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FLOW OF MATTER ONTO THE SURFACE OF A CRYSTAL UNDER CONDITIONS OF TURBULENT NATURAL CONVECTION

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In this work, the rate of growth of crystals from the gas phase under conditions of turbulent natural convection is determined.

The flow of matter onto the surface of a growing crystal is caused by a gradient, created by a corresponding temperature gradient in the medium, of the concentration of the active component in the gas phase. This could result in natural convection, whose effect on the rate of growth of the crystal also depends on the orientation of the system. The growth of crystals in vertical cylindrical ampuls is studied below. In this case, unstable stratification must exist in the gas phase in order for natural convection to arise, and this happens, for example, when the source ("hot" surface) is situated near the substrate ("cold" surface). It is assumed below that the natural convection is turbulent. We note that the conditions necessary for this are partially realized in practice. Like in [1], it is assumed that the gas phase is a binary mixture of active and inert components.

In the presence of turbulent pulsations the time-averaged rate of growth of the crystals, i.e., the velocity of the gas—solid interface, is given by the expression

$$\zeta = -\frac{\rho}{\rho^*}\left(v_n + \frac{1}{\rho}\langle\rho' v_n'\rangle\right), \quad \rho^* \gg \rho. \tag{1}$$

Here ρ and v_n are the average density and the component of the velocity of the gas phase normal to the front; ρ^* is the density of the crystal (ρ^* = const); and the prime indicates pulsation. Thus in order to find ζ it is necessary to know the velocity distribution in the gas phase and the correlation $\langle\rho' v_n'\rangle$. We shall confine our attention to the case when the gas density is a linear function of the temperature T, $\rho'/\rho = -\beta T'$ (β is the coefficient of volume expansion).

As the crystals grow the radial variation of the average temperature T is negligible compared with the variation along the axis. In addition, the Reynolds numbers, constructed based on the average velocity of the directed flow, are low (Re ∿ 1). Thus in this case the turbulence is determined by the effect of thermogravitational forces only. Under these conditions the balances of the second moments of the pulsations of the velocity and temperature [2, 3], written in the Boussinesq approximation, have the form

$$\frac{k}{l}E^{1/2}\langle v_x' v_r'\rangle + \beta g\langle v_r' T'\rangle = 0, \tag{2}$$

$$\frac{k}{l}E^{1/2}\langle v_x' v_\varphi'\rangle + \beta g\langle v_\varphi' T'\rangle = 0,\ \frac{k}{l}E^{1/2}\langle v_r' v_\varphi'\rangle = 0,$$

Moscow. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 4, pp. 76-78, 1986. Original article submitted May 30, 1985.

 0021-8944/86/2704-0546$12.50

$$\frac{k}{2l}E^{1/2}\left(\langle v_x'^2\rangle-\frac{2}{3}E\right)+\frac{b}{3}\frac{E^{3/2}}{l}+\beta g\langle v_x'T'\rangle=0,$$ (2)

$$\frac{k}{2l}E^{1/2}\left(\langle v_r'^2\rangle-\frac{2}{3}E\right)+\frac{b}{3}\frac{E^{3/2}}{l}=0,\ \frac{k}{2l}E^{1/2}\left(\langle v_\varphi'^2\rangle-\frac{2}{3}E\right)+\frac{b}{3}\frac{E^{3/2}}{l}=0,$$

$$\langle v_x'^2\rangle\frac{\partial T}{\partial x}+\beta g\langle T'^2\rangle+\frac{k_T}{l}E^{1/2}\langle v_x'T'\rangle=0,$$

$$\langle v_x'v_r'\rangle\frac{\partial T}{\partial x}+\frac{k_T}{l}E^{1/2}\langle v_r'T'\rangle=0,\ \langle v_x'v_\varphi'\rangle\frac{\partial T}{\partial x}+\frac{k_T}{l}E^{1/2}\langle v_\varphi'T'\rangle=0,$$

$$\langle v_x'T'\rangle\frac{\partial T}{\partial x}+\frac{b_T}{l}E^{1/2}\langle T'^2\rangle=0,\ E=\frac{1}{2}\left(\langle v_x'^2\rangle+\langle v_\varphi'^2\rangle+\langle v_r'^2\rangle\right)=0.$$

In the construction of the system (2) it is assumed [4-6] that

$$\nu\left\langle\frac{\partial v_i'}{\partial x_k}\frac{\partial v_j'}{\partial x_k}\right\rangle=\frac{b}{3}\frac{E^{3/2}}{l}\delta_{ij},\ a\left\langle\frac{\partial T'}{\partial x_k}\frac{\partial T'}{\partial x_k}\right\rangle=\frac{b_T}{l}E^{1/2}\langle T'^2\rangle,$$

$$-\left\langle\frac{p'}{\rho}\left(\frac{\partial v_i'}{\partial x_j}+\frac{\partial v_j'}{\partial x_i}\right)\right\rangle=\frac{k}{l}E^{1/2}\left(\langle v_i'v_j'\rangle-\frac{2}{3}E\delta_{ij}\right),$$

$$-\left\langle\frac{p'}{\rho}\frac{\partial T'}{\partial x_j}\right\rangle=\frac{k_T}{l}E^{1/2}\langle v_j'T'\rangle.$$

Here ν and a are the kinematic viscosity and thermal diffusivity of the medium; p is the pressure; g is the acceleration of gravity acting along the x axis; ℓ is the turbulence scale; and k, k_T, b, and b_T are constants.

