Investigation of the Convection Effect on the Inclusion Motion in Thermally Stressed Crystals

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Abstract. The conditions for occurrence of convective mass transfer in a liquid inclusion located in a thermally stressed crystal have been investigated. An estimated calculation of the Rayleigh number for a convective cell with solid boundaries has been performed. It is shown that the calculated value of the Rayleigh number can exceed the critical value. For such conditions, we have obtained analytical expressions for the perturbed velocity and temperature of a cylindrical convective cell with solid boundaries that exist in the inclusion. The theoretical model of induced transitions of atoms of the crystal matrix into solution and back is proposed, taking into account convective mass transfer.

Introduction

It is known that crystallization in a fluid medium (solutions, melts, high-density gaseous media) can be accompanied by convection, when heat and/or substance are trans-ported by hydrodynamic flow. Many modern technologies related to crystal growth inside a fluid medium have problems with convective heat and mass transfer control. The quality of grown crystals determined by the perfection of their structure and stoichiometry depends on understanding the mechanisms of convection during crystallization and the possibility to control it. The problem of controlling convective flows has become particularly important in connection with the growth of protein crystals from biological solutions. Protein crystallization plays a vital role in structural biology, protein purification, drug transport, etc. High-quality crystals are required for X-ray studies of protein molecules, which are essential for understanding protein functions.

3 Determination of the Parameters of the Viscous Incompressible Liquid Layer, at which Convection Can Occur

$$R = \frac{g^* \beta A h^4}{\nu \chi}$$

where $\nu = \eta / \rho$ is the kinematic viscosity of a liquid, determined by the values of its dynamic viscosity η and density ρ, $\chi = \kappa / \rho c_p$ is the thermal diffusivity of a liquid, which, in addition to density, also depends on thermal conductivity κ and specific heat capacity c_p, $\beta = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_p$ is the coefficient of thermal expansion, g^* is the "effective" acceleration acting on the element of liquid similar to the acceleration due to gravity.

$$\beta = 5.3 \cdot 10^{-4} K^{-1}, \chi = 1.74 \cdot 10^{-7} m^2/s,$$
$$\eta = 4.22 \cdot 10^{-4} Pa*s, \nu \approx 3.5 \cdot 10^{-7} m^2/s.$$

- Substituting these values into the expression for the Rayleigh number, we find that the order of magnitude for the investigated conditions corresponds to the critical Rayleigh number for the occurrence of convection with solid boundary conditions R ≈10 [4]

4 Basic Equations Describing Convection in Layers of Viscous Incompressible Liquid Bounded by Perfectly Heat-Conducting Solid Medium

$$\mathrm{v}_z(r,z,t) = \mathrm{v}(z) J_0(k_r r)\exp(-\lambda t),$$
$$T(r,z,t) = \vartheta(z) J_0(k_r r)\exp(-\lambda t),$$

$$(q^2-b)^3 = -a^3$$

$$q_{1,2} = \pm\sqrt{b-a};\ q_{3,4} = \sqrt{b+0.5a(1+i\sqrt{3})} = \pm(X_+ + iX_-);$$
$$q_{5,6} = \sqrt{b+0.5a(1-i\sqrt{3})} = \pm(X_+ - iX_-),$$

$$\mathrm{v}(z) = \sum_{m=1}^{6} C_m \exp(q_m z),$$

$$\mathrm{v}(0) = \mathrm{v}(1) = 0,\ \partial \mathrm{v}(0)/\partial z = \partial \mathrm{v}(1)/\partial z = 0,\ \vartheta(0) = \vartheta(1) = 0.$$

$$\mathrm{v}(z) = A_1\left[1 - ch\left(\left(z-\frac{1}{2}\right)X_+\right)ch^{-1}\left(\frac{1}{2}X_+\right)\right]\sin(n\pi z).$$

$$R_1^{rigid} = \alpha_1 \frac{\left(\pi^2 + (\beta_1 \cdot k_r)^{\mu_1}\right)^3}{(\beta_1 \cdot k_r)^{\mu_1}},$$

$$\vartheta(z) = -\frac{1}{\sqrt{b}}\int_0^z \mathrm{v}(\xi)\, sh\left(\sqrt{b}(\xi - z)\right)d\xi + \frac{1}{\sqrt{b}}\frac{sh(z\sqrt{b})}{sh(\sqrt{b})}\int_0^1 \mathrm{v}(\xi)\, sh\left(\sqrt{b}(\xi-1)\right)d\xi.$$

