Thermal and Solutal Influences of Continuous Fibers on Matrix Dendrite Growth in Composite Materials

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Numerical simulation was applied to evaluate the thermal and solutal influences of fibers on the dendrite growth of matrix alloy reinforced with continuous fibers. The results of temperature and solute distribution, and dendrite tip undercooling and tip radius were compared with experimental data obtained from directional solidification studies for polyvinylidene fluoride, Pyrex glass, and copper fiber reinforced succinonitrile-acetone alloy composites. In the polyvinylidene fluoride fiber/pure succinonitrile composites, dendrite in the composite region grew behind that in the bulk region, and in the direction of the heat flow. However, dendrite between copper fibers grew faster than that in the bulk region. A computer simulation revealed that a difference in thermal diffusivity influences the thermal distribution of specimens and the apparent dendrite tip location. The gap in dendrite tips between the composite and bulk regions increased as the composition of acetone increased or the fiber interstices became smaller. Numerical analysis revealed that tip composition and undercooling increased as the fiber interstices became smaller than the primary dendrite arm spacing. The constraint of growth on the secondary arm and the change of dendrite morphology were also shown in the analysis.

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1. Introduction

During liquid state fabrication for producing metal matrix composites (MMCs), molten alloy is solidified in the small gap of the fiber preform. The dendrite growth morphology of the primary phases could be determined by the influence of the fibers, as thermal and solute diffusion, and the natural convection at the solid/liquid interface are different from those in the bulk region. It is necessary to clarify the effects of fibers that control the morphology of the phases of MMCs in order to develop and optimize high-performance MMCs. The shape changes or the microsegregation of planar or cellular interfaces due to fibers,^{1,2)} and the influence of fibers on tip composition and undercooling³⁾ have been reported for alumina fiber/Al-Cu alloy composites. Also, the morphology of the primary phase in narrow channels has been reported⁴⁻⁶⁾ for succinonitrile (SCN)-acetone alloys as a model of MMCs. However, few papers have been concerned with the thermal, solutal and geometrical influences of fibers on dendritic growth, and the solidification mechanism is still unrevealed.

The purpose of the present study is to verify the influence of fibers on dendrite growth using a numerical simulation, as has been developed for microstructural analysis in bulk solidification. First, thermal distribution was calculated in the whole sample, then the dendrite morphology was simulated using a model based on solutal diffusion, tip curvature and local equilibrium, and finally dendrite parameters such as dendrite tip curvature and tip undercooling were compared to the results from fiber reinforced SCN-acetone alloy composites.

2. Experimental Procedure and Results⁶⁾

Pure SCN and SCN-2.2mass% acetone alloys, respectively, were used as the matrix alloys to investigate the ther-

mal and the solutal influences of fibers on dendrite growth. The SCN alloy was infiltrated in a 150 μm gap between glass slides (26 mm width and 86 mm length). Polyvinylidene fluoride (PVDF), Pyrex glass or a copper continuous fiber bunch was also placed between the slides, at the center, to form the composite region. The plate-like specimens were unidirectionally solidified at a fixed velocity of 10 or 150 $\mu\text{m/s}$ and a temperature gradient of 3 K/mm. The matrices in the composite region grew individually in the channel surrounded by fibers and the slide. The microstructure, and the dendrite tip position and tip radius were analyzed with an optical microscope and a video camera.

The dendrites in both the bulk and the composite regions showed steady state solidification. However, differences of the dendrite tip position between the bulk and the composite regions (L), were found in the direction of the heat flow. Figure 1 shows the value of L in relation to the fiber species and spacing. The tip positions of the dendrites, which solidified between the PVDF fibers, were located behind those in the bulk region, whereas, the dendrites between copper fibers grew ahead of those in the bulk region. Furthermore, the dendrite position gaps increased as the moving velocity of specimen increased. This remarkable difference of the dendrite tip position was not detected in glass fiber/SCN composites. Since the thermal diffusivity of PVDF fibers is smaller than that of the SCN matrix, and that of copper fibers is larger, these thermal properties can influence the dendrite tip position. In the case of the SCN-acetone alloy matrix, the dendrite tips go down to the cooler part, compared to pure SCN specimens, as shown in Fig. 2. Furthermore, the dendrite tip radius decreased as the fiber interstices became smaller, as shown in Fig. 3. In addition to the changes of these parameters, the growth of the secondary dendrite arms was constrained and almost stopped by the presence of the fibers.

