

OPTIMIZATION OF THE PROCESS OF CONTINUOUS MASS CRYSTALLIZATION OF SALTS FROM SOLUTIONS

A. I. Moshinskii

UDC 548.01:66.065

Crystallization of a polydisperse system of crystals in the case of continuous organization of the process in a cascade of crystallizers is investigated. The process is assumed to occur in a kinetic regime. The influence of fluctuations in the rate of crystal growth is taken into consideration. Basic computational formulas are obtained and certain limiting situations are analyzed.

Introduction. Any technological process being carried out always raises a question concerning its best organization, with different meanings being attached to the notion of optimality. So, the process is often considered to be optimum if: 1) the productivity, i.e., the production of a given amount of a target product in a minimum time is at the maximum; 2) the minimum unit cost is ensured; 3) the products put out have the best criteria of any of the parameters, etc. It is not unusual that several requirements can be imposed simultaneously on the characteristics of the process. And it often turns out that they (or some of them) contradict (exclude) each other; therefore, the formulation of what is meant by optimum organization of a process is frequently the subject of research by itself [1].

As regards the process of mass crystallization, it can be noted that its mathematical simulation is being intensely developed at the present time along the lines of the solution and analysis of more complex equations as well as in the direction of studying the influence of new effects (coagulation, recycle, stability, self-oscillations, etc.) [2-8]. In these cases the problems of the optimization of crystallization are left almost untouched and constitute only a minute fraction in the total number of relevant publications. It may be mentioned that the basic problems of the optimization of the crystallization process with different requirements on optimality are described in the fourth chapter of the monograph [7]. Some of the problems, methods of analysis, and discussion of optimum organization of mass crystallization are presented in [9-12]. Below, as a criterion of optimality we take a problem of determining the process parameters that ensure the least spread (closeness to monodispersity) in the sizes of particles with respect to the prescribed mean size. The characteristic of the "spread" will be understood to represent the standard deviation (dispersion) [7], although other criteria are also suited for this purpose (see, for example, [9]). The process of crystallization will be considered in a cascade of apparatuses.

Statement of the Problem. In addition to the foregoing, we will assume that: 1) crystals grow in a kinetic regime, i.e., the rate of the growth of crystals is independent of their sizes; 2) the influence of fluctuations in the rate of crystal growth is taken into account; 3) the process is isothermal; 4) the process is stationary, i.e., all transient phenomena are over; 5) the stationary regime is considered to be stable, i.e., there are no self-oscillations; 6) the crystal is taken to be of spherical shape, i.e., it is characterized only by one parameter – its equivalent radius.

Under these conditions, a system of equations for the crystal size distribution function \hat{f}_j will be written in the form

$$U_j (2d\hat{f}_j/dr - Dd\hat{f}_j^2/dr^2) = [\hat{f}_{j-1}(r) - \hat{f}_j(r)]/\theta_j, \quad (1)$$

$j = 1, 2, \dots, N$ is the number of the apparatus.

We assume that the magnitude of the distribution function at the inlet to the first crystallizer $\hat{f}_0(r)$ is equal to zero and that crystals nucleate only in the first apparatus, as given by the boundary condition

$$U_j (2\hat{f}_j - Dd\hat{f}_j/dr) |_{r=0} = J\delta_{j1}, \quad j = 1, 2, \dots, N, \quad (2)$$

where δ_{ij} is the Kronecker symbol ($\delta_{ij} = 1$ at $i = j$ and $\delta_{ij} = 0$ with $i \neq j$); U_j is half of the crystal growth rate value (to simplify the representation of subsequent expressions, a value that is not the entire rate is taken). One other condition specifies the tending of the functions \hat{f}_j to zero when $r \rightarrow \infty$. From Eq. (2) we see that the coefficient of the crystal growth rate fluctuation is expressed by the approved [13, 14] dependence (the growth rate proportionality with the proportionality coefficient $D/2$).