7 A Small Temperature Gradient.

$$\frac{\partial G}{\partial t} + V_z(r,z)\frac{\partial G}{\partial z} = \frac{\partial G}{\partial t} + \tilde{z}^2 A_1 \pi X_+ th\left(\frac{1}{2}X_+\right)\frac{\partial G}{\partial \tilde{z}}$$

where G takes values n_1, n_2, N.

$$\frac{\partial n_1}{\partial \xi} = \mu'(n_2 - n_1)N,\quad \frac{\partial n_2}{\partial \xi} = -\mu'(n_2 - n_1)N,\quad \frac{\partial N}{\partial \xi} = -\mu'(n_2 - n_1)N,$$

where A isapositiveconstant determined by the conditions of the problem, $\mu' = \mu_0 \lambda^{-1}$,

$\lambda = \left(1 + AA_1\pi X_+ th\left(\frac{1}{2}X_+\right)\right) > 0.$ $\xi(\tilde{z}, t) = t - A\tilde{z}^{-1} = C$ where C isaconstant.

$$V_S = \frac{d\tilde{z}}{dt} = -\frac{\tilde{z}^2}{A},$$

where $V_s = \frac{d\tilde{z}}{dt}$ is the velocity of theplaneofconstantphase. Expression (17) isvalid wherever $\tilde{z} \neq 0$ .If we assume $z = h$, then V_s determines the velocity of the plane of constant phase of the back inclusion wall.

$$V(h) = V_0 - \frac{(h-h_0)^2}{Ah_0^2} = \alpha + \beta h + \delta h^2,$$

where $\alpha = V_0 - \frac{1}{A}$, $\beta = \frac{2}{Ah_0}$, $\delta = \frac{1}{Ah_0^2}$

5 Influence of Convective Mass Transfer on the Velocity of Inclusion Motion

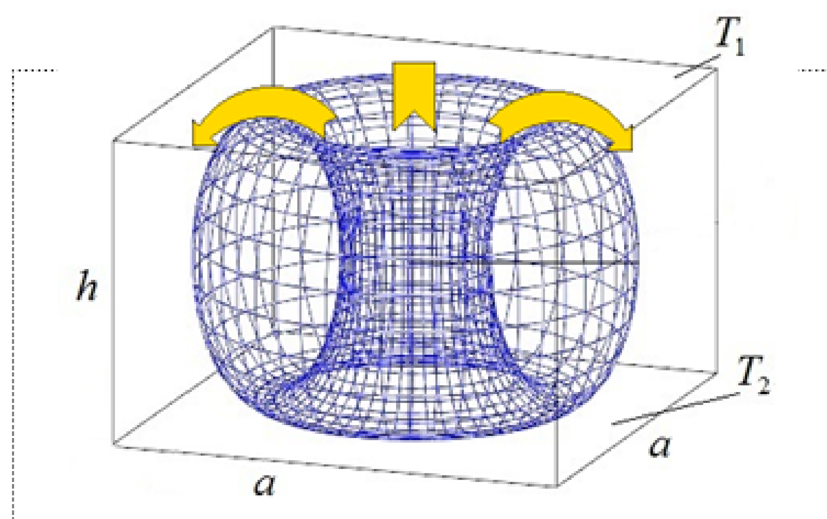


Fig. 1. Diagram of a cylindrical convective cell in the inclusion located in the temperature gradient field. The arrows show the direction of liquid flow in the cell.

The presence of convective mass transfer in the inclusion with velocities $\mathrm{v}_z(r,z,0)$ and $\mathrm{v}_r(r,z,0)$ creates solution flows washing the front and back inclusion surfaces. The rate of atoms emission/assimilation by the crystal surfaces depends on the temperature and its gradient. Therefore, in thermally stressed crystals, the convective process can accelerate the dissolution/growth processes on the inclusion surfaces,which can affect the velocity of its motion.

