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FREEZING OF GRANULES IN AN APPARATUS FOR CRYOGRANULATION OF LIQUID MATERIALS

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The numerical solution of the problem of cooling and freezing a spherical drop under conditions of convective heat transfer is studied.

Cryogranulation of liquid materials is the main element of cryodispersion technology for obtaining organic and inorganic powdered materials. The quality of the final product is determined, as a rule, by the degree of monodispersity of the obtained granules and the conditions under which the granules are frozen.

In cryodispersion technology, different types of drop generators [1, 2] as well as systems for maintaining and monitoring the pressure and temperature of the dispersed liquid are employed to obtain a flow of monodispersed drops.

Freezing of the flow of drops is one of the most important stages of cryodispersion technology. In the process of solidification and cooling of the drops of liquid, a crystalline structure is formed, and this ultimately determines the quality of the product. In the process the sizes of the crystals formed are determined by the rate of freezing. In addition, the rate of freezing also affects other specific properties of the granulated material (the possibility of segregation of the components of the solution and others).

Under the conditions of low-temperature treatment the monodispersity can be destroyed owing to merging of drops in flight as well as at the moment that the cooling agent arrives at the surface. To prevent this undesirable phenomenon it is proposed that the freezing of the drops in nitrogen vapor be organized so that there would be enough time for the particles to freeze before the cooling agent reaches the surface.

A diagram of the apparatus for cryogranulation of objects with freezing in a convective flow is shown in Fig. 1. The apparatus operates as follows. The working solution is injected under a fixed pressure from the preparation system 1 into a drop generator 2 and is forced through a draw plate. Oscillations of the piezoelectric element are superposed on the jet emerging from the drop generator, and in a certain range of frequencies the jet is broken up into monodispersed drops. The drops are then subjected to low-temperature treatment in the freezing unit 3. The drops are cooled with nitrogen vapors, whose temperature is close to the saturation temperature at atmospheric pressure. The frozen drops enter the cooled assembly 5. Low temperature is maintained in the freezing unit by feeding liquid nitrogen from a Dewar bottle 6 into a nitrogen jacket 4. In order to reduce heat inflow from the surrounding medium the unit is insulated with a layer of thermal insulation.

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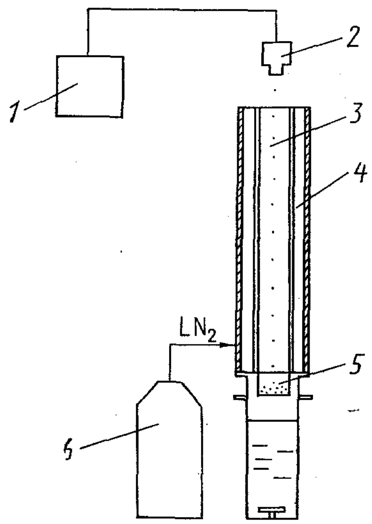

Fig. 1

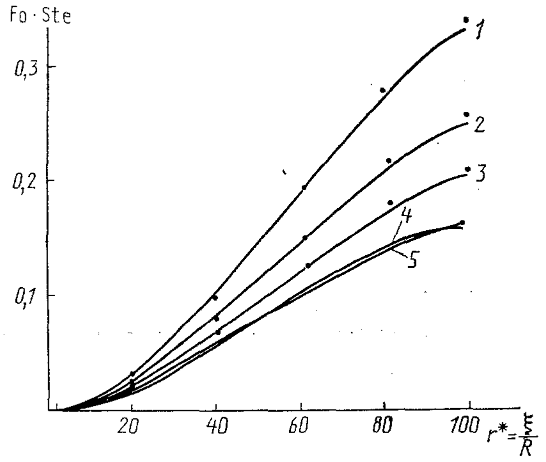

Fig. 2

Fig. 1. Line diagram of the apparatus for cryogranulation of liquid products.

Fig. 2. Comparison of the computational results (Bi → ∞): 1, 2, 3, 5) this work, Ste = 2.0, 1.0, 0.5, and 0.1; 4) calculation using Eq. (14); the dots are the data of [6].

