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## NONLINEAR OSCILLATIONS IN BULK CRYSTALLIZATION PROCESSES

V. V. Mansurov and I. A. Natalukha

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Nonlinear oscillations generated by losses of stability of steady-state bulk crystallization are investigated.

Heat- and mass-transfer processes, which are used extensively in chemical technology, metallurgy, and heat engineering and which result in the formation of particles of a new phase, are often accompanied by various kinds of oscillations and fluctuations of the characteristic parameters such as the temperature, concentration of the two-phase system, supersaturation, etc. [1-4]. These unstable regimes are manifested in diverse ways because of the vigorous ongoing development of highly promising technologies that make effective use of major departures of the system from equilibrium phase conditions and operate with metastable states. The investigation of mechanisms for the changeover of heat- and mass-transfer regimes is especially timely in the determination of ways to control transient processes, because the onset of instability can have a significant influence on the nature and the results of the processes involved.

In the present article we investigate the laws governing the loss of stability of steady-state regimes and the inception of highly nonlinear self-excited oscillatory regimes of bulk crystallization from supersaturated solutions or supercooled melts, where the twophase mixture is subjected to intense mixing, and the finished crystals are extracted from the system at a rate that depends on their dimensions.

# DERIVATION OF THE EVOLUTION EQUATION

For definiteness, we shall discuss crystallization from supersaturated solutions.

The physical mechanism of the formation of self-excited bulk crystallization regimes in a metastable medium under steady-state external conditions is described in [1]. The principal cause of instability of steady-state crystallization is a highly nonlinear

A. M. Gor'kii Ural State University, Sverdlovsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 54, No. 2, pp. 286-294, February, 1988. Original article submitted October 14, 1986.



Fig. 1. Neutral stability curves  $g_1 = S(R, b)$  in the plane of the parameters R,  $g_1$  for  $P_1 = 0$ ; values of  $g_1 > S(R, b)$ correspond to the stability domains of the steady-state regime.

Fig. 2. Comparison of theory (solid curve) and experiment [10] (points) in terms of the oscillation period T° along the neutral stability curve, b = 1.

dependence of the nucleation frequency on the degree of supersaturation, and this determines the fluctuation nucleation limit when a sufficiently high supersaturation level is attained.

Since a polydisperse ensemble of crystals is formed in crystallization and the volume of each crystal is much smaller than the volume of the carrier phase, the mass-transfer balance equation and the evolution equation for the distribution function f(t, r) of the crystals with respect to their radius r are written in the form

$$\frac{dc}{dt} = Q\left(\frac{c-c_0}{c_0}\right) - \rho \int_{r_0}^{\infty} \frac{dv}{dt} f(t, r) dr, \quad v = \frac{4}{3}\pi r^3; \tag{1}$$

$$\frac{\partial f}{\partial t} + \frac{d}{\partial r} \left( \frac{dr}{dt} f \right) + \gamma(r) f = 0, \quad \frac{dr}{dt} f \Big|_{r=r_*} = J \left( \frac{c-c_0}{c_0} \right). \tag{2}$$

The boundary condition in Eqs. (2) describes the nucleation process; the nucleation frequency in unit volume J is assumed to be a known function of the relative supersaturation and can be determined according to Frenkel'-Zel'dovich theory, Volmer's theory, or Mayer's phenomenology. The dependence of the effective extraction rate of crystals from the system  $\gamma$  on their size can be arbitrary in the context of the ensuing analysis.

We assume that the crystal growth rate is described by the relation

$$\frac{dr}{dt} = \frac{1}{\Psi(r)} \operatorname{X}\left(\frac{c-c_0}{c_0}\right).$$
(3)

Introducing the new variables

$$u = \frac{c - c_0}{c_0}, \quad \varphi = f \Psi^{-1}, \quad s = \int_0^r \frac{\Psi dr}{X(u_s)}$$
(4)

( $u_s$  is the steady-state supersaturation of the system; its existence will be established below), we reduce the highly nonlinear system (1)-(3) to a single evolution equation. In the variables (4), Eq. (2) and the boundary condition for it take the form

$$\frac{\partial \varphi}{\partial t} + \frac{X(u)}{X(u_s)} \frac{\partial \varphi}{\partial s} + \gamma [r(s)] \varphi = 0, \quad \varphi|_{s=0} \frac{J(u)}{X(u)}.$$
(5)

For simplicity, we let s = 0 in the boundary condition for (5) instead of

$$s = \int_{0}^{r_{*}} \frac{\Psi dr}{\mathbf{X}(u_{s})}.$$



Fig. 3. Characteristic curves of slightly nonlinear selfexcited oscillations: amplitude squared and frequency shift vs. parameter R and supercriticality ratio  $g_1/g_0^1 -$ 1 in the kinetic crystal-growth regime for power-law nucleation kinetics J = Cu<sup>n</sup>.