Fig. 1 Relative positions of dendrite tips in composite regions for PVDF, Pyrex glass and copper fiber/SCN composites. ($G = 3 \text{ K/mm}$)

Fig. 2 Relative positions of dendrite tips in composite regions for PVDF and copper fiber/SCN-2.2 mass% acetone alloy composites. ($G = 3 \text{ K/mm}$)

Fig. 3 Relationship between dendrite tip radius and fiber interstice for copper fiber/SCN-2.2 mass% acetone alloy composites. ($R = 10 \mu\text{m/s}$, $G = 3 \text{ K/mm}$)

3. Thermal Influence of Fibers

As the thermal diffusivity of each fiber can affect the cooling speed of the composite region, the temperature distribution of the whole specimen was analyzed. The temperature

distribution in 2-dimensions could be expressed as the valance of energy flux.

$$\frac{\partial(\rho h)}{\partial t} + \left\{ \frac{\partial}{\partial x}(\rho u h) + \frac{\partial}{\partial y}(\rho v h) \right\} = \left\{ \frac{\partial}{\partial x} \left(\frac{\lambda}{C_p} \frac{\partial h}{\partial x} \right) + \frac{\partial}{\partial y} \left(\frac{\lambda}{C_p} \frac{\partial h}{\partial y} \right) \right\} + S_h \quad (1)$$

where ρ is the density [kg/m^3], h is the enthalpy [J], t is a time [s], λ is the thermal conductivity [W/m K], C_p is the specific heat [J/kg K], S_h is the latent heat of fusion per unit volume [J/m^3], and u and v are the velocities [m/s] in the x - and y -directions, respectively. The temperature distribution was computed using the direct finite difference method (DFDM), in a 2-dimensional domain with following assumptions: (i) density, thermal conductivity and specific heat are constant, (ii) a specimen moves only to x -directions at a velocity of u during solidification; $v = 0$, (iii) boundaries of the left and right sides are adiabatic, and those of the up and down sides are constant for hotter or cooler temperatures, respectively, as shown in Fig. 4. From these assumptions, of steady state solidification, eq. (1) becomes;

$$\rho u \frac{\partial T}{\partial x} = \frac{\lambda}{C_p} \left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right) + \frac{S_h}{C_p} \quad (2)$$

where, T is temperature [K]. Figure 5 shows the temperature distribution in the PVDF and copper fiber reinforced composite specimens. The thermophysical properties used in the calculations are listed in Table 1.⁷⁾ Solidification velocity is set at 10 and 150 $\mu\text{m/s}$. In the case of the PVDF sample, the temperature in the composite region is slightly higher than that in the matrix (Fig. 5(a)). PVDF fibers, which have the smaller thermal diffusivity in the matrix, could maintain a higher temperature. On the other hand, in the copper fiber specimen, the composite region is a little cooler than the bulk because of the larger thermal diffusivity of the metal copper fibers (Fig. 5(b)). The thermal influence of the fibers increases as the moving velocity of specimen increases (Fig. 5(c), (d)). Consequently, the location of dendrite tips could be determined by the macroscopic temperature distribution due to the thermal diffusivity of the matrix and the fibers.

Fig. 4 Schematic view of the domain for the 2-dimensional simulation analysis.

Table 1 Thermophysical properties used in the thermal distribution simulation analysis.

	ρ [kg/m ³]	C_P [J/kg K]	λ [W/m K]	$\lambda/(\rho \cdot C_P)$ [m ² /s]
SCN ⁷⁾	0.988×10^3	1.95×10^3	0.223	1.16×10^{-7}
PVDF	1.79×10^3	7.71×10^2	0.130	9.42×10^{-8}
Copper	8.93×10^3	4.07×10^2	386.0	1.06×10^{-4}

Fig. 5 Influence of the thermal diffusivity of fibers and solidification velocity on temperature distribution.

4. Solutal Influence of Fibers

4.1 Mathematical description of the model

The dendrite morphology and the solutal distribution around the dendrite were numerically analyzed with rectangular coordinates in a two-dimensional domain using the model developed by Nastac.⁸⁾ The solute diffusion is given by following equations.