Solving (2), we find

$$\langle v_\varphi'^2\rangle=\langle v_r'^2\rangle=\frac{2}{3}E\left(1-\frac{b}{k}\right),\ \langle v_x'^2\rangle=\frac{2}{3}E\left(1+\frac{2b}{k}\right),$$

$$E=\alpha_E l^2 g\beta(\partial T/\partial x),\ \alpha_E=\frac{1}{k_T b}\left[\frac{2}{3}\left(1+\frac{2b}{k}\right)+\frac{b}{b_T}\right],$$

$$\langle v_x'T'\rangle=-\alpha l^2\sqrt{g\beta(\partial T/\partial x)},\ \alpha=b\alpha_E^{3/2},$$

$$\langle T'^2\rangle=\frac{b}{b_T}\sqrt{\alpha_E}\,l^2(\partial T/\partial x)^2,$$

$$\langle v_x'v_r'\rangle=\langle v_x'v_\varphi'\rangle=\langle v_r'v_\varphi'\rangle=\langle v_r'T'\rangle=\langle v_\varphi'T'\rangle=0.$$

Reynolds' equations for the case when $\mathbf{v}=(v_x,0,0)$ are written in the form

$$-\frac{1}{\rho}\frac{\partial p}{\partial x}+\nu\left(\frac{\partial^2 v_x}{\partial r^2}+\frac{1}{r}\frac{\partial v_x}{\partial r}\right)+\beta\frac{\partial T}{\partial x}xg=0,$$ (3)

$$-\frac{1}{\rho}\frac{\partial p}{\partial r}+\frac{1}{r}\frac{\partial}{\partial r}\langle v_r'^2\rangle+\frac{1}{r}\langle v_\varphi'^2\rangle=0,\ v_x(r=r_0)=0.$$

From here, assuming that the turbulence scale $\ell=l(r)$ and setting $\partial T/\partial x=\text{const}$, we find that the profile of the averaged velocity is distributed in the section according to a parabolic law

$$v_x=2v_0(1-R^2),\ R=r/r_0.$$ (4)

Based on [1] the average velocity

$$v_0=-\frac{D}{x_0}\frac{2H}{2-H},\ H=\frac{c_+-c_-}{1-c_-}.$$

Here c_- and c_+ are the mass concentrations of the active component on the substrate (x = 0) and at the source ($x=x_0$); r_0 is the radius of the ampul; and D is the diffusion coefficient.

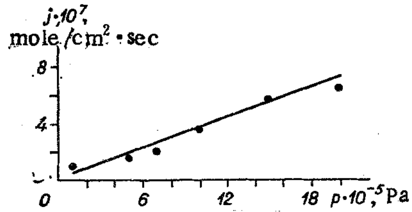

Fig. 1

Assuming that $b/k = 0.125m$ $b_T/k = 0.2$, $k_T/k = 0.9$, $k = 1.12$ and defining the turbulence scale according to Nekuradze $l/r_0 = 0.14 - 0.08R^2 - 0.06R^4$, after substituting (3) and (4) into (1) and averaging ζ over the section we obtain

$$\rho^*\zeta_0 = \rho\left[\frac{D}{x_0}\frac{2H}{2-H} + 0.248 r_0^2 \sqrt{g(\beta\partial T/\partial x)^3}\right].$$

Thus natural turbulent convection increases the rate of crystal growth.

Figure 1 shows the computational results for the growth of Ge crystals by means of chemical transport, based on the reaction

$$Ge_{(s)} + GeI_{4(g)} = 2GeI_{2(g)}. \tag{5}$$

The indices (s) and (g) denote the solid and gas phases. The turbulent flow (molar) of Ge given, in accordance to (5), by

$$j = \frac{j_0}{2M},\ j_0 = 2\int_0^1 \rho\beta\langle v_x' T'\rangle R\,dR,$$

where M is the molar mass of the active component GeI_2, is plotted along the ordinate axis, while the pressure is plotted along the abscissa axis. The figure also shows the experimental data of [7], consisting of the difference between the Ge flows onto the substrate under terrestrial conditions and under zero g. The pressure range $10^5 \le p \le 20\cdot10^5$ Pa corresponds to Grasshof numbers $Gr \cong 10^7 - 10^{10}$, where $Gr = g\beta x_0^4(\partial T/\partial x)/\nu^2$. The pressure dependence of the turbulent component of the flow of matter onto the substrate is determined by the pressure dependence of the density of the gas phase.

The data in the figure refer to an orientation of the system for which the substrate lies above the source. In addition to this, in [7] the opposite orientation, corresponding to a source lying above the substrate, was studied. In this case no differences were observed between the rates of growth of crystals measured under terrestrial conditions and under zero g. This is not surprising, since under the conditions of stable stratification, occurring in this case, there was no natural convection in the gas phase.

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