This is fully admissible insofar as the minimum crystal radius  $r_*$  only slightly exceeds the molecular diameter, and the main contribution to mass transfer is from cyrstals of radius much larger than  $r_*$ . Next, setting

$$\sigma = \int_{0}^{t} \frac{X(u[x]) dx}{X(u_s)}, \quad \Phi = \ln \varphi$$

and taking the Laplace transform of Eq. (5) with respect to the variable s, we arrive at the equation

$$\frac{\partial \Phi_p}{\partial t} + p \Phi_p - \ln \frac{J\left(u\left[t\left(\sigma\right)\right]\right)}{X\left(u\left[t\left(\sigma\right)\right]\right)} + \frac{X\left(u_s\right)\Gamma_p}{X\left(u\left[t\left(\sigma\right)\right]\right)} = 0,$$
(6)

where  $\Phi_p(\sigma, p)$  and  $\Gamma_p$  are the Laplace transforms of the functions  $\Phi(\sigma, s)$  and  $\gamma[r(s)]$ . Neglecting the term with the arbitrary constant in the general solution of Eq. (6), i.e., essentially analyzing the process in the fully developed asymptotic stage after the in-fluence of the initial conditions has completely disappeared, we have

$$\Phi_{p} = \int_{0}^{\sigma} \left[ \ln \frac{J(u[t(\sigma)])}{X(u[t(\sigma)])} - \frac{X(u_{s})\Gamma_{p}}{X(u[t(\sigma)])} \right] \exp\left[-p\left[(\sigma - \tau)\right]d\tau.$$
(7)

Once again taking the Laplace transform of Eq. (7) with respect to the variable  $\sigma$ , we obtain

$$\Phi_{pp'} = \left[ \ln \frac{J\left(u\left[t\left(\sigma\right)\right]\right)}{X\left(u\left[t\left(\sigma\right)\right]\right)} - \frac{X\left(u_{s}\right)\Gamma_{p}}{X\left(u\left[t\left(\sigma\right)\right]\right)} \right]_{\sigma \to p'} \frac{1}{p' + p}.$$

Taking the inverse Laplace transforms of p with respect to s and of  $p^\prime$  with respect to  $\sigma$  in succession, we have

$$\Phi = \ln \frac{J(u[t(\sigma - s)])}{X(u[t(\sigma - s)])} - \int_{0}^{s} \frac{X(u_s)\gamma[r(s - z)] dz}{X(u[t(\sigma - z)])}$$

or

$$f[r(s), t(\sigma)] = \frac{J(u[t(\sigma-s)])}{X(u[t(\sigma-s)])} \exp\left[-\int_{0}^{s} \frac{X(u_s)\gamma[r(s-z)]dz}{X(u[t(\sigma-z)])}\right] \Psi[r(s)].$$
(8)

Equation (8) specifies the distribution function of the crystals with respect to their radii in implicit form, since the function  $\sigma(t)$  cannot be inverted analytically. However, the time scale T of the crystallization process in real systems satisfies the condition  $\gamma_0 T >> 1$  [where  $\gamma_0$  is some characteristic value of the function  $\gamma(r)$ ], and so it is reasonable to assume that  $\sigma \approx t$ . The crystal-radius distribution function in this case acquires the form

$$f(t, r) = \frac{J\left(u\left[t-\int_{0}^{r} \frac{\Psi dr}{X(u_{s})}\right]\right)}{X\left(u\left[t-\int_{0}^{r} \frac{\Psi dr}{X(u_{s})}\right]\right)} \exp\left[-\int_{0}^{\kappa} \frac{X(u_{s})\gamma\left[r\left(\int_{0}^{r} \frac{\Psi dr}{X(u_{s})}-z\right)\right]dz}{X\left(u\left[t-\int_{0}^{r} \frac{\Psi dr}{X(u_{s})}\right]\right)}\right]\Psi(r), \qquad (9)$$
$$\varkappa = \int_{0}^{r} \frac{\Psi dr}{X(u_{s})}.$$