It is worthwhile to analyze problem (1), (2) in dimensionless form; for this purpose, we will introduce new variables and parameters:

$$f_j = \hat{f}_j D / (J\theta_j), \quad \kappa_j = D / (U\theta_j), \quad y = r/D, \quad (3)$$

in which relations (1) and (2) will be rewritten as follows:

$$\begin{aligned} 2df_j/dy - df_j^2/dy^2 &= \kappa_j [f_{j-1}(y) - f_j(y)], \\ (2f_j - df_j/dy) |_{y=0} &= \kappa_1 \delta_{j1}, \quad j = 1, 2, \dots, N. \end{aligned} \quad (4)$$

Solution of the Equations for the Crystallization Problem. Analysis of a problem of type (4) in the absence of terms that correspond to the influence of fluctuations of the crystal growth rate is traditionally carried out [7, 15] by operator methods, the most convenient of which is the Laplace transform applied directly. In our case, there is also an integral transformation convenient for seeking a solution of the problem, namely, the so-called generalized Fourier transform, whose direct and inverse formulas have the form [16]:

$$f^*(\nu) = \int_0^\infty f(y) Z(y, \nu) dy, \quad f(y) = \frac{2}{\pi} \int_0^\infty f^*(\nu) \frac{Z(y, \nu) d\nu}{1 + \nu^2}, \quad (5)$$

where $Z(y, \nu) = \sin(y\nu) + \nu \cos(y\nu)$; the starred quantities are those that were transformed according to the first formula in (5). Before applying direct transformation (5), it is necessary to pass to new desired functions by the formulas $f_j(y) = \exp(y)g_j(y)$, $j = 1, 2, \dots, N$ in order to impart a "self-conjugate" form to system (4). As a result, instead of Eq. (4), we have

$$\begin{aligned} g_j - dg_j^2/dy^2 &= \kappa_j [g_{j-1}(y) - g_j(y)], \\ (g_j - dg_j/dy) |_{y=0} &= \kappa_1 \delta_{j1}, \quad j = 1, 2, \dots, N. \end{aligned} \quad (6)$$

The application of direct transformation (5) to problem (6) yields

$$g_j^*(\nu) (1 + \kappa_j + \nu^2) = \kappa_j g_{j-1}^*(\nu) + \nu \kappa_1 \delta_{j1}, \quad (7)$$

$$j = 1, 2, \dots, N.$$

From this we find

$$g_j^*(\nu) = \nu \prod_{k=1}^j \kappa_k / (1 + \kappa_k + \nu^2). \quad (8)$$

The analytical form of inversion (8) depends on whether or not any parameters κ_j coincide at different indices. First, we will write out the results in the case when all the parameters x_j are different. The variants in which any of the indices coincide can often be easily obtained by means of the corresponding passage to the limit. Dividing expression (8) into partial fractions, we have

$$g_j^*(v) = v \sum_{k=1}^j \kappa_k R_k^j / (1 + \kappa_k + v^2), \quad R_k^j = \prod_{\substack{s=1 \\ s \neq k}}^j \kappa_s / (\kappa_s - \kappa_k), \quad R_1^1 = 1. \quad (9)$$

Now, having noted that using the tables of [17] it is easy to find the integral

$$K_1(y, \beta) = \frac{2}{\pi} \int_0^{\infty} \frac{vZ(y, v) dv}{(1 + \kappa + v^2)(1 + v^2)} = \frac{\exp(-y\beta)}{1 + \beta}, \quad (10)$$

in which $\beta = (1 + \kappa)^{1/2}$, we apply inversion formula (5) and obtain the desired solution of the problem

$$f_j(y) = \exp(y) \sum_{k=1}^j \kappa_k R_k^j \frac{\exp(-y\beta_k)}{1 + \beta_k}, \quad j = 1, 2, \dots, N. \quad (11)$$

When $j = N$, using relation (11) we determine the moments M_N^i of function $f_N(y)$ of an arbitrary order i :

$$M_N^i = \int_0^{\infty} y^i f_N(y) dy = i! \sum_{k=1}^N R_k^N / (\beta_k - 1)^i, \quad (12)$$

which can be used in the optimization problem. It should be noted that the moments of the crystal size distribution function reflect important characteristics of an aggregate of crystals. Thus, the zero moment is associated with the number of crystals, the first moment is associated with the mean size of crystals, the second – with the surface area, and the third – with the total (or mean) volume of crystals.