(a) in the liquid phase

$$\frac{\partial C_L}{\partial t} = \frac{\partial}{\partial x} \left(D_L \frac{\partial C_L}{\partial x} \right) + \frac{\partial}{\partial y} \left(D_L \frac{\partial C_L}{\partial y} \right) \quad (3)$$

(b) in the solid phase

$$\frac{\partial C_S}{\partial t} = \frac{\partial}{\partial x} \left(D_S \frac{\partial C_S}{\partial x} \right) + \frac{\partial}{\partial y} \left(D_S \frac{\partial C_S}{\partial y} \right) \quad (4)$$

where, D_L and D_S are the diffusion coefficients [m²/s] of the solute in the liquid and solid phases, respectively, and the influence of the convection is not considered. Local equilibrium at the solid/liquid interface is satisfied as:

$$C_S^* = k \cdot C_L^* \quad (5)$$

where, C_S^* and C_L^* are solute compositions [mass%] of the

solid and liquid phases at the solid/liquid interfaces and k is the equilibrium distribution coefficient. Furthermore, the conservation of mass is given at the solid/liquid interface as:

$$V_n^* C_L^* (k - 1) = \left\{ -D_S \left(\frac{\partial C_L}{\partial x} + \frac{\partial C_L}{\partial y} \right) + D_S \left(\frac{\partial C_S}{\partial x} + \frac{\partial C_S}{\partial y} \right) \right\} \cdot n \quad (6)$$

where, V_n^* is the normal velocity of the interface of the dendrite, n is the normal to the solid/liquid interface. From a consideration of the constitutional and the curvature undercoolings, the interface temperature T^* [K] is given as:

$$T^* = T_{eq} + (C_L^* - C_0)m_L - \Gamma \kappa f(\varphi, \theta) \quad (7)$$

where, T_{eq} is the equilibrium liquidus temperature [K], m_L is the liquidus slope [K/mass%], κ is the mean curvature of the solid/liquid interface [m⁻¹]⁹⁾ and Γ is the Gibbs-Thomson coefficient [K m]. Kinetics undercooling is not considered in this calculation. The function of $f(\varphi, \theta)$ expresses a coefficient that accounts for growth anisotropy, where θ is the growth angle, and φ is the preferential crystallographic orientation angle.⁸⁾ In the present calculation, θ is supposed to be zero, which means the growth direction is initially set parallel to the heat flow direction. Furthermore, the temperature gradient ($G = 3$ K/mm) and the cooling velocity ($V_C = 3 \times 10^{-2}$ K/s) are set up as the initial conditions in order to correspond with the experimental solidification condition.

4.2 Comparison between calculation and experimental data

Figure 6 shows examples of calculated columnar dendrites solidified for 120 s. The thermal and physical properties used in the calculations are shown in Table 2. In order to model the influence of small channel surrounded fibers and glass slides, the width of the domain was changed, where no solute diffusion occurred in either the right or left boundaries. A domain height of 5 mm, sufficient to remove the influence of the final transient stage, was set up. Figure 6 is a magnification of the

Fig. 6 Simulated columnar dendrite during unidirectional solidification in the composite region ($d =$ (a) 100 μm, (b) 200 μm, (c) 300 μm, (d) 600 μm), and in the bulk region (e).

Table 2 Thermophysical properties used in the dendrite morphology and solutal distribution simulation analysis.

	C_0 [mass%]	k [—]	m_L [K/mass%]	T_L [K]	D_L [m ² /s]	D_S [m ² /s]	Γ [Kμm]
SCN-Acetone ⁷⁾	2.2	0.1	-2.8	325	1.3×10^{-9}	$1.0 \times 10^{-12*}$	6.4×10^{-8}

*: assumption

bottom part of the domain. As shown in Fig. 6(e), the primary dendrite arm spacing (λ_1) of 350 μm was detected from the results of the present calculations. The dendrite morphology was modified according to the width of the specimen. When the fiber interstice (d) is smaller than λ_1 , the width of the dendrite arm (w), that is the distance of the secondary dendrite arm tips between the right and left sides, decreases with the decreasing of the fiber interstice as shown in Figs. 6(a) and (b). This phenomenon shows a good agreement with the experimental data of the SCN alloy composites. When d is set up from 1 to about 1.5 times λ_1 , w becomes larger than λ_1 and is almost the same as d (Fig. 6(c)). On the other hand, when w is nearly twice λ_1 , and two or more dendrites grow, w becomes smaller than λ_1 (Fig. 6(d)). Additionally, when d is much smaller than λ_1 , the dendrite shape becomes cellular-like (Fig. 6(a)). Transformations from dendrite to cellular are more clearly observed in alumina/Al-Cu alloy composites produced in the directional solidification study.³⁾ Figures 7(a) and (b) show back scattered electron images and copper composition mapping in a primary α phase surrounded by continuous alumina fibers in alumina/Al-15%Cu alloy composites. The solid line in Fig. 7(b) shows composition at 2.65%Cu, which is the maximum composition in solids at 881 K according to the equilibrium phase diagram. Since the sample was quenched at 881 K, the solid/liquid interface can be indicated by a solid line, and the true shape of the dendrite tip is elliptic or parabolic. A similar phenomenon is also observed in Al-4.5%Cu specimens (Figs. 7(c) and (d)). Therefore, in a limited space surrounded by fibers, dendrite grows away from the fibers and changes to an elliptic or parabolic interface.

