



EFFECTS OF THE ORIENTATION OF MICROGRAVITY ON THE CONCENTRATION DISTRIBUTION IN CRYSTALLIZATION FROM SOLUTION

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ABSTRACT

A quasi-steady state growth and dissolution in a 2-D rectangular enclosure is numerically investigated. This paper is an extension to indicate the effects of the orientation of gravity on the concentration field in crystallization from solution under microgravity, especially on the lateral non-uniformity of concentration distribution at the growth surface. The thermal and solute convection are included in this model.

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1. INTRODUCTION

The solute concentration distribution at the crystal surface has a profound influence on the structure and the quality of the crystal growing from solution. Thus, it is necessary to have a thorough understanding of the concentration distribution at the crystal surface for optimization of crystal growth process design and crystal quality. In our previous paper (Wang and Hu, 1996), we numerically examined the effects of the microgravity level on the lateral non-uniformity of concentration distribution at the growth surface using the diffusion-reaction theory with finite interface kinetics coefficient, while the microgravity orientation is fixed on the direction parallel to the growth interface. However, in space many factors such as atmospheric drag, basic orbital attitude motions and attitude corrections can result in the change of not only the microgravity level but also the microgravity orientation (Iwan and Alexander *et al.*, 1989). Xu *et al.* (1993) have numerically studied the effects of the orientation of microgravity on the solute concentration field using the pure diffusion model to deal with the growth process. It is impossible for their work to reveal the characteristics of the supersaturation and the non-uniformity at the growth surface.

This present investigation extends our work (Wang and Hu, 1996) by further considering the change of the microgravity orientation and its effect on the concentration field in solution crystal growth, especially on the lateral non-uniform solute distribution at the growth surface. We believe the results obtained here will be helpful for the proper design of experimental conditions for crystal growth under microgravity. With the thermal and solute convection, the concentration field is obtained by numerically solving the Navies-Stokes equations, mass convection-diffusion equation and heat transfer equation.

2. MATHEMATICAL MODEL AND NUMERICAL SIMULATION

The physical system considered here involves solution crystal growth via temperature gradient technique. In order to simplify an actual crystal growth system and retain the essential physical features of the system, similar to reference (Wang and Hu, 1996), we consider a quasi-steady state growth and dissolution in a 2-D rectangular enclosure, in which one side wall is the growth surface of crystal and the opposite one the dissolution surface, as a numerical model as depicted in Figure 1. Sodium chlorate (NaClO_3) aqueous solution is contained inside the

cavity. The left vertical source interface of the cavity is maintained at a higher temperature T_s^* compared to T_c^* at the right vertical seed crystal interface. In view of the lower growth and dissolution rates comparing with the typed flow velocity, and our focal attention on the influence of the buoyancy-induced convection on the concentration field, we simplify the realistic moving source and seed interfaces as the fixed plates.

By introducing the stream function ψ and the vorticity ω , with the Boussinesq approximation, the steady governing equations in dimensionless form appear as follow

$$\begin{cases} \frac{1}{Pr} \nabla \cdot (V \omega) = \nabla^2 \omega + R_{ac} \left(\frac{\partial \phi}{\partial X} \cos \gamma + \frac{\partial \phi}{\partial Y} \sin \gamma \right) - R_{at} \left(\frac{\partial \mathcal{G}}{\partial X} \cos \gamma + \frac{\partial \mathcal{G}}{\partial Y} \sin \gamma \right) \\ \nabla \cdot (V \phi) = \frac{1}{Le} \nabla^2 \phi \\ \nabla \cdot (V \mathcal{G}) = \nabla^2 \mathcal{G} \\ \nabla^2 \psi = \omega \end{cases} \quad (2.1)$$

where V , \mathcal{G} and ϕ are the dimensionless velocity, temperature and concentration, respectively. γ is the orientation angle between the vector of gravity and the -Y axis. \mathcal{G} and ϕ are defined as $\mathcal{G} = (T^* - T_c^*) / (T_s^* - T_c^*)$ and $\phi = (C^* - C_c^*) / (C_s^* - C_c^*)$, respectively, where C_s^* and C_c^* are the equilibrium solubility at T_s^* and T_c^* , respectively. The characteristic parameters used for non-dimensionalization are a for length and $V_0 = \kappa / a$ for velocity (where κ is the thermal diffusivity). Then, the thermal Rayleigh number R_{at} , solute Rayleigh number R_{ac} , Prandtl number Pr and Lewis number Le become

$$\begin{cases} R_{at} = \frac{\beta a^3 g g_0 (T_s^* - T_c^*)}{\nu \kappa}, & R_{ac} = \frac{\alpha a^3 g g_0 (C_s^* - C_c^*)}{\nu \kappa} \\ Pr = \frac{\nu}{\kappa}, & Le = \frac{\kappa}{D} \end{cases} \quad (2.2)$$

where ν is kinematic viscosity, D is diffusion coefficient, g is terrestrial gravity acceleration, g_0 is the ratio of local gravity to earth's gravity, $\alpha = \frac{1}{\rho^*} \frac{\partial \rho^*}{\partial C^*}$ is solutal expansion coefficient and $\beta = -\frac{1}{\rho^*} \frac{\partial \rho^*}{\partial T^*}$ is thermal expansion coefficient.