The length of the freezing unit is calculated from the condition that the drops must be completely solidified before they enter the collector, i.e.,

$$l = v(\tau_0 + \tau_s).$$

In order to determine the length the problem of cooling and freezing of the drops must be solved. This problem is described by the equation

$$\frac{\partial T}{\partial \tau} = a\left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r}\right) \tag{1}$$

and the conditions

$$\tau = 0,\ T = T_i; \tag{2}$$

$$r = 0,\ \frac{\partial T}{\partial r} = 0; \tag{3}$$

$$r = R,$$

$$\lambda\frac{\partial T}{\partial r} = \alpha(T - T_c) \text{ — before the drops start to freeze;} \tag{4a}$$

$$\lambda'\frac{\partial T}{\partial r} = \alpha(T - T_c) \text{ — after the drops start to freeze;} \tag{4b}$$

$$r = \xi,\ \lambda'\frac{\partial T}{\partial r} - \lambda\frac{\partial T}{\partial r} = \rho q\frac{\partial \xi}{\partial \tau}. \tag{5}$$

The exact analytic solution of this problem is known only for the stage of cooling of a drop prior to the start of crystallization [4]:

$$\Theta = \sum_{n=1}^{\infty}\frac{2(\sin\mu_n - \mu_n\cos\mu_n)\sin\mu_n R}{(\mu_n - \sin\mu_n\cos\mu_n)\mu_n R}\exp(-\mu_n^2\mathrm{Fo}), \tag{6}$$

where $\Theta = (T - T_c)/(T_i - T_c)$ and μ_n are the roots of the equation $\tan\mu = -\mu/(\mathrm{Bi} - 1)$.

We shall study some limiting cases of the problem (1)-(5).

For Bi ≪ 1 the solution for the cooling stage prior to the start of crystallization can be obtained from Eq. (6):

$$\Theta = \frac{\sin\sqrt{3\mathrm{Bi}}R}{\sqrt{3\mathrm{Bi}}R}\exp(-3\mathrm{Bi\,Fo}) \tag{7}$$

or

$$\tau_0 = -\frac{Rc_p\rho}{3\alpha}\ln\Theta_{cr} \tag{8}$$

For the stage of crystallization with Bi ≪ 1 and Ste ≪ 1 the crystallization time of a drop can be found from heat balance

$$\int_0^R \rho' q 4\pi r^2 dr = \int_0^{\tau_{cr}} \alpha(T_i - T_{cr})\, 4\pi R^2 d\tau, \tag{9}$$

$$\tau_{cr} = \frac{1}{3}\frac{q\rho' R}{\alpha(T_i - T_{cr})}. \tag{10}$$

In addition, for this stage with Ste ≪ 1 a more accurate expression can be obtained by using the quasistationary approximation. In this case, for the solidified part of the drop we have the stationary equation of heat conduction

$$\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r} = 0 \tag{11}$$

with the boundary conditions (2)-(5).

This approach makes it possible to obtain the dependence of the temperature on the radius of a drop for any position of the front:

$$T = T_{cr} + \frac{\alpha(T_c - T_{cr})}{\left(\frac{\alpha}{R} - \frac{\lambda}{R^2} - \frac{\alpha}{\xi}\right)}\left(\frac{1}{r} - \frac{1}{\xi}\right) \tag{12}$$

and the time dependence of the position of the freezing front:

$$\tau = \frac{q\rho}{\lambda'\alpha(T_{cr} - T_c)}\left(\frac{\alpha\xi^2}{2} - \frac{\xi^3}{3}\left(\frac{\alpha}{R} - \frac{\lambda}{R^2}\right)\right). \tag{13}$$

For τ_{cr} we obtain from Eq. (13)

$$\tau_{cr} = \frac{q\rho}{\lambda'\alpha(T_{cr} - T_c)}\left(\frac{\alpha R^2}{6} + \frac{\lambda R}{3}\right). \tag{14}$$

The problem in the formulation (1)-(5) can be solved by using the approximate analytical methods of [5], which are quite complicated, or some numerical method [3, 6].