Of the numerous cases of coincidence of κ_j for different indices j , we need only the variant in which all κ_j coincide, i.e., $\kappa_j = \kappa = \text{const}(j)$. It is evident from expression (8) that the inversion formula will have the factor $(1 + \kappa + v^2)$ to the power j . This will require generalization of integral (10). Note that, the desired integrals can be obtained, in particular, from Eq. (10) by differentiating both sides of formula (10) with respect to κ and allowing for the corresponding factor and sign (minus for an odd number of differentiations and plus for an even one). A detailed calculation (which is omitted) results in the following expression:

$$K_{N+1}(y, \beta) = \frac{2}{\pi} \int_0^{\infty} \frac{vZ(y, v) dv}{(1 + v^2)(\beta^2 + v^2)^{N+1}} = \frac{\pi \exp(-\beta y)}{2\beta^{2N+1}} \left\{ \sum_{k=0}^N \frac{(N+k)!}{(N-k)! k!} \times \right. \\ \left. \times \frac{(\beta y)^{N-k}}{2^k} - \frac{1}{(1+\beta)} \sum_{j=0}^N \frac{[(1+\beta)y]^j}{j!} \sum_{k=0}^{N-j} \frac{(N+k)!}{k! 2^k} \left(\frac{\beta}{1+\beta} \right)^{N-k} \right\}. \quad (13)$$

Thus, in this case the solution of the problem will be as follows

$$f_j(y) = \kappa^j \exp(y) K_j(y, \beta), \quad j = 1, 2, \dots, N. \quad (14)$$

Selection of Optimum Values of the Parameters in the Solution Obtained. Let us now pass to the optimization problem. According to the foregoing, as a criterion of optimality (estimation of the homogeneity of crystal composition), we selected a requirement for attaining the minimum value of crystal size dispersion square in the last apparatus relative to the mean size

$$\sigma^2 = \int_0^{\infty} (y - y_*)^2 f_N(y) dy,$$

where $y_* = M_N^1 M_N^0$ is the prescribed mean size of crystals. Using the notation introduced, the formula for the dispersion square can be transformed as

$$\sigma^2 = M_N^2 - (M_N^1)^2 / M_N^0 = M_N^2 - y_*^2 M_N^0.$$

Since we have a single restriction on the parameters: $M_N^1 = y_* M_N^0$ (i.e., a problem in conditional extremum), we introduce the auxiliary Lagrange function

$$\Phi = \sigma^2 - \lambda (M_N^1 - y_* M_N^0) = M_N^2 - y_*^2 M_N^0 - \lambda (M_N^1 - y_* M_N^0),$$

where λ is the Lagrangian multiplier. From this it is clear that in a general case we must determine the first three moments of the function $f_N(y)$.

The basic problem is the determination of the parameters κ_j proceeding from the requirements of the optimum conditions. Therefore, attention should be given to the fact that by virtue of their definition and the known facts [7], these parameters depend on the supersaturation of the solution, temperature, and the mean residence time of crystals in the apparatus. For example, for the growth rate U_j the following formula was obtained [7]: $U_j = A \cdot C^m \varepsilon_j^n \exp(-E/RT_j)$, where A , m , n are constants, E is the activation energy, C is the supersaturation, ε_j is the power required for agitation, which can also be used for the regulation of κ_j , and T_j is the temperature.

Since the parameters κ_j enter identically (symmetrically) into formulas (12) for M_N^i , we see that the "constant" solution $\kappa_j = \kappa = \text{const}(j)$ satisfies the system of equations used for determining the optimum parameters obtained by differentiation of the function Φ with respect to all parameters κ_j and equating the derivatives to zero:

$$\partial [M_N^2 - y_*^2 M_N^0 - \lambda (M_N^1 - y_* M_N^0)] / \partial \kappa_j = 0, \quad j = 1, 2, \dots, N.$$

The value of κ itself can be determined from the superimposed relation: $M_N^1 = y_* M_N^0$. Thus, in order to establish the functional dependence between the optimum value of κ and mean size of crystals y_* , it is necessary to calculate the zero and first moments of the function $f_M(y)$, determined by formula (14) at $j = N$.

It should be noted that usually in problems concerning the extremum of σ^2 -type functions, the solution obtained gives a minimum, although, strictly speaking, this fact needs verification. Generally such verification can turn out to be nontrivial; therefore, for the most part, research workers rely on "physical considerations" and particular cases in which elucidation of the type of extremum involves no difficulties. Here we will not investigate the character of the extremum of the function Φ .