The top and bottom walls are adiabatic and impermeable. The linear diffusion-reaction theory with finite interface kinetics coefficient is used to describe the growth process. The solution concentration at source interface is assumed as C_s^* .

Then, the boundary conditions may be given in the form of

$$\begin{cases} X=0, & V_x = V_y = 0, & \mathcal{G} = 1, & \phi = 1 \\ X=1, & V_x = V_y = 0, & \mathcal{G} = 0, & \frac{\partial \phi}{\partial X} = -K_c \phi \\ Y=0, & V_x = V_y = 0, & \frac{\partial \mathcal{G}}{\partial Y} = \frac{\partial \phi}{\partial Y} = 0 \\ Y=\lambda, & V_x = V_y = 0, & \frac{\partial \mathcal{G}}{\partial Y} = \frac{\partial \phi}{\partial Y} = 0 \end{cases} \quad (2.3)$$

where $\lambda = b/a$ is the aspect ratio of the cavity, $K_c = \frac{k_c a}{D}$ the dimensionless crystal interface kinetics coefficient, k_c the crystal interface kinetics coefficient.

The Eq. (2.1) associated with boundary conditions Eq. (2.3) are numerically solved using the finite difference method. Near the source and crystal interfaces we used a finer grid than in the bulk solution. More detailed description of our simulation can be seen in reference (Wang and Hu, 1996).

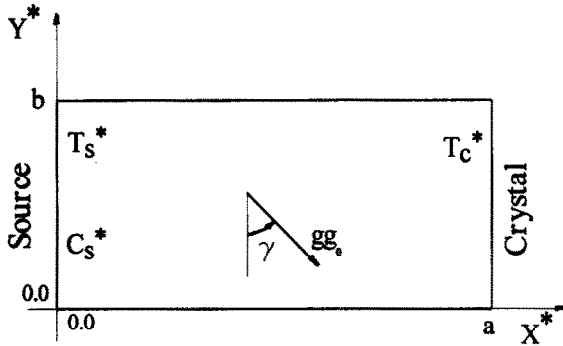


Fig. 1. Schematic representation of the physical and coordinate systems.

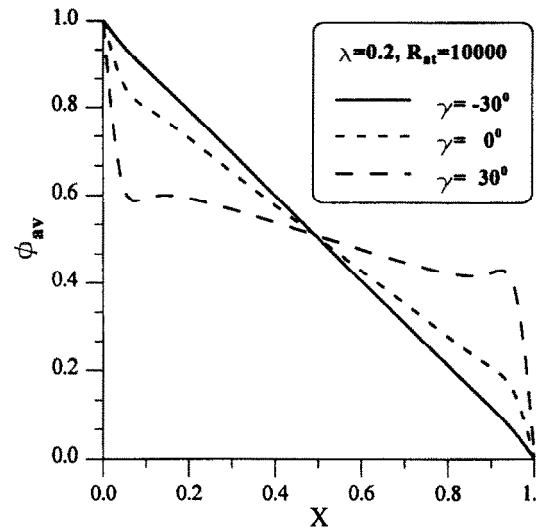


Fig. 2. Laterally averaged dimensionless concentration ϕ_{av} versus X for $\lambda = 0.2$, $R_{at} = 10000$.

3. RESULTS

The thermophysical properties and operation conditions used in this work are the same as that in the reference (Wang and Hu, 1996). R_{at} and R_{ac} are behalf on the gravity level at constant temperature difference. Thus, we have a relationship as follow $R_{at} = 5.175 \times 10^7 g_0$.

