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INFLUENCE OF SEED CRYSTAL DIMENSION ON CRYSTAL GROWTH FROM SOLUTION UNDER MICROGRAVITY

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The influence of the dimension of seed crystal on the characteristics of crystal growth from solution under microgravity is studied. The dimensionless average velocity of fluid $V_{\rm av}$, the dimensionless maximum velocity of fluid $V_{\rm max}$, the temperature distribution index S_θ, the concentration distribution index S_ϕ and the dimensionless average growth rate of crystal $\overline{V}_{\rm cg}$ are calculated by only taking into account the variation of the solution density caused by the temperature change, that caused by the concentration change being neglected. In certain regions of the parameter (Ra, Pr, Sc and μ) space, some scaling laws are generated: the scales of S_ϕ and $\overline{V}_{\rm cg}$ are given by power functions of μ with negative exponents. It is shown that the characteristics of crystal growth for small seed crystal are different from those for large seed crystal.

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I. INTRODUCTION

Great attention has been paid to the problems of crystal growth in space recently. Eckert *et al.*[1] have reviewed the problems of numerical computation of crystal growth in space. In a series of our previous papers[2−12], the characteristics of crystal growth from solution under microgravity were analyzed in detail with a cuboidal container model. In Refs.[3–11], a flat plate of crystal was chosen to form one wall of the container (the plane of $x = 0$) in the model. But in Ref.[2], two cases were taken into account. The first case was the same as in Refs.[3–11]. In the second case, that wall was not made of the crystal, but of the same material as that of the top and bottom wall except that a narrow strip of width d in the middle of the wall was still of the crystal. This corresponds to the case of small seed crystal. In Ref.[2], the two cases were compared, but the width of the crystal strip was not changed. In this paper, the influence of the relative dimension of seed crystal on crystal growth from solution under microgravity will be studied by only taking into account the variation of the solution density caused by the temperature change, that caused by the solute concentration change being neglected. This neglect is valid under the condition of

$$\left|\left(\frac{\partial\rho}{\partial T}\right)_0 (T - T_0)\right| \gg \left|\left(\frac{\partial\rho}{\partial c}\right)_0 (c - c_0)\right|, \qquad (1)$$

where ρ is the solution density, T is the solution temperature, c is the solute concentration, c_0 is a suitable concentration between c_s and c_i (see below), and T_0 is the corresponding temperature. It can be shown by our numerical calculation that when the ratio of the two terms of the inequality (1) is equal to or larger than 50:1, the error which results from the neglect is less than 1%. So it is a quite accurate approximation.

The model employed here is the same as that used in case Ⅱ of Ref.[2]. A cuboidal container is filled with the parent solution. The dimensions of the container and the strip of seed crystal in the z-direction is assumed to be long enough to simplify the mathematical problem as a two-dimensional problem. The edges of the container, designated as the x-axis and y-axis, are a and b in length, respectively. The left wall of the container (the plane of x=0) is made of the same material as that of the top and bottom wall except that a narrow strip of seed crystal of width d is placed in the middle of the wall. The gravity orients to the y-direction.

The concentration of the solute at the growth face of crystal is taken as c_s, the equilibrium solubility at the temperature of T_s, while at the surface of the opposite wall (the plane of $x = a$), which is made of the material of solute source, the solute concentration is taken as c_i, the equilibrium solubility at the temperature of T_i. The condition $c_i > c_s$ can be realized by choosing suitable temperatures T_s and T_i. Figure 1 illustrates the Cartesian coordinate system for the cuboidal container model.

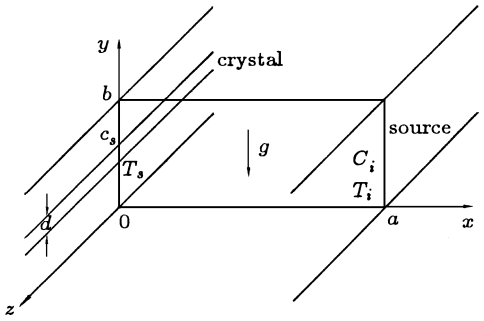


Fig.1. Coordinate system for the model used in the text.