Formulas for the moments of the distribution function $f_N(y)$ are most easily obtained after constructing a system of equations for moments, as is often done in practice, although they can also be found from relation (12) after passage to the limit. Upon integrating Eq. (4) over y in the limits $(0, \infty)$, we obtain

$$\kappa (M_{j-1}^0 - M_j^0) = \kappa \delta_{j1}, \quad j = 1, 2, \dots, N, \quad (15)$$

whence, allowing for the fact that $M_0^0 = 0$ and for the properties of the Kronecker symbols, we find: $M_j^0 = \text{const}(j) = 1, j \geq 1$. Thus, the zero moments are determined. Now, we multiply Eq. (4) by y and again integrate over y in the limits $(0, \infty)$. As a result we have

$$\kappa (M_{j-1}^1 - M_j^1) = -2M_j^0 - f_j^0, \quad f_j^0 = f_j(0), \quad j = 1, 2, \dots, N. \quad (16)$$

Summing up all Eqs. (16) and performing certain calculations, we obtain

$$\kappa M_N^1 = 2N + \sum_{j=1}^N f_j^0 = 2N + \kappa/2 - \kappa^{N+1} F_N(\kappa), \quad (17)$$

where

$$F_N(\kappa) = \frac{2}{\pi} \int_0^\infty \frac{v^2 dv}{(1 + \kappa + v^2)^N (1 + v^2)^2}.$$

For the summation of the quantities f_j^0 , we will avail ourselves of the well-known formula for the sum of a geometric progression [17]. In this case the value of f_j^0 was represented by the inverse integral expansion (5) at $y = 0$ for our variant $\kappa_j = \kappa$, and formula (8) was used. The integral $F_N(\kappa)$ can be calculated by differentiating the expression for $F_1(\kappa) = 1/(2\kappa) - [(1 + \kappa)^{1/2} - 1]/\kappa^2$ with respect to the parameter κ ($F_1(\kappa)$ is found directly):

$$F_N(\kappa) = \frac{(-1)^{N-1} d^{N-1}}{(N-1)! d\kappa^{N-1}} F_1(\kappa).$$

The calculation leads to the following formula for the function $F_N(\kappa)$:

$$F_N(\kappa) = \frac{1}{\kappa^{N+1}} \left[N + \frac{\kappa}{2} + (1 + \kappa)^{1/2} \sum_{k=0}^{N-1} \frac{(N-k)(2k-3)!!}{k! \cdot 2^k} \left(\frac{\kappa}{1 + \kappa} \right)^k \right], \quad (18)$$

where $(2k+1)!! = 1 \cdot 3 \cdot 5 \cdot \dots \cdot (2k+1)$ and it is assumed by definition that $(-3)!! = -1$ and $(-1)!! = 1$. Relation (18) allows one to write the expression for M_N^1 in final form as

$$\kappa M_N^1 = N - (1 + \kappa)^{1/2} \sum_{k=0}^{N-1} \frac{(N-k)(2k-3)!!}{k! \cdot 2^k} \left(\frac{\kappa}{1 + \kappa} \right)^k. \quad (19)$$

It might appear that the function $F_N(\kappa)$ has a singularity when $\kappa \rightarrow 0$, but the Taylor expansion of this function in the vicinity of zero

$$F_N(\kappa) = \frac{1}{2^{N+1} (N-1)!} \sum_{k=0}^{\infty} \frac{(2N+2k-1)!!}{(N+k)(N+k+1)k!} \left(\frac{-\kappa}{2} \right)^k \quad (20)$$

indicates the boundedness of the function $F_N(\kappa)$ at the given point. Series (20) has a convergence radius equal to unity. The case $\kappa \rightarrow 0$ corresponds to a small influence of the crystal growth rate fluctuations [see Eq. (3)]. Confining ourselves to one term of the series, we obtain from Eq. (17) the following approximation of the function M_N^1 in the vicinity of zero:

$$\kappa M_N^1 \simeq 2N + \kappa/2 - [\kappa^{N+1} (2N-1)!!] / [2^{N+1} (N+1)!], \quad \kappa \rightarrow 0. \quad (21)$$

From this we see that formula (20) turns out to be especially useful at large values of N and rather small values of κ , because the effect of κ exerted on the result and associated with the function $F_N(\kappa)$, begins from the term of the order of κ^{N+1} , which has certain advantages over dependence (19) in spite of the fact that Eq. (19) is a finite expression, while Eq. (20) is a sum of an infinite series.