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Fig. 4. Characteristic curves of strong nonlinear self-vibrations: a) amplitude A and frequency  $\omega$  of vibrations with respect to supersaturation  $u/u_s$  vs supercriticality ratio, b = 1; R = 0.5;  $g_1^0$  = 2.95;  $w^0$  = 1.08; J = Cu<sup>n</sup>; b) shape of substantial non-linear self-vibrations.

If necessary, Eq. (9) can be replaced by relation (8) by a numerical procedure for inverting the function  $\sigma(t)$ .

We note that a finite-dimensional system of ordinary differential equations for the moments of the crystal-radius distribution function has been used in several well-known theoretical studies [3-7] for the analysis of bulk crystallization, but this system can only be obtained when the crystal growth rate does not depend on the size of the crystals and  $\gamma = \text{const}$ , which corresponds to different residence times of crystals of any radius in the apparatus. The second assumption is used in many papers and, on the whole, is fairly consistent with the real physical picture; however, the first assumption is not always valid by any means and requires the conformation of definite additional conditions in the crystallizer [8]. The advantages of the proposed approach over existing methods are obvious in light of these considerations. Using relations (8) or (9), we can easily calculate the moments of the crystal-radius distribution function (which are global characteristics of the process: the mean radius, surface area, and mass of the finished crystals) directly for specific functions  $\gamma(r)$ ,  $\Psi(r)$ , and X(u), making it possible to solve a broad category of problems associated with the implementation of crystallization both in steady-state and in transient (time-dependent) regimes.

Substituting Eq. (8) in (1) and introducing the dimensionless variables  $\Theta = \gamma_0 t$  and  $\gamma' = \gamma/\gamma_0$ , we obtain the evolution integrodifferential equation for the relative super-saturation

$$c_{0}\gamma_{0}\frac{du}{d\Theta} = Q\left(u\right) - 4\pi\rho\gamma_{0}^{-1}X\left(u\left[\Theta\right]\right)\int_{0}^{\infty} \frac{J\left(u\left[\Theta\left(\sigma'-\tau\right)\right]\right)}{X\left(u\left[\Theta\left(\sigma'-\tau\right)\right]\right)} \exp\left[-\int_{0}^{\tau} \frac{X\left(u_{s}\right)\gamma'\left[r\left(\tau-z\right)\right]dz}{X\left(u\left[\Theta\left(\sigma'-z\right)\right]\right)}\right]\frac{r^{2}\left(\tau\right)X\left(u_{s}\right)d\tau}{\Psi\left[r\left(\tau\right)\right]},$$

$$\sigma' = \int_{0}^{\Theta} \frac{X\left(u\left[x\right]\right)dx}{X\left(u_{s}\right)}, \quad \tau = \gamma_{0}^{-1}\int_{0}^{\tau} \frac{\Psi\left(r\right)dr}{X\left(u_{s}\right)}.$$
(10)

The need to take into account the specific dependences of the kinetics of crystal extraction from the volume on the size of the crystals has been indicated in many recent papers [7-9]; this can be a result of technological considerations (the necessity of sorting the finished crystals) and also be a departure from the normal operating regime of the reactor (clogging of the exit opening). We now give some typical dependences  $\gamma(r)$  as examples:

1) Randolph's M-z model [7]

$$\gamma = \begin{cases} M, & 0 < r < r_1, \\ l < M, & r_1 < r < r_2, \\ l < z < M, & r > r_2; \end{cases}$$
(11)

2) Born model [9]

 $\gamma = \frac{a_1}{1 + a_2 r^l}; a_1, a_2 > 0, l > 1;$   $\gamma = \frac{1 + c_1 r^l}{1 + c_2 r^l}; c_1, c_2 > 0, l > 1.$ (12)