We propose a modification of the finite-difference method for solving the problem (1)-(5). This modification permits taking into account the temperature dependence of the thermophysical properties and the initial temperature distribution in the drop and it is at the same time very convenient and quite simple. The method consists of the following.

We divide the volume of the drop into n equal layers. For each layer we write the heat balance in the differential form

$$\begin{gathered} dQ_\alpha = dQ_{\lambda,1}, \\ dQ_{\lambda,1} = dQ_{\lambda,2} + dQ_{c_p,1}, \\ \cdots\cdots\cdots\cdots \\ dQ_{\lambda,i} = dQ_{\lambda,i+1} + dQ_{c_p,i}, \\ dQ_{\lambda,n-1} = dQ_{c_p,n-1}, \end{gathered} \tag{15}$$

where $dQ_\alpha \approx \alpha(T_1 - T_c)R_1^2 4\pi\Delta\tau$ is the heat removed from the surface of a drop owing to convection over the time $\Delta\tau = \tau_{k+1} - \tau_k$; $dQ_{\lambda,i} \approx \lambda_i(T_{i+1,k} - T_{i,k})4\pi R_i R_{i+1}(\Delta\tau/\Delta R)$ is the heat transferred through the first layer owing to heat conduction over the time $\Delta\tau$ taking into account the stationary distribution of the temperature in a spherical layer; $dQ_{c_p,t} \approx 4/3\pi(R_t^3 - R_{i+1}^3)\rho_i c_{pi}\Delta T$ is the heat released in the first layer accompanying cooling for the temperature difference

$$\Delta T = \frac{T_{i,k-1} + T_{i+1,k-1} - T_{i,k} - T_{i+1,k}}{2}.$$

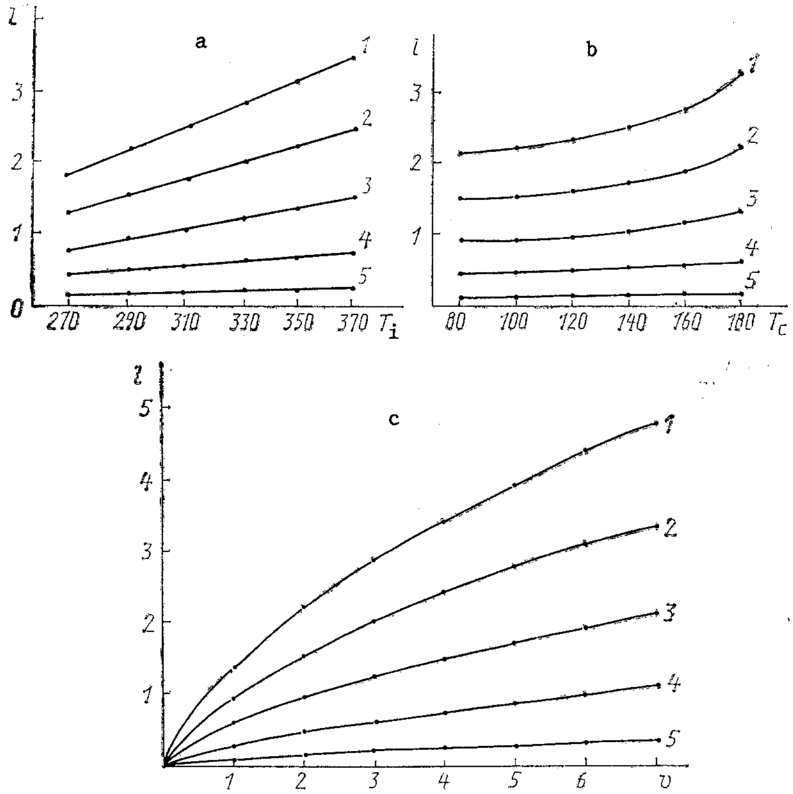

Fig. 3. Length of the freezing unit for drops with diameter D = 500 (1), 400 (2), 300 (3), 200 (4), and 100 (5) μm as a function of the initial temperature of the drops (a) (v = 2 m/sec; T_c = 100 K), the temperature of the nitrogen vapors (b) (v = 2 m/sec; T_i = 293 K), and the drop velocity (c) (T_i = 293 K; T_c = 100 K). ℓ, m; v, m/sec; and T_i and T_c, K.