Figure 2 reveals the effects of the gravity orientation on the concentration field in the cavity, in which $\phi_{av} = \int_0^1 \phi(X, Y) dY / \lambda$. For case $\gamma = -30^\circ$, the solute transport is mainly controlled by diffusion process. As γ is raised, convection becomes stronger. When $\gamma = 30^\circ$, there are considerably steeper interfacial concentration gradients, and the concentration distribution in the bulk solution tends to uniformity. Figure 3 shows the concentration distribution at the growth interface for various gravity orientations γ , which is our focus in this work. One sees that the concentration distribution is non-uniform across the crystal face. As expected, the supersaturation in the region facing the incoming solution is higher than that in the other region. For $\gamma = -30^\circ$, concentration changes gradually along the face, and the value of supersaturation is smaller. As γ is raised, supersaturation overall the interface first increases, and the increment in the region facing the incoming solution is greater. For case $\gamma = 30^\circ$, stronger convection causes an evident non-uniformity of supersaturation, and the position of maximum supersaturation moves upper from the corner of the cavity. This plot also shows that the maximum supersaturation at the crystal interface does not increase as γ further increases. In this paper, we also define the measure $\zeta = (\phi_{cmax} - \phi_{cmin}) / \phi_{ac}$ to characterize the non-uniformity of the supersaturation at the growth face as in reference (Iwan and Alexander *et al.*), where ϕ_{cmax} , ϕ_{cmin} and $\phi_{ac} = \int_0^1 \phi_c dY / \lambda$ are the maximum, minimum and average concentration at the growth interface, respectively. The effects of the gravity orientations γ on the non-uniformity of the supersaturation ζ for various R_{at} and λ can be seen in Figure 4. As indicated in Figure 4, ζ asymmetrically changes with γ , and due to weaker convection, the smaller R_{at} and/or the smaller λ , the smaller the effects of γ on ζ . Furthermore, one sees that in the range of

$-80^\circ < \gamma < 80^\circ$, there is a maximum measure ζ_{\max} at a certain orientation γ_c depending on R_{at} and λ . As R_{at} increases, γ_c first increases, then decreases to a value near 0° after reaching maximum. In addition, γ_c in the case $\lambda = 0.4$ is smaller than that in the case $\lambda = 0.2$.

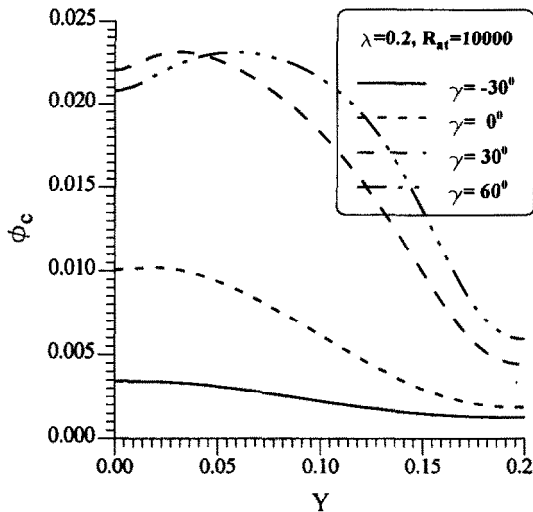


Fig. 3. Lateral dimensionless concentration ϕ_c versus Y for $\lambda = 0.2$, $R_{at} = 10000$.

4. SUMMARY

In this paper, a quasi-steady state growth and dissolution via temperature gradient technique in a 2-D rectangular enclosure filled with sodium chlorate (NaClO_3) aqueous solution is chosen as a model, and the effects of microgravity orientation on the solute concentration field in crystallization from solution, especially on the lateral non-uniformity of concentration distribution at the growth surface, are numerically investigated. Our results indicate that for small R_{at} and λ , the effects of the gravity orientation γ on the concentration field in the solution region and the concentration distribution at the growth interface is so small that can be neglected. But as R_{at} and λ are raised, concentration field remarkably changes along with γ . In the range of $-80^\circ < \gamma < 80^\circ$, as γ is increased, due to stronger convection, the interface concentration gradient becomes steeper. Moreover, there is a maximum measure of the non-uniformity of supersaturation ζ_{\max} at the growth surface at a certain angle γ_c depending on R_{at} and λ . For large R_{at} and/or large λ , γ_c is the minus value near to 0° .

In our present work, due to the absence of the kinetic coefficient for the dissolution process, we treated the dissolution process as pure diffusion process. And for convenience we assumed the real moving interfaces to be fixed. Next paper will exhibit evidence that the results obtained with this simple operation is nearly similar to that of more realistic model associating with the diffusion-reaction theory and moving interfaces.

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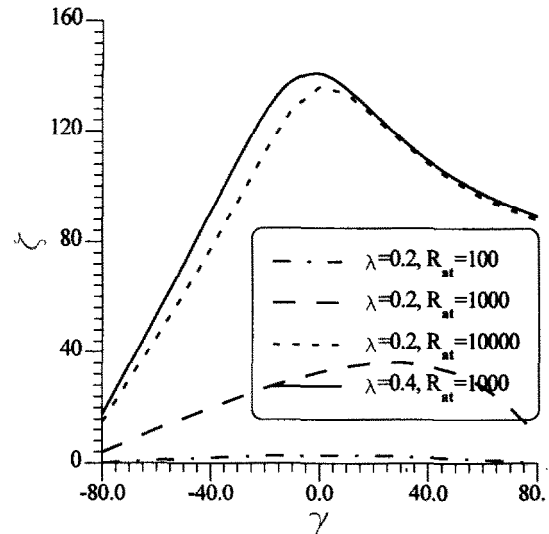


Fig. 4. The measure of concentration non-uniformity ζ at the growth interface versus the gravity orientation γ .