and $\varepsilon_\lambda(P)$

$\alpha_\lambda^{(1)}(P)$, $\beta_\lambda^{(1)}(P)$, $\gamma_\lambda^{(1)}(P)$, $\delta_\lambda^{(1)}(P)$ and $\varepsilon_\lambda^{(1)}(P)$ are defined by summations:

$$\begin{aligned}\alpha_\lambda^{(1)}(P) &= \sum_{ij}^{i_M} \frac{e^{-P \cdot r_L}}{2 \cdot r_S} \\ \beta_\lambda^{(1)}(P) &= \sum_{ij}^{i_M} \frac{e^{-P \cdot r_L}}{r_L} \left\{ \frac{1}{2} - \frac{1}{4 \cdot r_S} + \frac{r_{ij}^2}{4 \cdot r_S^3} + \frac{r_{ij}^2}{4 \cdot r_S^2} \left[P + \frac{1}{r_L} \right] \right\} \\ \gamma_\lambda^{(1)}(P) &= \sum_{ij}^{i_M} \frac{e^{-P \cdot r_L}}{2 \cdot r_S} \left\{ \frac{P}{2} - \frac{P}{4 \cdot r_S} - \frac{1}{4 \cdot r_S^2} + \frac{r_{ij}^2}{4} \left[\frac{P^2}{r_S^2} + \frac{3P}{r_S^3} + \frac{3}{r_S^4} \right] \right\} \\ \delta_\lambda^{(1)}(P) &= \sum_{ij}^{i_M} \frac{e^{-P \cdot r_L}}{2 \cdot r_S \cdot r_L} \left\{ 1 - \frac{r_{ij}^2}{2} \left[\frac{P}{r_S} + \frac{1}{r_S^2} + \frac{1}{r_S \cdot r_L} \right] \right\} \\ \varepsilon_\lambda^{(1)}(P) &= \sum_{ij}^{i_M} E_1(P \cdot r_L)\end{aligned}$$

All integers i and j should be summed for $\sqrt{i^2 + j^2} \leq i_M$. Functions for thermal field are given if P is replaced by P_L .

A2-(2) Contribution to $\alpha_\lambda(P)$, $\beta_\lambda(P)$, $\gamma_\lambda(P)$, $\delta_\lambda(P)$ and $\varepsilon_\lambda(P)$, integrated in $i_M < \sqrt{i^2 + j^2} < \infty$.

$$\begin{aligned}\alpha_\lambda^{(2)}(P) &= \text{FF1} \\ \beta_\lambda^{(2)}(P) &= \left(\frac{P}{4} + \frac{1}{2} \right) \text{GG1} - \frac{P}{16} \text{GF2} - \frac{1}{16} \text{GF3} + \frac{1}{4} \text{GG2} - \frac{1}{16} \text{GGF} \\ \gamma_\lambda^{(2)}(P) &= \left(\frac{P^2}{4} + \frac{1}{2} P \right) \text{FF1} + \left(\frac{3}{4} P - \frac{1}{4} P \right) \text{FF2} - \left\{ \frac{1}{4} + \frac{1}{4} \left(\frac{P^2}{4} - 3 \right) \right\} \text{FF3} - \frac{3P}{16} \text{FF4} - \frac{3}{16} \text{FF5} \\ \delta_\lambda^{(2)}(P) &= \frac{1}{4} \text{GF1} + \frac{P}{16} \text{GF2} + \frac{1}{16} \text{GF3} - \frac{P}{4} \text{GG1} - \frac{1}{4} \text{GG2} + \frac{1}{16} \text{GGF} \\ \varepsilon_\lambda^{(2)}(P) &= \frac{2\pi}{\lambda^2} \left\{ \frac{r_0}{P} E_2 \left(P r_0 + \frac{P}{2} \right) + \frac{1}{P^2} E_3 \left(P r_0 + \frac{P}{2} \right) \right\}\end{aligned}$$

Functions FF1, etc., and r_0 and $E_1(x)$ are defined in Table 1. For thermal functions $\alpha_\lambda^{(2)}(P_L)$ etc., replace P by P_L .

pared with four kinds of experiment in the following special points:

- Al-0.55 mass%Cu alloy; data are given in the growth region from the growth rate of transition from the planar interface to cellular to the growth rate of absolute stability. The dependency of primary arm spacing on the growth rate will be studied.
- Al-6.0 mass%Fe alloy; applicability to the different species of alloy and different physical property, especially this alloy has smaller thermal conductivity in solid than that in liquid.
- Al-25.7 mass%Cu alloy; applicability of the model to the solidification of alloy with high solute concentration will be studied. The effect of high solute concentration is taken into account by the dependency of liquidus temperature on the average solute concentration in the model.
- SCN-0.0516 mass%Argon; applicability to the alloy with very dilute average concentration of solute. Dilute

solute concentration will result to the large growth rate from the planar interface to cellular and the small growth rate of absolute stability. In the model the dilute solute concentration makes small the loops corresponding to D-solution. At very dilute concentration the D-solution will disappear and C-solution remains. This alloy is the case. Solute trapping is also taken into account in some solidification. In the model the solute trapping merely makes small the growth rate of absolute stability, but the predicted primary arm spacing changes a little.

The physical properties used in the calculations are given in Table 4. The diffusion of solute in solid is assumed to be very small, $D_S = D/10000$, in all calculations. Calculations are done by taking $i_M = 20$ after the study of convergence of summation of (i, j) . There, the contributions inside the circle with radius $i_M = 20$ in (i, j) plane are summed individually and ones outside of this circle are approximated by the integrated value.

Table 3 Convergence of summations in (i, j) in $\alpha_\lambda(P)$ etc.

(a) $P = 0.1, \lambda = 1.0$								
	$i_M = 10$		$i_M = 20$		$i_M = 40$		$i_M = 100$	
$\alpha_\lambda(P)$	17.59	28.57	24.49	28.53	27.98	28.53	28.53	28.53
	10.98		4.04		0.54		1.4×10^{-3}	
$\beta_\lambda(P)$	16.73	28.23	23.94	28.18	27.61	28.18	28.18	28.18
	11.50		4.23		0.57		1.4×10^{-3}	
$\gamma_\lambda(P)$	1.284	1.915	1.690	1.912	1.882	1.912	1.912	1.912
	0.631		0.222		2.9×10^{-2}		7.2×10^{-5}	
$\delta_\lambda(P)$	0.8668	0.3452	0.5443	0.3472	0.3743	0.3743	0.4373	0.3473
	-0.521		-0.1970		-2.7×10^{-2}		-6.7×10^{-5}	
$\varepsilon_\lambda(P)$	133.9	284.8	222.3	284.3	275.1	284.3	284.3	284.3
	151.0		61.9		9.2		2.5×10^{-2}	
(b) $P = 0.01, \lambda = 1.0$								
	$i_M = 20$		$i_M = 40$		$i_M = 100$		$i_M = 1000$	
$\alpha_\lambda(P)$	55.2	311.1	101.6	311.1	196.1	311.1	311.1	311.1
	255.9		209.5		115.0		1.42×10^{-2}	
$\beta_\lambda(P)$	53.6	310.8	100.2	310.8	159.2	310.8	310.8	310.8
	257.2		210.5		115.6		1.54×10^{-2}	
$\gamma_\lambda(P)$	0.7816	2.131	1.052	2.131	1.548	2.131	2.131	2.131
	1.350		1.079		0.584		7.14×10^{-5}	
$\delta_\lambda(P)$	1.616	0.3681	1.402	0.3682	0.9403	0.3682	0.3682	0.3682
	-1.248		-1.035		-0.5721		-7.10×10^{-5}	
$\varepsilon_\lambda(P)$	2.03×10^3	3.110×10^4	5.36×10^3	3.110×10^4	1.500×10^4	3.110×10^4	3.110×10^4	3.110×10^4
	2.907×10^4		2.574×10^4		1.611×10^4		2.61	
(c) $P = 0.001, \lambda = 1.0$								
	$i_M = 20$		$i_M = 100$		$i_M = 1000$		$i_M = 10000$	
$\alpha_\lambda(P)$	60.7	3.139×10^3	297.3	3.139×10^3	1.983×10^3	3.139×10^3	3.138×10^3	3.139×10^3
	3.078×10^3		2.841×10^3		1.155×10^3		0.143	
$\beta_\lambda(P)$	58.8	3.138×10^3	295.6	3.138×10^3	1.982×10^3	3.138×10^3	3.138×10^3	3.138×10^3
	3.079×10^3		2.843×10^3		1.156×10^3		0.143	
$\gamma_\lambda(P)$	0.5381	2.155	0.7191	2.154	1.576	2.155	2.155	2.155
	1.617		1.436		0.5784		7.13×10^{-5}	
$\delta_\lambda(P)$	1.871	0.3703	1.784	0.3703	0.9476	0.3703	0.3709	0.3703
	-1.500		-1.413		-0.5773		-7.13×10^{-5}	
$\varepsilon_\lambda(P)$	4.77×10^3	3.138×10^6	3.16×10^4	3.138×10^6	1.517×10^6	3.138×10^6	3.138×10^6	3.139×10^6
	3.134×10^6		3.067×10^6		1.52×10^6		2.63×10^2