The processes of transitions of crystal atoms into solution and back have a probabilistic nature. Therefore, to describe such transitions, similarly to the transitions in crystals with inhomogeneous defect distribution, a two-level model can be used.

6 Two-Level Model of Induced Transitions of Crystal Atoms into Solution and Back

$$\frac{\partial n_1}{\partial t} + V_r(r,z)\frac{\partial n_1}{\partial r} + V_z(r,z)\frac{\partial n_1}{\partial z} = \mu_{12}(r,z)(n_2 - n_1)N,$$
$$\frac{\partial n_2}{\partial t} + V_r(r,z)\frac{\partial n_2}{\partial r} + V_z(r,z)\frac{\partial n_2}{\partial z} = -\mu_{12}(r,z)(n_2 - n_1)N,$$
$$\frac{\partial N}{\partial t} + V_r(r,z)\frac{\partial N}{\partial r} + V_z(r,z)\frac{\partial N}{\partial z} = -\mu_{12}(r,z)(n_2 - n_1)N,$$

where $V_r(r,z) = \mathrm{v}_r(r,z,0)$, $V_z(r,z) = \mathrm{v}_z(r,z,0)$; $\mu_{12}(r,z)$ is the probability of induced transitions of atoms between energy levels, in the general case, a function of coordinates r, z.

We consider two cases of dependence of the probability of induced transitions on coordinate z: a small temperature head ($\Theta << T_1$) or gradient, when the probability of induced transitions is a constant value - $\mu(z) = \mu_0 = \text{const}$; a great temperature head ($\Theta >> T_1$) or gradient, when the probability of induced transitions is a linear function of z, i.e. $\mu(z) = \mu_1 + \mu_2 z, |\mu_2| << \mu_1$.

Fig. 1. Dependence of the maximum inclusion velocity on their size at a small temperature gradient ($A \approx 8 \cdot 10^2$ K/m).