Ⅱ. EQUATIONS OF MOTION

The Navier-Stokes equation, the continuity equation and the equation of motion for convection-diffusion have been given in Ref.[4], neglecting the variation of solution density caused by the solute concentration change, that is, letting α=0 in Eq.(5) of Ref.[4]. It is convenient to introduce the stream function ψ and the vorticity ξ.[2] By introducing dimensionless parameters λ, μ, dimensionless variables $X, Y, \boldsymbol{V}$ and dimensionless functions Ψ, Ξ, θ, ϕ as follows[13,14]:

$$\lambda = b/a, \quad \mu = d/b, \tag{2}$$

$$X = x/a, \quad Y = y/a, \tag{3}$$

$$\boldsymbol{V} = (a/\chi), \boldsymbol{v}, \tag{4}$$

$$\Psi = (1/\chi)\psi, \tag{5}$$

$$\Xi = (a^2/\chi)\xi, \tag{6}$$

$$\phi = (c_i - c)/(c_i - c_s), \tag{7}$$

$$\theta = (T_i - T)/(T_i - T_s), \tag{8}$$

where $\boldsymbol{v}$ is the fluid velocity, χ is the thermal diffusion coefficient,and transforming the equations of motion into their dimensionless form, we obtain

$$\nabla_R^2 \Xi = \frac{1}{Pr}\left(\frac{\partial \Psi}{\partial Y}\frac{\partial \Xi}{\partial X} - \frac{\partial \Psi}{\partial X}\frac{\partial \Xi}{\partial Y}\right) - Ra\frac{\partial \theta}{\partial X}, \tag{9}$$

$$\nabla_R^2 \theta = \frac{\partial \Psi}{\partial Y}\frac{\partial \theta}{\partial X} - \frac{\partial \Psi}{\partial X}\frac{\partial \theta}{\partial Y}, \tag{10}$$

$$\nabla_R^2 \Psi = \Xi, \tag{11}$$

$$\nabla_R^2 \phi = \frac{Sc}{Pr}\left(\frac{\partial \Psi}{\partial Y}\frac{\partial \phi}{\partial X} - \frac{\partial \Psi}{\partial X}\frac{\partial \phi}{\partial Y}\right), \tag{12}$$

where the operator ∇_R^2 is defined by

$$\nabla_R^2 = \frac{\partial^2}{\partial X^2} + \frac{\partial^2}{\partial Y^2}. \tag{13}$$

While the Rayleigh number Ra, the Prandtl number Pr and the Schmidt number Sc are defined as follows:

$$Ra = a^3 \beta g_0 g (T_i - T_s)/(\nu\chi), \tag{14}$$

$$Pr = \nu/\chi, \tag{15}$$

$$Sc = \nu/D, \tag{16}$$

where ν is the kinematic viscosity, D is the solute diffusion coefficient, g_0 is the terrestrial acceleration of gravity, g is the gravity level and β is the thermal expansion coefficient, which is given by

$$\beta = -\frac{1}{\rho_0}\left(\frac{\partial \rho}{\partial T}\right)_0, \tag{17}$$

where

$$\rho_0 = \rho(c_0, T_0). \tag{18}$$

The boundary conditions associated with Eqs.(9)–(12) are similar to those in Refs.[2] and [11]. They are expressed by

$$\Psi(0,Y) = \Psi(1,Y) = \Psi(X,0) = \Psi(X,\lambda) = 0, \tag{19}$$

$$\left.\frac{\partial \Psi(X,Y)}{\partial X}\right|_{X=0} = \left.\frac{\partial \Psi(X,Y)}{\partial X}\right|_{X=1} = \left.\frac{\partial \Psi(X,Y)}{\partial Y}\right|_{Y=0} = \left.\frac{\partial \Psi(X,Y)}{\partial Y}\right|_{Y=\lambda} = 0, \tag{20}$$

$$\theta(0,Y) = 1, \quad \theta(1,Y) = 0, \tag{21}$$

$$\theta(X,0) = \theta(X,\lambda) = 1 - X, \tag{22}$$

$$\left.\frac{\partial \phi(X,Y)}{\partial Y}\right|_{Y=0} = \left.\frac{\partial \phi(X,Y)}{\partial Y}\right|_{Y=\lambda} = 0, \tag{23}$$

$$\left.\frac{\partial \phi(X,Y)}{\partial X}\right|_{X=0} = 0, \qquad \text{if} \quad Y < \frac{\lambda(1-\mu)}{2} \quad \text{or} \quad Y > \frac{\lambda(1+\mu)}{2}, \tag{24}$$

$$\left.\frac{\partial \phi(X,Y)}{\partial X}\right|_{X=0} = K(\phi - 1), \qquad \text{if} \quad \frac{\lambda(1-\mu)}{2} \le Y \le \frac{\lambda(1+\mu)}{2}, \tag{25}$$

$$\left.\frac{\partial \phi(X,Y)}{\partial X}\right|_{X=1} = -K'\phi, \tag{26}$$

where $K$and K' are the dimensionless interface kinetics coefficients[11−15], $K = ka/D$ and $K' = k'a/D$, k is the linear interface kinetics coefficient for crystallization and k' is that for dissolution. Here, the interface kinetics of both crystallization and dissolution at the planes of $X = 0$ (for $\lambda(1-\mu)/2 \le Y \le \lambda(1+\mu)/2$) and $X = 1$ is taken into account and the diffusion-reaction theory with finite interface kinetics coefficient is used.[15]