Fig. 1 Convergence of $\alpha_\lambda(P)$ as a function of i_M for summations in (i, j) plane.

3.1 Al-0.55 mass% Cu alloy with $G_L = 2.7 \times 10^4$ K/cm²)

This is a case with low solute concentration and high temperature gradient. In the calculation all physical properties are assumed to be constant, because of low solute concentra-

tion. Solute trapping is not assumed in this calculation. The experiments have been performed by horizontal solidification by YAG laser remelting and solidification of this alloy, and the primary spacing is observed by SEM.

In Fig. 2 the predicted primary spacing is compared with experimental results. Predicted spacing given by a solution given by BB-model and that given by D-solution are shown by lines. The shaded area shows the region of predicted primary spacing corresponding to

$$\lambda_1(\Delta T_{\min}) \leq \lambda_1 \leq \lambda_1(\Delta T_{\min} \times 1.05)$$

for D-solution. This area shows that small deviation of undercooling of tip will result large dispersion of spacing.

In small growth rate, the spacing is well predicted by a solution of BB-model. In high growth rate, the models predict smaller spacing than those given by experiment. Near the absolutely stable growth rate, the predicted spacing by a solution of BB-model and D-solution increases in tandem.

Experimental results show the transition from cellular in-

Table 4 Physical properties used in calculation.

Property	Alloy	Al-4.0 mass%Cu	Al-6.0 mass%Fe	Al-25.7 mass%Cu	SCN-0.0516 mass%Argon
melting point T_M [K]		399.6	933.6#	933.6#	331.24
thermal diffusivity α_L [cm ² /s]		0.376	0.763	0.437	1.16×10^{-3}
ratio of thermal conductivity K_S/K_L {—}		2.33	0.556	2.33	1.008
latent heat/heat capacity $\Delta H/c_p$ [K]		447	211.1	420	24.84
diffusivity of solute D [cm ² /s]		5.0×10^{-5}	##	##	8.8×10^{-6}
cappillary coefficient $\gamma/\Delta S$ [K·cm]		1.04×10^{-5}	1.0×10^{-5}	2.4×10^{-5}	6.222×10^{-6}
redistribution coefficient k {—}		0.140	##	##	0.20
liquidus slope m_0 [K/mass%]		-2.50	#	#	-4.70

#Equilibrium liquidus temperature and liquidus slope:

$$\text{Al-6.0 mass\%Fe; } T [\text{K}] = 933.6 - 7.2911C + 0.3067447C^2 - 0.7033261C^3 \\ + 2.80184 \times 10^{-3}C^4 - 4.361545 \times 10^{-5}C^5$$

$$\text{Al-25.7 mass\%Cu; } T [\text{K}] = 933.6 - 5.76C - 0.34367C^2 + 0.04315C^3 \\ - 2.2054 \times 10^{-3}C^4 + 3.7162 \times 10^{-5}C^5$$

where $C = C [\text{at\%}]$, and liquidus slope is defined by

$$m_0 = \frac{dT}{dC}.$$

##Diffusion coefficient and redistribution coefficient:

$$D = D_0 \exp\left(-\frac{Q}{RT}\right)$$

$$\text{Al-6.0 mass\%Fe; } D_0 = 1.10 \times 10^{-3} [\text{cm}^2/\text{s}], \quad Q = 2.38 \times 10^4 [\text{J/mol}]$$

$$\text{Al-25.7 mass\%Cu; } D_0 = 2.95 \times 10^{-3} [\text{cm}^2/\text{s}], \quad Q = 4.00 \times 10^4 [\text{J/mol}]$$