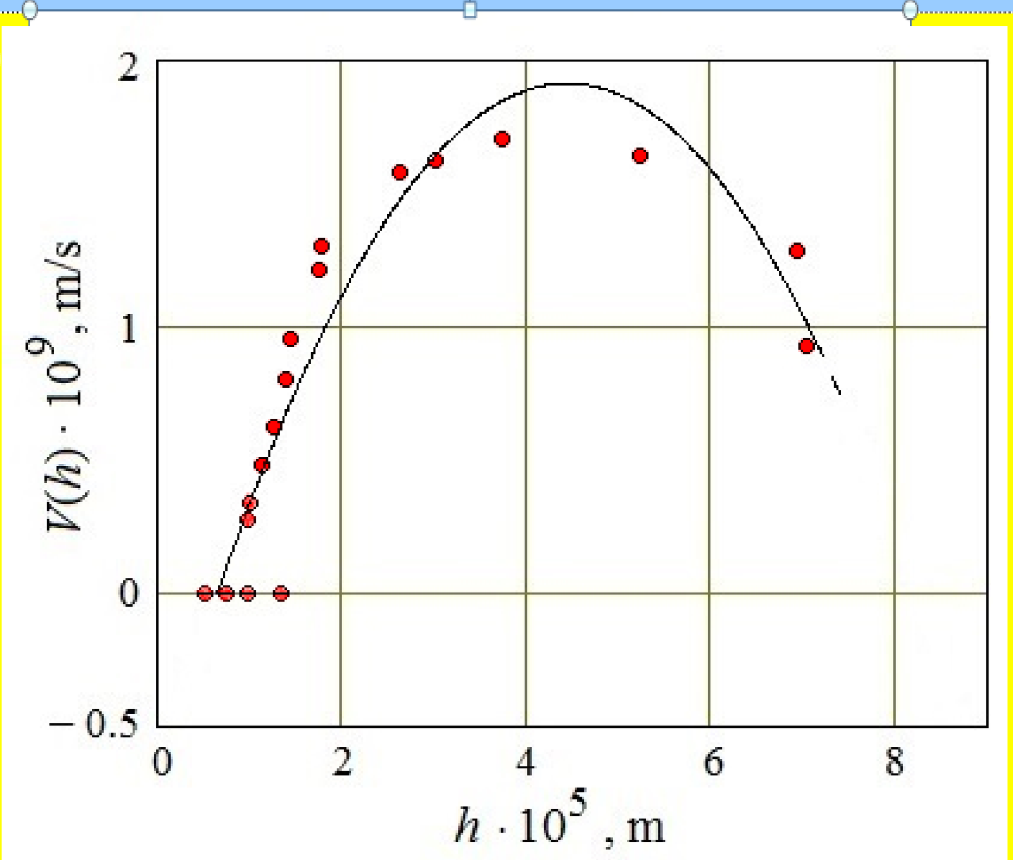
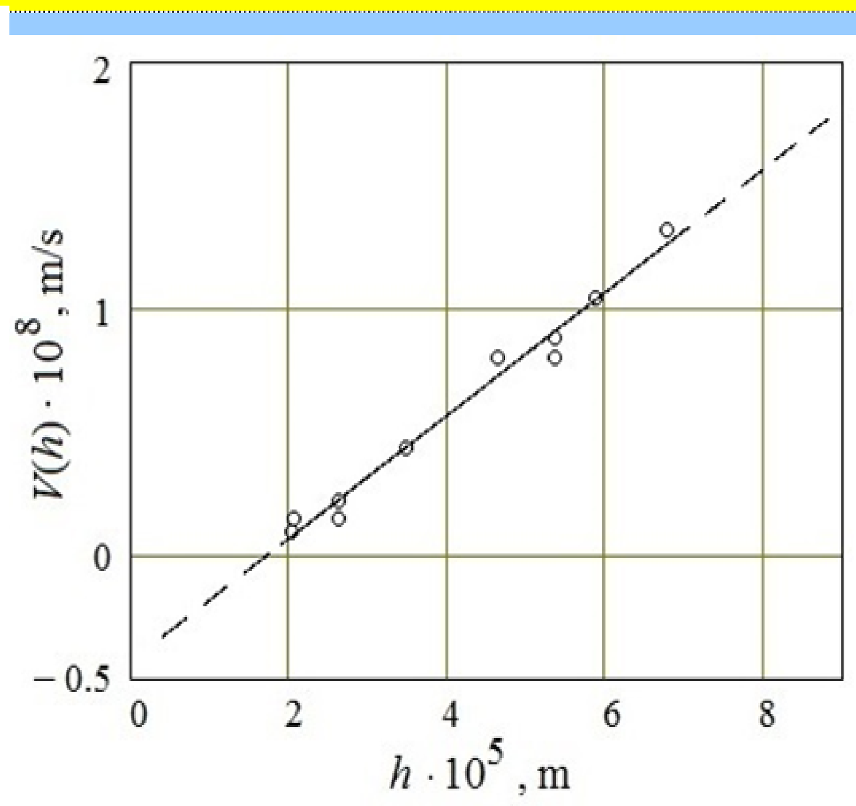


Fig. 3. Dependence of the maximum inclusion velocity on their size at a great temperature gradient .



Conclusion. The paper analyzes the appearance of convection mass transfer in a liquid inclusion, located in a thermally stressed crystal. We have analyzed papers that consider the motion of inclusions and the effect of convective mass transfer on their motion parameters. A number of papers have noted that the inclusion velocity is insignificantly sensitive to its orientation. In the opinion of the authors of these papers, that is a demonstration of the convective effect, because it is sensitive to the direction of the acceleration due to gravity. Contrary to previous researchers, this paper analyzes the forces acting in the inclusion. It is shown that due to the difference in osmotic pressure on the front and back inclusion surfaces, its own "effective" acceleration arises, which is directed along the temperature gradient. An estimated calculation of the Rayleigh number for a cell with solid boundaries is carried out, and its critical value is calculated. For such conditions, there are derived the expressions for the perturbed velocity and temperature of a cylindrical convective cell with solid boundaries that exists in the inclusion. The model of induced transitions of atoms of the crystalline matrix into the solution and back is proposed, taking into account convective mass transfer. Based on the proposed model, the analytical expressions for the dependence of inclusion velocity on their size at small and great temperature gradients are obtained. A good agreement between the theoretical model and the experimental data is shown for both cases. The proposed theoretical model for describing the dependence of the inclusion velocity on its size in the presence of convective mass transfer can be used to determine the characteristic parameters of both the kinetics of inclusion motion and the crystal itself.