#### STABILITY ANALYSIS OF STEADY-STATE CRYSTALLIZATION

Equation (10) admits a steady-state solution corresponding to crystallization with constant supersaturation and a constant nucleation frequency:

$$Q(u_s) = 4\pi\rho X(u_s) J(u_s) \gamma_0^{-1}\Omega,$$
$$\Omega = \int_0^\infty \exp\left[-\int_0^\tau \gamma' [r(\tau - z)] dz\right] \frac{r^2(\tau) d\tau}{\Psi[r(\tau)]}$$

Setting  $u = u_s(1 + \xi)$ , where  $|\xi| << 1$ , and linearizing Eq. (10), we obtain the perturbation evolution equation (we assume from now on that the inequality  $\gamma_0 T >> 1$  holds)

$$\frac{d\xi}{d\Theta} - P_{1}\xi + Rd_{1}\xi + (g_{1} - Rd_{1})\Omega^{-1}\int_{0}^{\infty}\xi(\Theta - \tau) \exp\left[-\int_{0}^{\tau}\gamma'\left[r\left(\tau - z\right)\right]dz\right]\frac{r^{2}\left(\tau\right)d\tau}{\Psi\left[r\left(\tau\right)\right]} + Rd_{1}\Omega^{-1} \times \\ \times \int_{0}^{\infty}\left[\int_{0}^{\tau}\gamma'\left[r\left(\tau - z\right)\right]\xi\left(\Theta - z\right)dz\right]\exp\left[-\int_{0}^{\tau}\gamma'\left[r\left(\tau - z\right)\right]dz\right]\frac{r^{2}\left(\tau\right)d\tau}{\Psi\left[r\left(\tau\right)\right]} = 0,$$
(13)

in which we have introduced the parameters

$$R = \frac{Q(u_s)}{c_0 \gamma_0 u_s}, \quad P_1 = \frac{Q'(u_s)}{c_0 \gamma_0}, \quad g_1 = R \frac{J'(u_s) u_s}{J(u_s)}, \quad d_1 = \frac{X'(u_s) u_s}{X(u_s)}$$

The spectral analysis of Eq. (13) yields the criterion of neutral stability

$$i\omega - P_{1} + Rd_{1} + (g_{1} - Rd_{1})\Omega^{-1}\int_{0}^{\infty} \exp\left[-i\omega\tau - \int_{0}^{\tau}\gamma'\left[r\left(\tau - z\right)\right]dz\right] \times$$

$$\times \frac{r^{2}(\tau)d\tau}{\Psi\left[r\left(\tau\right)\right]} + Rd_{1}\Omega^{-1}\int_{0}^{\infty}\left[\int_{0}^{\tau}\gamma'\left[r\left(\tau - z\right)\right]\exp\left(-i\omega z\right)dz\right] \exp\left[-\int_{0}^{\tau}\gamma'\left[r\left(\tau - z\right)\right]dz\right]\frac{r^{2}(\tau)d\tau}{\Psi\left[r\left(\tau\right)\right]} = 0.$$
(14)

The complex equation (14) specifies the surface of neutral stability in the space of parameters  $g_1$ ,  $d_1$ , R,  $P_1$ . Information on the functions  $\gamma'(r)$ , X(u), and  $\Psi(r)$  is needed in order to obtain specific results. As a simple example, we take

$$\gamma' = \text{const}, \quad \frac{dr}{dt} = \frac{\beta c_0 u}{r^{b-1}}, \tag{15}$$

where  $\beta$  and b are kinetic coefficients (b = 1 corresponds to the kinetic regime of crystal growth, and b = 2 to diffusion-limited growth). The curves traced by the neutral stability surface on the plane of the parameters g<sub>1</sub>, R is shown in Fig. 1. The comparison of the theoretical results with the experimental data [10] in terms of the oscillation period along the neutral stability curve is illustrated in Fig. 2. We note that the case g<sub>1</sub> < 0, which corresponds to the Tammann effect, has little bearing on applications and will therefore be disregarded here.

It is evident from Fig. 1 that steady-state crystallization is stable here, i.e., any fluctuation of the supersaturation decays exponentially with time, and instability sets in only when the ascending branch of the J(u) curve becomes sufficiently steep, despite the fact that for  $g_1 < S(R, b)$  a very strong metastability occurs in the system, since the Gibbs number  $G \sim 10$  in the indicated domain [the parameters R and  $g_1$  are proportional to  $J(u_S)$  and  $J'(u_S)u_S$ , respectively, with the same proportionality factor]. At the intersection of a neutral stability decreases); however, this is caused by an increase of  $J'(u_S)$  and is not associated with a variation of the nucleation frequency, so that the net effect is to promote an increase of instability. Consequently, the physical cause of instability is the highly nonlinear dependence of the nulceation rate on the supersaturation; in other words, instability can set in only when a large number of seed crystals are present simultaneously in the system, where they promote a diminution of supersaturation during the growth process.