$$k = \frac{k_e + a_0 \frac{V}{D}}{1 + a_0 \frac{V}{D}}$$

$$\text{Al-6.0 mass\%Fe; } k_e = 0.16792 - 1.5244 \times 10^{-4} T,$$

$$a_0 = 1.0 \times 10^{-7} [\text{cm}]$$

$$\text{Al-25.7 mass\%Cu; } k_e = 1.137138 \times 10^4 - 66.96962T + 0.15815355T^2 - 1.8679168 \times 10^{-4}T^3$$

$$+ 1.1023244 \times 10^{-7}T^4 - 2.6001967 \times 10^{-11}T^5,$$

$$a_0 = 5.0 \times 10^{-7} [\text{cm}]$$

Fig. 2 Comparison of the predicted primary spacing with that given by experiment for Al-0.55 mass%Cu alloy. Open circles show cell and closed circles do dendrite. Full line shows the predicted primary spacing given by D-solution in AB-model. The dot line shows the predicted spacing given by BB-model.

terface to cellular-dendritic one in low growth rate, where as the models predict the transition from a solution of BB-model to D-solution of AB-model.

3.2 Al-6.0 mass%Fe alloy with $G_L = 5.0 \times 10^4 \text{ K/cm}^5$)

Experiments have been performed by solidification through CO_2 laser remelting and solidification of this alloy. The temperature gradient is very large compared with that of usual solidification. In predictions the liquidus temperature is assumed to be dependent on average solute concentration, and the diffusion coefficient of solute is dependent on liquidus temperature.⁵⁾ Solute trapping is also assumed in the prediction. The dependency relationship is shown in Table 4.

In Fig. 3(a) the predicted primary spacing is compared with experimental one. The corresponding tip undercooling is shown in Fig. 3(b) with growth rate.

The primary spacing is fairly well predicted. The predicted tip undercooling decreases at high growth rate near the absolutely stable growth rate as expected by solute trapping.⁵⁾

Figure 3(a) shows that in the solidification with high solute concentration the predicted spacing by C-solution differs from that of D-solution in the high growth rate. And the primary spacing is fairly well predicted by C-solution. The tip undercooling given by C-solution is larger than that given by

Fig. 3 (a) Comparison of the predicted primary spacing with that given by experiment for Al-6.0mass%Fe alloy. Predicted primary spacings given by C-solution and D-solution in AB-model are shown in this Figure. Predicted primary spacing given by C-solution is similar to one given by BB-model. (b) Predicted tip undercooling with growth rate for Al-6.0 mass%Fe alloy.

D-solution (Fig. 3(b)). The best solution for comparison with experiment will be discussed in the next section.

S.-Z. Lu *et al.*⁸⁻¹¹⁾ have analyzed this solidification by numerical simulation. The predictions given by them are very similar to ones given in this work. (Compare Fig. 3(a) in this text with Fig. 7 in Ref. 8)).

3.3 Al-25.7 mass%Cu alloy with $G_L = 5.0 \times 10^4 \text{ K/cm}^{3,4)}$

Experiments have been also performed by solidification by CO₂ laser through remelting and solidification of this alloy. This is an experiment with high temperature gradient and high solute concentration. In predictions the liquidus temperature is assumed to be dependent on the average solute concentration, and the solute diffusivity is dependent on the liquidus temperature. Solute trapping is also assumed in the prediction. The dependency relationship is shown in Table 4.⁴⁾

The predicted primary spacing given by C-solution fairly well corresponds to the experimental data, but slightly larger than that given by experiment (Fig. 4). The predicted primary spacing given by D-solution is larger than that of C-solution.

3.4 SCN-0.0516 mass%Argon alloy with $G_L = 5.0 \times 10^2 \text{ K/cm}^{6,7)}$

This experiment has been performed by solidification where a special equipment is used for in-situ observation of rapid solidification. The solute concentration is very dilute and the temperature gradient is large. Observed morphology is cellular and the primary spacing is measured with different growth rate.

In Fig. 5 the predicted primary spacing is compared with

Fig. 4 Comparison of the predicted primary spacing with that given by experiment for Al-25.7mass%Cu alloy.

Fig. 5 Comparison of the predicted primary spacing with that given by experiment for SCN-0.0516 mass%Argon alloy.

that given by experiments. In this solidification the primary spacing is predicted by the BB-model (or C-solution in AB-model) alone. There is no D-solution in this case.

