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The effect of oscillatory flow on nucleation and grain growth in the undercooled melt

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ABSTRACT

The present paper investigates the effect of the oscillatory flow induced by vibration on nucleation and grain growth in the undercooled melt in terms of the analytical method. The analytical solution shows that the oscillatory flow stimulates the meta-stable crystalline embryos to grow rapidly and facilitates to form a great number of nuclei in the undercooled melt during the initial stage of nucleation. As a grain grows, the oscillatory flow alternately facilitates and inhibits the growth of the grain such that the formed nuclei survive. For the low frequency and low acceleration, the interface temperature of a grain rises rapidly immediately after nucleation and then gradually decreases. For the high frequency, the interface temperature shows no significant difference with the increase of frequency. For the high acceleration, the interface temperature oscillates with the oscillatory flow. The oscillation acts as a stimulating and inhibitory effect and facilitates to produce a number of crystalline sites in the undercooled melt. The grains are refined with increasing vibration acceleration under a certain vibration frequency.

1. Introduction

Melt convection influences nucleation and subsequent grain growth during solidification, and determines to great extent interfacial morphologies and microstructures. Thermo-soluto capillary effects, buoyancy, density change upon phase transition induce natural convection in the melt, frequently leading to spatial and time-dependent non-uniform interfacial morphologies. Convection in the melt can be induced by external forcing means to gain better control over the interfacial morphologies, among which vibration is an effective way for inducing convection in the melt. There have been a number of experiments and simulations that indicate the effect of melt convection induced by vibration on nucleation and interfacial morphologies during solidification [1–7]. Sugiura et al. [8] investigated the refining mechanism of Sn-10mass%Pb alloy imposed in an electromagnetic vibration and confirmed that the electromagnetic vibration can induce the oscillatory flow in the melt, and the intense electromagnetic vibration induces nucleation in the melt and the refinement of solidified microstructures occurs in the initial stage of solidification. Mizutani et al. [9] and Zhang et al. [10] observed respectively the grain refinement effect under vibrations with various vibration frequency and acceleration and found that grain size decreases as vibration intensity increases. Lan et al. [11,12] performed the numerical simulations and revealed the effects of angular vibration on the flow, segregation and interface morphology in vertical Bridgman crystal growth. These experiments and simulations have gained deep insight into grain refinement during solidification, however, further theoretical investigation is still required to understand the mechanism of grain refinement. This paper aims to study the effect of oscillatory flow induced by vibration on nucleation and grain growth in the initial stage of solidification. By the matched asymptotic method and multiple-variable expansion method [13,14] we find the asymptotic solution for the temperature field and grain growth and explore the mechanism of grain refinement in the convective undercooled melt.

2. Model formulation

During solidification, nucleation occurs in the initial stage of transformation. As the liquid temperature decreases in the undercooled melt, the crystallite embryo of the solid phase from the meta-stable liquid phase occurs. The crystallite embryos are small transient clusters of the forming solid phase, dispersed in the liquid phase. The crystallite embryo may grow to enough size to form a nucleus or decay to vanish in the melt, which depends on its ambient change of temperature. Vibration exerted on the melt can induce the oscillatory flow in the melt, resulting in the change of temperature [8]. As the temperature

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decreases below the liquidus, the crystallite embryo grows and surpasses the critical nucleation radius and then a nucleus of the forming solid phase occurs. As the temperature further decreases, the nucleus continuously grows in size to form a grain. A large number of nuclei in the melt occur and grow in size until all the liquid phase is consumed. For the analytical analysis, we consider the formation of one nucleus as a particle in the undercooled melt. According to Mullins and Sekerka's view [15], the field that is several wavelengths away from the particle is taken as the far field. It is assumed that the far field temperature is T_∞ ($T_\infty < T_M$, T_M is the melting temperature for the pure substance) and an oscillatory flow far from the particle induced by melt vibration is in the downward direction pointing to the particle. For simplicity, the liquid phase and solid phase have equal densities and equal specific heats, and the buoyancy effects are neglected. $\mathbf{U}_L$ and P denote the convective velocity and pressure in the liquid phase, respectively. T_L and T_S denote the temperatures in the liquid phase and solid phase, respectively. $R = R(\theta, \varphi, t)$ denotes the interface of the particle in the spherical coordinate (r, θ, φ) whose origin is set at the center of the sphere. The interface of the particle separates the solid phase $r < R(\theta, \varphi, t)$ from the liquid phase $r > R(\theta, \varphi, t)$. The nucleation and grain growth in the undercooled melt are governed by the coupled continuity, momentum equation and heat conduction equations:

$$\nabla\cdot\mathbf{U}_L = 0, \tag{2.1}$$

$$(\mathbf{U}_L\cdot\nabla)\mathbf{U}_L = -\frac{1}{\rho_L}\nabla P + \upsilon\nabla^2\mathbf{U}_L, \tag{2.2}$$

$$\frac{\partial T_L}{\partial t} + (\mathbf{U}_L\cdot\nabla)T_L = \kappa_L\nabla^2 T_L, \tag{2.3}$$

$$\frac{\partial T_S}{\partial t} = \kappa_S\nabla^2 T_S, \tag{2.4}$$

where ρ_L is the density in the liquid phase, υ is the kinematical viscosity, κ_T and κ_S are respectively the thermal diffusivities in the liquid and solid phases, which are subject to the following boundary conditions.

At the interface, the total mass conservation condition and the tangential non-slip condition hold,

$$\rho_L(\mathbf{U}_L\cdot\mathbf{n} - U_I) = \rho_S(\mathbf{U}_S\cdot\mathbf{n} - U_I),\ \ \mathbf{U}_L\cdot\boldsymbol{\tau} = \mathbf{U}_S\cdot\boldsymbol{\tau}.$$

where ρ_S are the density in the solid phase, $\mathbf{n}$ and $\boldsymbol{\tau}$ are the exterior unit vectors normal and tangential to the interface, respectively. As the convective velocity in the solid phase is assumed to be constant, $\mathbf{U}_S = 0$, the densities in the liquid and solid phases are equal, the total mass conservation condition and the tangential non-slip condition are expressed as

$$\mathbf{U}_L\cdot\mathbf{n} = 0,\ \ \mathbf{U}_L\cdot\boldsymbol{\tau} = 0. \tag{2.5}$$

The interface conditions also include the thermal equilibrium condition, the Gibbs-Thomson condition and energy conservation condition:

$$T_L = T_S, \tag{2.6}$$

$$T_S = T_M\left(1 + \frac{2\gamma_0}{\Delta H}K\right) - \Delta T_K, \tag{2.7}$$

$$\Delta H U_{\mathrm{I}} = (k_S\nabla T_S - k_L\nabla T_L)\cdot\mathbf{n}, \tag{2.8}$$

where K is the local mean curvature at the interface, γ_0 is the isotropic surface energy, ΔT_K is the kinetic undercooling which is necessary for the attachment of atoms to the interface, $\Delta T_K = (1/\mu)U_{\mathrm{I}}$, μ is the interfacial kinetics coefficient, U_{I} is the normal growth velocity of the interface, k_L and k_S the heat conduction coefficients in the liquid phase and solid phase, respectively.

The far-field temperature condition is expressed as

$$T_L \to T_\infty\ as\ r \to \infty. \tag{2.9}$$

The oscillatory flow induced by melt vibration is expressed as a harmonic function of amplitude A and frequency Ω in the downward direction pointing to the particle,

$$\mathbf{U}_L \to -A_D\cos\Omega t\mathbf{k}\ as\ r \to \infty, \tag{2.10}$$

where A_D and Ω are the amplitude and frequency of the oscillatory flow, respectively, $A_D = A\Omega$ and $\mathbf{k}$ is the third unit vector of Cartesian rectangular coordinates.

We choose the initial radius of the particle r_0 as the length scale, the characteristic velocity of the interface $V = k_L\Delta T/(r_0\Delta H)$ as the velocity scale, r_0/V as the time scale, $\rho_L k_L V/r_0$ as the pressure scale and $\Delta H/(c_{pL}\rho_L)$ as the temperature scale, in which ΔH is the latent heat per unit volume, c_{pL} is the specific heat in the liquid phase. With the transformation

$$\overline{\mathbf{U}}_L = \frac{\mathbf{U}_L}{V} = \frac{1}{V}(u, v, w),\ \ \overline{P} = \frac{P}{\rho_L k_L V/r_0},$$

$$\overline{T}_L = \frac{T_L - T_M}{\Delta H/(c_{pL}\rho_L)},\ \ \overline{T}_S = \frac{T_S - T_M}{\Delta H/(c_{pL}\rho_L)},\ \bar{r} = \frac{r}{r_0},\ \bar{t} = \frac{t}{r_0/V}, \tag{2.11}$$