#### NONLINEAR OSCILLATIONS

We now prove that weakly nonlinear self-excited supersaturation oscillations of small relative amplitude occur in the system for a slight ingress into the domain of instability.

Obviously, the supersaturation perturbations grow exponentially in the instability domain in the linear approximation. It is clear, however, that unbounded growth of the perturbations does not take place in reality. Exponential growth of the perturbations occurs only in the initial stage; very quickly, the perturbations are no longer small, and neither do they obey a linear equation. The ensuing analysis shows that the principal factor in the stabilization of perturbations and the onset of self-excited oscillations during crystallization is the highly nonlinear dependence of the nucleation frequency on the supersaturation level.

Retaining terms up to and including third-order in powers of  $\xi$  in Eq. (10) [11] and taking Eqs. (15) into account, we obtain the equation

$$\frac{d\xi}{d\Theta} - P_{1}\xi - P_{2}\xi^{2} - P_{3}\xi^{3} + (g_{1} - R)J_{10}^{0} + (g_{2} - g_{1} + R)J_{20}^{0} + + (g_{3} - g_{2} + g_{1} - R)J_{30}^{0} + RJ_{01}^{1} + (g_{1} - R)J_{11}^{1} + \frac{R}{2}J_{01}^{2} + + (g_{2} - g_{1} + R)J_{21}^{1} + \frac{1}{2}(g_{1} - R)J_{11}^{2} + \frac{R}{6}J_{01}^{3} - -RJ_{02}^{1} - (g_{1} - R)J_{12}^{1} - RJ_{4} + RJ_{03}^{1} + R\xi + (g_{1} - R)\xi J_{10}^{0} + + (g_{2} - g_{1} + R)\xi J_{20}^{0} + R\xi J_{01}^{1} + (g_{1} - R)\xi J_{11}^{1} + \frac{R}{2}\xi J_{01}^{2} - R\xi J_{02}^{1} = 0,$$
(16)

in which we have introduced the parameters

$$P_{k} = \frac{Q^{(h)} u_{s}^{h-1}}{k! c_{0} \gamma}; \quad g_{k} = \frac{RJ^{(h)} u_{s}^{h}}{k! J(u_{s})}; \quad Q^{(h)} = \frac{d^{k}Q}{du^{h}}; \quad J^{(h)} = \frac{d^{k}J}{du^{h}}; \quad (17)$$

$$I_{kn}^{m} = \Gamma^{-1}(3/b) \int_{0}^{\infty} \xi^{h}|_{\Theta-\tau} \left[\int_{0}^{\tau} \xi^{n}|_{\Theta-z} dz\right]^{m} \exp(-\tau) \tau^{-1+3/b} d\tau, \quad n, \quad m, \quad k = 1, 2, 3;$$

$$J_{4} = \Gamma^{-1}(3/b) \int_{0}^{\infty} \left[\int_{0}^{\tau} \xi|_{\Theta-z} dz\right] \left[\int_{0}^{\tau} \xi^{2}|_{\Theta-z} dz\right] \exp(-\tau) \tau^{-1+3/b} d\tau,$$

where  $\Gamma(\mathbf{x})$  is the gamma function.

We write the relative supersaturation in the Fourier series form:

$$u = u_{s} \left[ 1 + \sum_{n=-2}^{2} \Phi_{n} \exp(in\omega\Theta) \right], \ \Phi_{-n} = \Phi_{n}^{*},$$
  
$$\Phi_{0} \sim \Phi_{1} \Phi_{-1} = \Phi_{1}^{2} = q, \ q \sim g_{1}/g_{1}^{*} - 1,$$
(18)