The correspondence between prediction and experiment is fairly similar both in the magnitude and the dependency on the growth rate.

4. Discussion and Conclusion

Predicted needle dimensions which are given by the models of arrayed needles under solidification with high temperature gradient, are compared with those given by solidification of alloys; Al-Cu, Al-Fe and SCN-Argon. Correspondences between predicted primary spacing and experimental ones are shown to be very high under a high temperature gradient.

Positive features of the model are as follows;

- (a) prediction of the dependency on thermal diffusivity: the thermal diffusivity of metallic alloy (Al-Cu and Al-Fe) is, for example, two order of magnitude larger than that of organic alloy (SCN-Argon), but the dependency of primary arm spacing on growth rate is well predicted.
- (b) dependence of dimension on average solute concentration: the primary arm spacing for alloy with very dilute solute concentration (SCN-0.0516 mass%Argon) is properly predicted as well as the alloy with high solute concentration (Al-27.8 mass%Cu).
- (c) dependency on temperature gradient: the temperature gradient for metallic alloy (Al-Cu and Al-Fe) is about two order of magnitude larger than that for the organic alloy (SCN-Argon). The larger temperature gradient

makes the predicted primary arm spacing smaller in the prediction.

These features show that the addition of the Ivantsov function well describes the field of temperature and solute in the solidification with a high temperature gradient. The model has a positive solution (C-solution) that does a good job of predicting the experimental data but another solution (D-solution) has smaller tip undercooling than that of C-solution. If a needle that has smaller tip undercooling grows preferentially, then a needle given by D-solution grows easily. The principle of preferably choosing the C-solution is not supported in the proposed models.

The reason why the D-solution in the model has smaller tip undercooling than that of C-solution will be explained by the effect of diffusion from secondary arms. When the diffusion of solute from secondary arms is taken into account in the model, it will result the more pile-up of solute on the tip of needle. This pile-up of solute makes the undercooling of D-solution large, while the undercooling of C-solution remains same because of no secondary arms of cellular interface.

The proposed model should be improved in the following points.

- (1) by making a model in which both BB-model and AB-

model are satisfied,

- (2) by taking the proper tip shape of needle into account,
- (3) by taking into account of the stability of the arrayed needle against some perturbations.

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Appendix A: How to solve and predict tip radius of curvature and spacing (Numbered equations correspond to those in Table 1B in Part I)

Define

$$\begin{aligned}\tilde{G}_L &= \left[\frac{\Delta H}{c_p} P_L + \frac{\rho}{2} G_L \right] / \left(\frac{K_S}{K_L} \right) & \text{and} \quad a_{\omega 2}^L &= \alpha_\lambda(P_L) \left(\lambda_L - \frac{\omega^2}{4} \right) - \beta_\lambda(P_L) \lambda_L \\ a_{\omega 0}^L &= \left[-\frac{\rho}{2} G_L \left(\lambda_L - \frac{\omega^2}{4} \right) + \lambda_L \tilde{G}_L \right] / a_{\omega 2}^L & \text{and} \quad a_{\omega 1}^L &= \frac{\omega^2}{4} \lambda_L / a_{\omega 2}^L \\ b_{\omega 0}^L &= \left[\frac{\rho}{2} G_L \beta_\lambda(P_L) - \alpha_\lambda(P_L) \tilde{G}_L \right] / a_{\omega 2}^L & \text{and} \quad b_{\omega 1}^L &= -\frac{\omega^2}{4} \alpha_\lambda(P_L) / a_{\omega 2}^L\end{aligned}$$

Then eqs. (1), (4), (5) and (6) give

$$B_\omega^S e^{\lambda_S} = B_U^S / B_D^S \quad (a) \quad A_0^L = a_{\omega 0}^L + a_{\omega 1}^L B_\omega^S e^{\lambda_S} \quad (b) \quad B_\omega^L e^{-\lambda_L} = b_{\omega 0}^L + b_{\omega 1}^L B_\omega^S e^{\lambda_S} \quad (c) \quad P_S B_0^S e^{-P_S} = -\tilde{G}_L + \lambda_S B_\omega^S e^{\lambda_S} \quad (d)$$