Of particular interest is also the opposite limiting case $\kappa \rightarrow \infty$. If we make use of a finite sum

$$\sum_{k=1}^{N-1} \frac{(N-k)(2k-3)!!}{k! \cdot 2^k} = N - \frac{(2N-1)!!}{2^{N-1} (N-1)!}, \quad (22)$$

we obtain from formula (19) the following asymptotic dependence for M_N^1

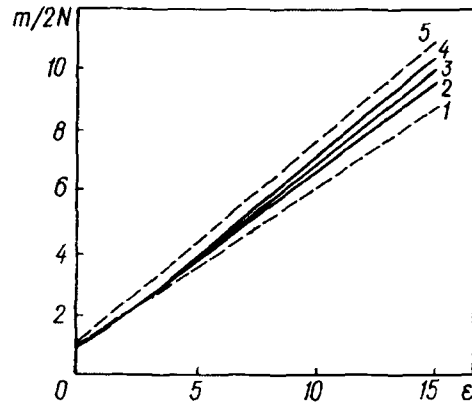


Fig. 1. Lines of optimum connection between the dimensionless parameters at different values of N : 1) $N = 1$ (it is also the asymptotic line for $\varepsilon \rightarrow 0$ and any value of N); 2) 3, 3) 4, 4) 5, limiting asymptotic line for $\varepsilon \rightarrow \infty$ and $N \rightarrow \infty$.

$$\kappa M_N^1 = N + \frac{(2N-1)!!}{2^{N-1}(N-1)!} (1+\kappa)^{1/2}, \quad \kappa \rightarrow \infty. \quad (23)$$

To calculate the sum (22), we used the well-known [17] formula

$$\frac{(2k-3)!!}{(k-1)! \cdot 2^k} = \frac{1}{\pi} \int_0^\infty \frac{dv}{(1+v^2)^k}, \quad k = 1, 2, 3, \dots,$$

and performed summation under the integral sign, etc. The relation between the parameter κ and the mean size of particles is given by the equation $M_N^1 = y_* M_N^0$, from which the basic formula follows:

$$\kappa y_* = 2N + \kappa/2 - \kappa^{N+1} F_N(\kappa). \quad (24)$$

First of all we note that according to Eq. (3) in the optimum regime the condition $\kappa_j = \kappa = \text{const}(j)$ leads to equality of the products of the residence time and the crystal growth rate in each apparatus of the cascade:

$$U_j \theta_j = \rho = \text{const}(j). \quad (25)$$

Here ρ has the dimensionality of length, whereas physically the parameter $\kappa = D/\rho$ is the ratio of two lengths, i.e., it characterizes the relative role of crystal-growth-rate fluctuations and size ρ , which corresponds to the scale of the crystals grown in the apparatus. Relation (25) shows that, as required, it is possible to change U_j or θ_j in a specific apparatus, but so that the product of these parameters remains constant and equal to ρ . Our statement of the problem assumes that the crystal mean size r_* is known or, according to Eq. (3), in dimensionless form $y_* = r_*/D = M_N^1/M_N^0$, while the parameter κ (which is equivalent to ρ) is the desired characteristic. However, we note that in formula (24) ρ , rather than D , plays the role of a natural scale for the crystal mean size, since $\kappa y_* = r_*/\rho$. This is especially significant when $D/\rho \rightarrow 0$ (which is often the case), i.e., when the parameter κ is a small quantity. Of course, on passing to the fluctuationless case of $\kappa \rightarrow 0$, the size should be scaled by the parameter ρ . In this connection, it may be useful to consider formula (24) as the relationship between parameters $m = r_*/\rho$ and $\varepsilon = D/r_*$ for determining the parameter ρ . Just as in the first case of the functional relationship between y_* and κ , in the variant noted we have in essence inverse functions. This is relation (24), which gives an explicit dependence of y_* on κ , as well as the following equation

$$m = 2N + \varepsilon m/2 - (\varepsilon m)^{N+1} F_N(\varepsilon m), \quad (26)$$

which follows from Eq. (24). Here, in performing tabulation, we can first assign the product εm , then determine the value of m from Eq. (26) and then, having divided the assigned value of εm by m , we obtain the value of ε , i.e., we virtually couple the parameters m and ε . When $N = 1$ and $N = 2$, we easily obtain analytical dependences between the parameters:

$$m = 2 + \varepsilon \quad \text{or} \quad \rho = r_*/(2 + D/r_*) \quad (N = 1), \quad (27)$$

$$m = [9\varepsilon^2 + 16\varepsilon - 4 + (81\varepsilon^4 + 288\varepsilon^3 + 312\varepsilon^2 + 128\varepsilon + 16)^{1/2}]/(8\varepsilon) \quad (N = 2).$$

The corresponding graphs for the values $N = 3, 4, 5$ are given in Fig. 1. Useful information, including that obtained on the graphical representation of the dependences $m = m\varepsilon$, is given by the following asymptotic formulas:

$$m/(2N) = 1 + \varepsilon/2, \quad (\varepsilon \rightarrow 0); \quad (28)$$

$$\frac{m}{2N} = 1 + \frac{\varepsilon}{2N} \left(\frac{(2N-1)!!}{2^{N-1} (N-1)!} \right)^2, \quad (\varepsilon \rightarrow \infty).$$