Ⅲ. CALCULATION AND RESULTS

The partial differential Eqs.(9)–(11) with the boundary conditions of Eqs.(19)–(22) constitute a complete set which can be solved. The equations are reduced to a large set of difference equations and the numerical solutions to the equations are computed using an iteration method with relaxation parameters.[16] Then, with the obtained solution of $\Psi(X,Y)$, Eq.(12) with the boundary conditions of Eqs.(23)–(26) can be solved in a similar procedure. In computation, the parameters are taken as follows: $\lambda = 0.2$; $\mu = 0.05$–1; $Ra = 6\times10^3$, 6×10^4, 6×10^5; $Sc = 10$, 10^2, 10^3; $Pr = 0.6$, 6, 60. Crystals usually dissolve much faster than they grow, and dissolution rates are much greater than growth rates under equal driving force (the deviation of concentration from the equilibrium solubility); five-fold differences are not uncommon.[17] Therefore, the following values are taken in our computation: K=500, K'=2500. In order to describe the temperature distribution and concentration distribution compactly, we define an index S_ζ for each function $\zeta(X,Y)$(ζ stands for θ or ϕ)as follows:

$$S_\zeta = \frac{1}{\lambda}\int_0^1 \mathrm{d}X \int_0^\lambda \mathrm{d}Y|\zeta(X,Y) - \zeta_\mathrm{d}(X,Y)|, \quad (27)$$

where
$$\zeta_\mathrm{d}(X,Y) = 1 - X, \quad (28)$$

which represents the temperature distribution or concentration distribution for Ra=0, that is, there exists only diffusion without convection. The S_θ and S_ϕ are two composite indexes which represent synthetically the deviation of the temperature distribution and the concentration distribution from $\theta_\mathrm{d}(X,Y)$ and $\phi_\mathrm{d}(X,Y)$, respectively.

In addition to S_θ and S_ϕ, the dimensionless average fluid velocity V_av, dimensionless maximum fluid velocity V_max and dimensionless average growth rate of crystal $\overline{V}_\mathrm{cg}$ are computed. The $\overline{V}_\mathrm{cg}$ is given by[6,7]

$$\overline{V}_\mathrm{cg} = -\frac{\eta}{\lambda}\frac{Pr}{Sc}\int_0^\lambda \left.\frac{\partial\phi(X,Y)}{\partial X}\right|_{X=0}\mathrm{d}Y, \quad (29)$$

where

$$\eta = (c_i - c_s)/\rho_\mathrm{c}, \quad (30)$$

ρ_c being density of the growing crystal.

It has been shown in Ref.[11] that the interface kinetics does not affect the scaling laws of the fluid velocity and the temperature distribution index S_θ because the boundary conditions of Eqs.(23)–(26) have nothing to do with Eqs.(9)–(11), so the change of the function ϕ does not affect the solutions of Ψ, Ξ and θ. For the same reason, the dimension of the seed crystal does not affect the scaling laws of V_av, V_max and S_θ.

Figure 2 shows how S_ϕ depends on μ for $\lambda = 0.2$, $Pr = 0.6$, $Sc = 10^2$ and different values of Ra. It can be seen from Fig.2 that S_ϕ is a power function of μ with a negative exponent in a certain range of small values of μ, the dimensionless width of the seed crystal. This range for large value of Ra is greater than that for small one, and the scaling law of S_ϕ for $Ra = 6\times10^3$ is valid only in a quite small range of μ values.

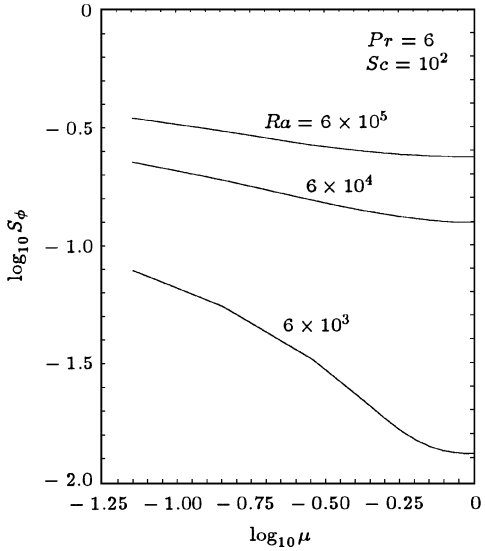


Fig.2. Plot of $\log_{10}S_\phi$ versus $\log_{10}\mu$ for three values of Ra and Pr=6, $Sc = 10^2$.