where

$$\begin{aligned}B_U^S &= -a_{\omega 0}^L [P_L \beta_\lambda(P_L) - \gamma_\lambda(P_L) - \delta_\lambda(P_L)] - b_{\omega 0}^L \left[(P_L - \lambda_L) \left(\lambda_L - \frac{\omega^2}{4} \right) - \frac{\omega^2}{4} \right] \\ B_D^S &= a_{\omega 1}^L [P_L \beta_\lambda(P_L) - \gamma_\lambda(P_L) - \delta_\lambda(P_L)] + b_{\omega 1}^L \left[(P_L - \lambda_L) \left(\lambda_L - \frac{\omega^2}{4} \right) - \frac{\omega^2}{4} \right] - \left[\left(P_L + \frac{K_S}{K_L} \lambda_S \right) \left(-\lambda_S - \frac{\omega^2}{4} \right) - \frac{K_S}{K_L} \frac{\omega^2}{4} \right]\end{aligned}$$

For solute parameters, define

$$\begin{aligned}a_{\omega 2}^P &= -[(1-k)P \varepsilon_\lambda(P) - \alpha_\lambda(P)] \left(\lambda_P - \frac{\omega^2}{4} \right) + \beta_\lambda(P) [(1-k)P - \lambda_P] \\ a_{\omega 0}^P &= (1-k)P C_\infty^L \left(\lambda_P - \frac{\omega^2}{4} \right) / a_{\omega 2}^P \quad a_{\omega 1}^P = - \left[\left(\lambda_P - \frac{\omega^2}{4} \right) \frac{D_C}{D} \lambda_C - [(1-k)P - \lambda_P] \left(-\lambda_C - \frac{\omega^2}{4} \right) / k \right] / a_{\omega 2}^P \\ b_{\omega 0}^P &= -(1-k)P C_\infty^L \beta_\lambda(P) / a_{\omega 2}^P \quad b_{\omega 1}^P = \left[\beta_\lambda(P) \frac{D_C}{D} \lambda_C - [(1-k)P \varepsilon_\lambda(P) - \alpha_\lambda(P)] \left(-\lambda_C - \frac{\omega^2}{4} \right) / k \right] / a_{\omega 2}^P\end{aligned}$$

Then eqs. (8), (9) and (10) give

$$B_\omega^C e^{\lambda_C} = B_U^C / B_D^C \quad (e) \quad A_0 = a_{\omega 0}^P + a_{\omega 1}^P B_\omega^C e^{\lambda_C} \quad (f) \quad B_\omega e^{-\lambda_P} = b_{\omega 0}^P + b_{\omega 1}^P B_\omega^C e^{\lambda_C} \quad (g)$$

where

$$B_U^C = -a_{\omega 0}^P [P\beta_\lambda(P) - \gamma_\lambda(P) - \delta_\lambda(P)] - b_{\omega 0}^P \left[(P - \lambda_P) \left(\lambda_P - \frac{\omega^2}{4} \right) - \frac{\omega^2}{4} \right]$$

$$B_D^C = a_{\omega 1}^P [P\beta_\lambda(P) - \gamma_\lambda(P) - \delta_\lambda(P)] + b_{\omega 1}^P \left[(P - \lambda_P) \left(\lambda_P - \frac{\omega^2}{4} \right) - \frac{\omega^2}{4} \right] - \left[\left(P + \frac{D_C}{D} \lambda_C \right) \left(-\lambda_C - \frac{\omega^2}{4} \right) - \frac{D_C}{D} \frac{\omega^2}{4} \right]$$

Derivation of tip radius: For a given G_L , a given C_∞^L and a given growth rate V , and for some given λ , assume tip radius ρ , then (a) and (d) give $P_S B_0^S e^{-P_S}$ and $B_\omega^S e^{\lambda_S}$ and (e) gives $B_\omega^C e^{\lambda_C}$.

Insert these into (12) for a given growth rate V_0 ,

$$P_S B_0^S e^{-P_S} + \left(-\lambda_S - \frac{\omega^2}{4} \right) B_\omega^S e^{\lambda_S} = \frac{\gamma}{\Delta S} \frac{2}{\rho} + \frac{m_0}{k} \left(-\lambda_C - \frac{\omega^2}{4} \right) B_\omega^C e^{\lambda_C}$$

Choose ρ to be satisfied eq. (12), then ρ and C_t^L is determined for a given λ .

Calculate for other λ 's, and choose a λ to minimize C_t^L .

Then a set or sets of ρ , λ and C_t^L is determined.

This procedure is shown below by flow chart for calculation.