where  $g_0^1$  is the value of  $g_1$  on the neutral stability surface. Substituting Eq. (18) in (16) and separating terms with different harmonics, we obtain for expressions for  $\Phi_0$  and  $\Phi_2$  for n = 0.2. The complex equation for n = 1, in which the values of  $\Phi_0$  and  $\Phi_2$  must be included, is used to determine the amplitude squared of the fundamental  $\Phi_2^1$  and the frequency shift  $\omega - \omega^\circ$  ( $\omega^\circ$  is the frequency of the oscillations on the neutral stability surface). Omitting these formidable calculations, we give the results of the calculations of q and  $\omega - \omega^\circ$  in Fig. 3 for Mayer nucleation kinetics  $J = Cu^n$  [8]. It is evident that q > 0 in the entire domain of parameters of the system, corresponding to "soft" destabilization and Hopf normal bifurcation of the steady-state regime [11]. As the supercriticality ratio is increased, the oscillation amplitude increases, and the frequency decreases. The time-average supersaturation perturbation  $\langle \xi \rangle = \Phi_0$  is negative in the instability domain and decreases with increasing supercriticality. The self-excited supersaturation oscillations lead to destabilization and oscillations of the global characteristics of bulk crystallization, i.e., the average radius, surface, and mass yield of the crystals. Calculations according to an equation given in the Appendix show that the average yield of the crystals increases a few percent when crystallization takes place in the self-excited regime, even for a small supercriticality, in comparison with the mass yield in the steady-state regime; this fact has been confirmed experimentally in several papers [12]. An analysis also shows that this effect is strongest for the kinetic regime of crystal growth.

After deep ingress into the instability domain, the theory developed above for weakly nonlinear oscillations is invalid. We have analyzed Eq. (10) numerically for this case. We used a modified Aitken-Steffensen iterative method [13], which has ultrafast secondorder convergence and differs from other iterative procedures in that it does not require computation of the derivative of the kernel in each iteration step. It follows from Fig. 4 that the generated large-amplitude oscillations are far from harmonic. The theory also indicates the possibility of self-excitation occurring for deep ingress into the instability domain. However, the oscillation frequency is close to zero in the indicated domains.

We note in conclusion that external periodic actions afford practical tools for the intensification and stabilization of unstable bulk crystallization regimes characterized by strong nonlinearities during crystal nucleation and growth. For example, periodic admission of the feed solution into the system tends to broaden the stability domains of the steadystate regime and, close to the instability threshold, can produce resonance effects associated with an increase in the mass yield and average size of the crystals [14].

# APPENDIX

Here we give an expression for the moments of the distribution function of the crystals with respect to their radii in the weakly nonlinear self-excited crystallization regime for the kinetics of crystal growth and extraction from the volume (15):

$$\begin{split} m_{h} &= \int_{0}^{\infty} fr^{h} dr = \left(\frac{b\beta c_{0} u_{s}}{\gamma}\right)^{h/b} \gamma^{-1} J\left(u_{s}\right) \Gamma\left(1 + kb^{-1}\right) \left\{1 + \Phi_{0}\left(g_{1}^{\circ} R^{-1} + kb^{-1}\right) + 2q\left(g_{2} R^{-1} - g_{1}^{\circ} R^{-1} - kb^{-1}\right) + 2q\omega^{-2} \left[1 - \operatorname{Re} A\left(\omega\right)\right] + 2q\left(g_{2} R^{-1} - g_{1}^{\circ} R^{-1} - kb^{-1}\right) + 2q\omega^{-2} \left[1 - \operatorname{Re} A\left(\omega\right)\right] + \omega^{-1} \operatorname{Im} \Phi_{2} + 2q \operatorname{Re} A\left(2\omega\right) \left[g_{2} R^{-1} - g_{1}^{\circ} R^{-1} + 1\right] - \omega^{-1} \operatorname{Re} \Phi_{2} \operatorname{Im} A\left(2\omega\right) - \left(2\omega\right) - \omega^{-1} \operatorname{Im} \Phi_{2} \operatorname{Re} A\left(2\omega\right) - 2q\left(g_{1}^{\circ} R^{-1} - 1\right)\omega^{-1} \left[\operatorname{Im} A\left(\omega\right) - \operatorname{Im} A\left(2\omega\right)\right] + q\omega^{-2} \left[2\operatorname{Re} A\left(\omega\right) - \operatorname{Re} A\left(2\omega\right) - 1\right] - 2q\omega^{-1} \operatorname{Im} A\left(2\omega\right) + 4q\omega^{-2} \left[2\operatorname{Re} A\left(\omega\right) - \operatorname{Re} A\left(2\omega\right) - 1\right] - 2q\omega^{-1} \operatorname{Im} A\left(2\omega\right)\right] + sin 2\omega\Theta\left(2\left(1 - g_{1}^{\circ} R^{-1}\right)\left[\operatorname{Im} \Phi_{2} \operatorname{Re} A\left(2\omega\right) + Re \Phi_{2} \operatorname{Im} A\left(2\omega\right)\right] + w^{-1} \operatorname{Re} \Phi_{2} - 2q\left(g_{2} R^{-1} - g_{1}^{\circ} R^{-1} + 1\right) \operatorname{Im} A\left(2\omega\right) + \omega^{-1} \operatorname{Im} \Phi_{2} \operatorname{Im} A\left(2\omega\right) - \omega^{-1} \operatorname{Re} \Phi_{2} \operatorname{Re} A\left(2\omega\right) + q\omega^{-2} \left[\operatorname{Im} A\left(2\omega\right) - 2\operatorname{Im} A\left(2\omega\right)\right] + 2\sqrt{q} \operatorname{Sin} \omega\Theta \times 2\sqrt{q} \operatorname{Sin} \omega\Theta + \omega^{-1} \operatorname{Im} A\left(\omega\right) \left[1 - g_{1}^{\circ} R^{-1}\right] - \omega^{-1} \operatorname{Re} A\left(\omega\right) + \omega^{-1}\right] + o\left(q\right)\right\}, A\left(x\right) = \left(1 + ix\right)^{-3/b}. \end{split}$$