Figures 3 and 4 show the curves of $\log_{10}(\overline{V}_\mathrm{cg}/\eta)$ versus $\log_{10}\mu$ for λ=0.2, Pr=6 and different values of Ra and Sc. It is known from Figs.3 and 4 that the scaling law of $\overline{V}_\mathrm{cg}$ is valid in a certain range of μ values. The $\overline{V}_\mathrm{cg}$ is a power function of μ with a negative exponent in a wide range of μ values for $Ra = 6\times10^3$ and $Sc = 10^2$. The range of μ values in which the scaling law of $\overline{V}_\mathrm{cg}$ is valid, is greater for small values of Ra or Sc than for large ones. The scaling law of $\overline{V}_\mathrm{cg}$ for $Ra = 6\times10^5$, $Sc = 10^2$ and for $Ra = 6\times10^4$, $Sc = 10^3$ is valid only in a quite small range of μ values.

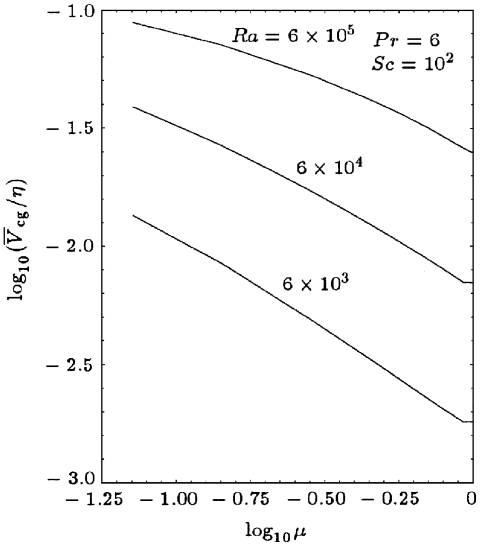


Fig.3. Plot of $\log_{10}(\overline{V}_{cg}/\eta)$ versus $\log_{10}\mu$ under the same conditions as for Fig.2.

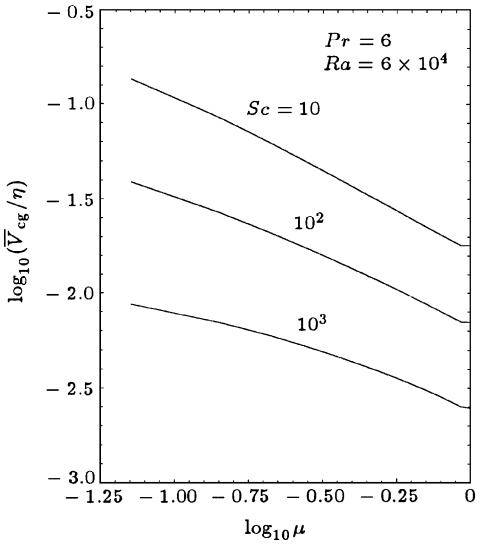


Fig.4. Plot of $\log_{10}(\overline{V}_{cg}/\eta)$ versus $\log_{10}\mu$ for three values of Sc and $Pr = 6$, $Ra = 6 \times 10^4$.

IV. DISCUSSION AND CONCLUSION

Finally, we come to a conclusion that the dimension of seed crystal does affect the characteristics of crystal growth from solution under microgravity. The S_ϕ and $\overline{V}_{cg}$ depend on μ, but S_θ, V_{av} and V_{max} do not.

In certain regions of the parameter (Ra, Pr, Sc and μ) space, some scaling laws are generated: the scales of the concentration distribution index S_ϕ and the average growth rate of crystal $\overline{V}_{cg}$ are given by power functions of μ with negative exponents.

The S_ϕ represents the deviation of the concentration distribution $\phi(X, Y)$ from $\phi_d(X, Y)$, the concentration distribution for the case that there exists only diffusion without convection, and only the boundary condition of ϕ at the plane of X=0 depends on the value of μ. Therefore, the solute diffusion at the plane of X=0 changes with the value of μ, but the latter does not affect directly the fluid convection in the bulk. The larger the Rayleigh number Ra, that is, for instance, the larger the gravity level g, the larger the temperature difference $T_i - T_s$; or the smaller the viscosity ν, the stronger the fluid convection. It follows that when the value of Ra is large, S_ϕ is a power function of μ with a negative exponent in a large range of μ values; when the value of Ra is small, that scaling law of S_ϕ is valid only in a quite small range of μ values. The dimensionless average growth rate of orystal $\overline{V}_{cg}$ depends strongly on boundary condition of ϕ at the plane of X=0. Therefore, the behavior of the scaling law of $\overline{V}_{cg}$ is quite different from that of the scaling law of S_ϕ.

In the range of small values of μ, the scaling laws are valid in rather wide regions of parameter (Ra, Pr and Sc) space. But only in narrow regions of parameter (Ra, Pr and Sc) space, the scaling laws can be valid in a wide range of μ values. This means that the characteristics of crystal growth for small seed crystal are different from those for large seed crystal.

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