we transfer Eqs. (2.1)–(2.10) to the dimensionless equations. Further, we introduce a slow variable $\rho = \varepsilon\bar{r}$, where ε is the relative undercooling parameter,

$$\varepsilon = \frac{\Delta T}{\Delta H/(c_{pL}\rho_L)},$$

with $\Delta T = T_M - T_\infty$ being the undercooling in the melt. When the variables $\bar{r}, \theta, \varphi$ and ρ are formally treated as the independent variables,

$$\frac{\partial}{\partial\bar{r}} \to \frac{\partial}{\partial\bar{r}} + \varepsilon\frac{\partial}{\partial\rho},$$

the continuity equation, momentum equation and heat conduction equations in Eqs. (2.1)–(2.10) are expressed as the dimensionless equations

$$\overline{\nabla}\cdot\overline{\mathbf{U}}_L + \varepsilon\frac{\partial\bar{u}}{\partial\rho} = 0, \tag{2.12}$$

$$\begin{aligned}\varepsilon(\overline{\mathbf{U}}_L\cdot\overline{\nabla})\overline{\mathbf{U}}_L + \varepsilon^2\bar{u}\frac{\partial\overline{\mathbf{U}}_L}{\partial\rho} &= -\nabla\overline{P} - \varepsilon\left(\frac{\partial\overline{P}}{\partial\rho}, 0, 0\right)\\ &\quad + \mathrm{Pr}\left(\overline{\nabla}^2\overline{\mathbf{U}}_L + 2\varepsilon\frac{\partial^2\overline{\mathbf{U}}_L}{\partial r\partial\rho} + \varepsilon^2\frac{\partial^2\overline{\mathbf{U}}_L}{\partial\rho^2} + \varepsilon\frac{2}{r}\frac{\partial\overline{\mathbf{U}}_L}{\partial\rho}\right),\end{aligned} \tag{2.13}$$

$$\varepsilon\frac{\partial\overline{T}_L}{\partial t} + \varepsilon(\overline{\mathbf{U}}_L\cdot\overline{\nabla})\overline{T}_L + \varepsilon^2\bar{u}\frac{\partial\overline{T}_L}{\partial\rho} = \overline{\nabla}^2\overline{T}_L + 2\varepsilon\frac{\partial^2\overline{T}_L}{\partial r\partial\rho} + \varepsilon^2\frac{\partial^2\overline{T}_L}{\partial\rho^2} + \varepsilon\frac{2}{r}\frac{\partial\overline{T}_L}{\partial\rho}, \tag{2.14}$$

$$\varepsilon\lambda_S\frac{\partial\overline{T}_S}{\partial t} = \overline{\nabla}^2\overline{T}_S + 2\varepsilon\frac{\partial^2\overline{T}_S}{\partial r\partial\rho} + \varepsilon^2\frac{\partial^2\overline{T}_S}{\partial\rho^2} + \varepsilon\frac{2}{r}\frac{\partial\overline{T}_S}{\partial\rho}, \tag{2.15}$$

which are subject to the interface conditions:

$$\overline{\mathbf{U}}_L\cdot\mathbf{n} = 0,\ \ \overline{\mathbf{U}}_L\cdot\boldsymbol{\tau} = 0, \tag{2.16}$$

$$\overline{T}_L = \overline{T}_S, \tag{2.17}$$

$$\overline{T}_S = 2\varepsilon\Gamma\overline{K} - \varepsilon E^{-1}M\overline{U}_I, \tag{2.18}$$

$$\varepsilon\overline{U}_I = (k\overline{\nabla}\overline{T}_S - \overline{\nabla}\overline{T}_L)\cdot\mathbf{n} + \varepsilon\frac{\partial}{\partial\rho}(k\overline{T}_S - \overline{T}_L), \tag{2.19}$$

where $\bar{u}$ is the first component of $\overline{\mathbf{U}}_L = (\bar{u}, \bar{v}, \bar{w})$, Pr is the Prandtl number,

$$\mathrm{Pr} = \frac{\upsilon}{\kappa_L},\ \lambda_S = \frac{\kappa_L}{\kappa_S},\ K = \frac{\overline{K}}{r_0},\ \nabla = \frac{1}{r_0}\overline{\nabla},\ \overline{U}_I = \frac{U_I}{V},$$