### NOTATION

c, solute concentration; c<sub>o</sub>, thermodynamic-equilibrium concentration;  $\rho$ , crystal density; t, time, r, crystal radius; f(t, r), distribution function of crystals with respect to radii; J, J<sup>(k)</sup>, nucleation frequency and its k-th derivative; r<sub>\*</sub>, minimum crystal radius; c, n, Mayer nucleation kinetic constants; v, crystal volume;  $\gamma(r)$ , kinetic function for extraction of crystals from volume; Q, mass flow rate; u, dimensionless super-saturation level; u<sub>s</sub>, steady-state supersaturation level;  $\xi$ , dimensionless supersaturation perturbation;  $\gamma'(r)$ , dimensionless rate of crystal extraction from system;  $\Theta$ , dimensionless time; T, characteristic time scale of the process;  $\phi_n$ , amplitude of n-th harmonic of supersaturation perturbation;  $\omega$ , oscillation frequency; q, amplitude squared of fundamental

harmonic of supersaturation perturbation; A, amplitude of highly nonlinear self-excited oscillations; G, Gibbs number;  $m_k$ , k-th moment of crystal-size distribution function. The index ° signifies that the corresponding quantity is evaluated on the surface of neutral stability; angle brackets denote time average; the asterisk denotes complex conjugates.

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### VIBRATIONS AND NONUNIFORM HEATING OF A SHAFT IN A RADIAL BEARING

Yu. A. Buevich and M. I. L'vov

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The article examines the problem of vibrations of the axis of a shaft in a radial bearing due to the imbalance of the system as a result of nonuniform heating of the shaft.

Trouble-free operation, longer life, and reliability of turbine rotors and rotating units of other installations require careful balancing with the object of minimizing the level of perturbing forces and moments acting on the plant. In practice it is impossible to eliminate these fluctuations completely; as a result, the axis of rotation, and also the axes of bearing shafts do not take up a strictly fixed position, instead they vibrate [1]. If the random effects on the system are negligibly small, then to such vibrations there correspond periodic motions of the point of intersection of the shaft axis with the plane of the bearing along some closed path, usually close to elliptical [2]. When the regime of rotation changes, it is possible that the characteristic linear dimension of the path (the vibration amplitude) abruptly changes; this prevents the normal functioning of friction units and may even lead to their destruction, or even to the breakdown of the installation itself or some of its parts [3]. It is therefore of interest to find the causes of such vibrations and the dependences of their characteristics on the physical and regime parameters.

One of the causes of imbalance of some plant (which would be perfectly balanced under isothermal conditions) may be the bending of shafts in bearings due to their thermal

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