The factor in front of ε , which depends on N in the second formula of Eq. (28), changes from $1/2$ at $N = 1$ [see the first formula in Eq. (27)] to the limiting value $2/\pi \cong 0.636620$, for $N \rightarrow \infty$, obtained from the Stirling formula. When $\varepsilon \rightarrow 0$, the tangent of the slope of the straight line (the first equation of (28)) is also equal to $1/2$. From this it is seen (confirmed by the graphs and formulas (27)) that the curves $m = m(\varepsilon)$ in the coordinates $m/(2N)$ and ε tend to occupy a rather narrow sector with rays, whose tangents of the slope are equal to $1/2$ and $2/\pi$.

NOTATION

C , supersaturation of solution; D , coefficient of crystal growth rate fluctuations; \hat{f}_j, f_j , functions of crystal size distribution in j -th apparatus, dimensional and dimensionless, respectively; $F_N(\kappa)$, function determined by formula (17); $g_j(y) = \exp(-y)$, $f_j(y)$, new desired functions; J , nucleation rate in 1st crystallizer; M_N^i , moments of function $f_N(y)$ of order i ; N , number of crystallizers; Q , volumetric flow rate of liquid through system; r , crystal radius; V_j , volume of j -th apparatus; $y = r/D$, dimensionless crystal radius; $y_* = M_N^1/M_N^0$, mean size of crystals; $\beta = (1 + \kappa)^{1/2}$; σ^2 , square of the dispersion of crystal sizes in last apparatus of cascade; Φ , Lagrange function; $\kappa_j = D/(U_j \cdot \theta_j)$; ν , parameter of integral transformation (5); ρ , scale of grown crystals; $\theta_j = V_j/Q$, mean resistance time of particles in j -th apparatus.

REFERENCES

1. N. N. Moiseev, *Mathematical Problems of a System Analysis* [in Russian], Moscow (1981).
2. Yu. A. Buevich, V. V. Mansurov, and I. A. Natalukha, *Inzh.-Fiz. Zh.*, **49**, No. 2, 233-242 (1985).
3. Yu. A. Buevich, V. V. Mansurov, and I. A. Natalukha, *Inzh.-Fiz. Zh.*, **55**, No. 2, 275-284 (1988).
4. S. P. Fedorov, *Zh. Fiz. Khim.*, **62**, No. 12, 3251-3256 (1988).
5. A. I. Moshinskii, *Inzh.-Fiz. Zh.*, **64**, No. 2, 199-203 (1993).
6. A. I. Moshinskii, *Prikl. Mekh. Tekh. Fiz.*, No. 6, 75-83 (1993).
7. V. V. Kafarov, I. N. Dorokhov, and E. M. Kol'tsova, *Systems Analysis of Processes of Chemical Technology. Processes of Mass Crystallization from Solutions and the Gas Phase* [in Russian], Moscow (1983).
8. O. M. Todes, V. A. Seballo, and A. D. Gol'tsiker, *Mass Crystallization from Solutions* [in Russian], Leningrad (1984).
9. A. M. Tapilin, Yu. K. Tovbin, A. I. Gorbunov, and M. G. Slin'ko, *Dokl. Akad. Nauk SSSR*, **259**, No. 2, 423-426 (1981).
10. E. T. Klimenko, L. I. Khurgin, and A. Shnaider, *Teor. Osn. Khim. Tekhnol.*, **13**, No. 4, 538-545 (1979).

11. A. I. Moshinskii and M. I. Sibirev, *Zh. Prikl. Khim*, **62**, No. 4, 874-877 (1989).
12. A. I. Moshinskii, *Zh. Prikl. Khim.*, **67**, No. 9, 1454-1460 (1994).
13. A. D. Randolph and E. T. White, *Chem. Eng. Sci.*, **32**, 1067-1076 (1977).
14. I. V. Melikhov and B. D. Nebylitsyn, in: *Crystal Growth*, Vol. 12 [in Russian], Erevan (1977), pp. 108-115.
15. A. D. Randolph and M. A. Larson, *Am. Inst. Chem. Eng. J.*, **8**, No. 5, 639-645 (1962).
16. I. Sneddon, *Fourier Transforms* [Russian translation], Moscow (1955).
17. G. V. Dvayt, *Tables of Integrals and Other Mathematical Formulas* [Russian translation], Moscow (1973).