The variations of the differences of the dendrite tips between the composite and bulk regions (L) are summarized in Fig. 8. The results of copper fiber/SCN alloy composites were selected, since they were less thermally influenced at a low solidification velocity. The data are standardized by λ_1 because of the elimination of any size dependency. Although in Fig. 8 the absolute value of L in the SCN alloy specimens is a little larger than the calculation, the phenomena, that the tip position goes back to a cooler part of the specimen as the fiber interstices become smaller, is qualitatively the same. Figure 9 shows a comparison of dendrite tip radii in relation to the ratios of the fiber interstices and dendrite arm spacings (d/λ_1). Tip radii as calculated have a good agreement with the experimental data, in that the tip radii decrease as the fiber interstices decrease. From Fig. 9, the tip radii changed from about 8.5 μm to 6.5 μm, the curvature undercooling (ΔT_R : as shown in eq. (8)) was increased from 0.0075 K to 0.0098 K by the presence of the fibers, and the contribution of ΔT_R could be negligible.

$$\Delta T_R = \Gamma \cdot \frac{1}{r} \quad (8)$$

Additionally, excess tip composition as defined in eq. (9) in-

Fig. 7 Longitudinal section through dendrite tips in composite regions of alumina/Al-15 mass%Cu alloy composites ((a), (b), $G = 2.5$ K/mm, $R = 40$ μm/s), and the transverse section of Al-4.5 mass%Cu alloy specimen ((c), (d), $R = 2$ μm/s). Figures (a) and (c) are back-scattered electron images, and (b), (d) are copper composition maps with EPMA analysis. The solid line in Figs. (b) and (d) show the morphologies of the growing dendrites.

Fig. 8 Relationship between the relative positions of dendrite tips and the ratios of fiber interstices and dendrite arm spacings (d/λ_1). ($R = 10$ μm/s, $G = 3$ K/mm)

dicates how the solute piles up in front of the dendrite tips, and increases as (d/λ_1) becomes smaller as shown in Fig. 10.

$$f_c = \frac{C_L^{**} - C_0}{C_L^* - C_0} \quad (9)$$

where, C_L^{**} and C_L^* are the tip compositions in the composite

Fig. 9 Relationship between the dendrite tip radius and the ratio of fiber interstice and dendrite arm spacing (d/λ_1). ($R = 10 \mu\text{m/s}$, $G = 3 \text{ K/mm}$)

Fig. 10 Relationship between the excess tip composition and the ratio of fiber interstice and dendrite arm spacing (d/λ_1). ($R = 10 \mu\text{m/s}$, $G = 3 \text{ K/mm}$)

and the bulk regions, respectively. The dendrite tip undercooling in the composite region is 0.45 K larger than that in the bulk. Therefore, the difference of tip position is due mainly to an increase of the tip composition. The limitation of the fields in dendrite growth by continuous fibers can cause an increase in the solute composition around a dendrite tip and in the undercooling of the dendrite tip.

5. Conclusion

The thermal and solutal influences of continuous fiber on the growth of matrix alloy reinforced composites are evaluated by a numerical simulation, and are compared with the data of SCN alloy composites. The results can be summarized as follows:

(1) A difference of dendrite tip position between composites and bulk regions was found along the heat flow direction depending on the species and the spacing of the fibers. The heat flow simulation indicates that the thermal diffusivity of fibers could change the heat flux, the temperature distribution in the composite region, and consequentially the dendrite tip positions in SCN alloy matrix composites.

(2) The 2-dimensional simulation indicated that the change of dendrite morphology was related to the fiber interstices. The position of the dendrite tip, the tip radius, the tip composition and the tip undercooling are mutually related. The presence of fibers can cause an increase of solute pile up and dendrite tip undercooling at the dendrite tip in the composite region, especially when the fiber interstice becomes smaller than the primary dendrite arm spacing.

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