At the center of a spherical drop the condition (3) is satisfied, and assumes the form

$$T_{n,k} = T_{n+1,k}. \tag{16}$$

Thus we obtain a conservative implicit difference scheme (15) and (16) for the process of cooling of the drop. The system contains n + 1 equations and n + 1 unknowns.

After the temperature of the surface of the drop reaches the temperature of the phase transition T, Eqs. (15) and (16) no longer change, except for the equations for the frozen layer, to which the term

$$dQ_{q,i} = \frac{4}{3}\pi (R_i^3 - R_{i+1}^3)\rho_i' q \tag{17}$$

is added. The solution of the system (15)-(17) was determined by the method of iterations. The heat-transfer coefficient is determined from the relation [7]

$$\mathrm{Nu} = 2 + 0{,}03\,\mathrm{Re}^{0,54}\mathrm{Pr}^{0,33} + 0{,}35\,\mathrm{Re}^{0,58}\mathrm{Pr}^{0,36}. \tag{18}$$

The thermal conductivity and viscosity of the nitrogen vapors was calculated using the formula of Sutherland [8]:

$$\lambda(T) = 2{,}397\cdot 10^{-2}\,\frac{377}{T+104}\left(\frac{T}{273}\right)^{3/2}, \tag{19}$$

$$\eta(T) = 167\cdot 10^{-7}\,\frac{377}{T+104}\left(\frac{T}{273}\right)^{3/2}. \tag{20}$$

The density of the vapor was determined from the relation

$$\rho(T) = 344/T. \tag{21}$$

The computational results obtained using the proposed method agree satisfactorily with the data of [6] as well as (for Ste $\ll$ 1) with the data obtained using the relation (14) (Fig. 2).

The program developed was used to perform calculations of the cooling and freezing of drops in the freezing unit and to determine the dependence of the length of the unit on the temperature of the nitrogen vapor T_c, the initial temperature of the drops T_i, the velocity of efflux of the drop flow v in the following range of values of the parameters: T_c = 80-180 K, T_i = 273-373 K, v = 0-7 m/sec, D = 100-500 μm (Fig. 3).

On the basis of the calculations presented in Fig. 3 it can be concluded that in order to reduce the length of the granule-freezing unit and therefore the flow rate of liquid nitrogen required to cool the system and to maintain the working temperature the following are required:

1) The rate of efflux of the drop flow must be reduced to the lowest values for which monodispersed drops can still be obtained.

2) The starting temperature of the dispersed liquid in the precooling stage must be reduced to a temperature close to the crystallization temperature. This is important for liquids having a high heat capacity, such as, for example, water, since when $T_i > T_{cr}$ the required increase in length of the freezing unit $\Delta \ell \sim (c_p \Delta T)/q$.

3) The temperature of the nitrogen vapor must be reduced as much as possible. This is especially important for drops with diameter D > 300 μm in the temperature interval studied (Fig. 3b).

A periodic-action apparatus for cryogranulation of liquid materials was developed on the basis of the calculations.

NOTATION

v, average velocity of the drops of liquid in the freezing unit; τ_0, cooling time before the drops start to crystallize; τ_s, solidification time of the drops; λ, thermal conductivity of the liquid phase; a, thermal diffusivity of the liquid phase; ρ, density of the liquid phase; λ', thermal conductivity of the solid phase; a', thermal diffusivity of the solid phase; ρ', density of the solid phase; q, heat of crystallization; α, average heat-transfer coefficient at the surface of the drop; R, radius of a drop; T_c, temperature of the nitrogen vapor; T_i, initial temperature of a drop; ξ, coordinate of the freezing front; Bi, Biot's number; Fo, Fourier's number; $T_{i,k}$, temperature at the outer boundary of the i-th layer at the k-th moment in time; i, layer number; i = 1, 2,...,n; and R, radius of the i-th layer.

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