DISSOLUTION OF A POLYDISPERSE SYSTEM OF CRYSTALS WITH ALLOWANCE
FOR DISSOLUTION RATE FLUCTUATIONS

A. I. Moshinskii   UDC 66.061:539.21

The paper analyzes the formulation of the dissolution problem and its solution when
the growth rate of the particles fluctuates.

Models based on the introduction of the size distribution function of the particles are
used extensively in the mathematical description of a number of processes that involve the
growth of bubbles, drops, crystals, etc. [1-4]. There are two reasons for this: first, the
results obtained in such models are entirely satisfactory for practical purposes in regard
to accuracy and give all the necessary information and, second, the more detailed and rigor-
ous methods of the mechanics of multiphase systems [5, 6] lead to substantially more compli-
cated calculations and often, because of the inaccuracies and the difficulties of completing
the fundamental equations, cannot provide advantages over the approach based on the distri-
bution function.

For the case when the system is in a confined volume and no particles enter or leave the
volume, the dissolution problem of the basis of the distribution function has been analyzed
in [7-9] as it pertains to two dissolution laws: a diffusion law, when the dissolution rate
is inversely proportional to the crystal size, and a kinetic law, when the rate does not
depend on the crystal size. In both examples the dissolution rate is proportional to the
difference between the equilibrium concentration and the current concentration.

As is known [10-14], rate of change in the crystal size can fluctuate greatly, this
being attributed to kinetic phenomena on the crystal surface during the attachment/separation
of new particles and also to the variability of the hydrodynamic conditions in the neighbor-
hood of the crystal. Isolating the systematic (average) part of the rate of change r and
the fluctuational part, which is assumed to be Gaussian white noise, we arrive at the
Fokker–Planck equation [12-14] for the size distribution function of the crystals,

$$\frac{\partial f}{\partial t} = \frac{\partial}{\partial r}\left(vf + D\frac{\partial f}{\partial r}\right). \tag{1}$$

Phenomena similar to the dissolution of crystals can also occur in disperse systems of
another type, i.e., some of the results obtained below have more general significance but in
order to make the exposition more specific, we shall deal only with dissolution and the re-
verse process, crystallization, for which we obtained Eq. (1) with a fluctuation coefficient
and acquired some experience in determining this coefficient.

We should immediately take note of some disadvantages that ensue from the proposed
idealization in the description of the growth/dissolution of crystals. When the fluctua-
tional component of the growth rate is approximated by white noise, which is due to the small
time scale of the fluctuations in comparison with the characteristic time of variation of the
distribution function, a parabolic equation is obtained; its characteristic property, which
is realized in crystallization and dissolution problems, will consist in the "smearing" of
the initial profile of the distribution function that really exists in a bounded interval of
sizes along the entire r axis.* This corresponds to the fact that some crystals will grow

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*It is known (see, e.g., [15]) that for nonlinear equations of the parabolic type [Eq. (1)
is nonlinear, see below] there may exist a situation when a perturbation propagates with
finite velocity in the presence of a sharp front, ahead of which the solution does not change.
Our assertion is based on an analysis of the known exact solutions [12, 13, 16, 17] of the
crystallization equation and, apparently, is of a general nature for the relations used at
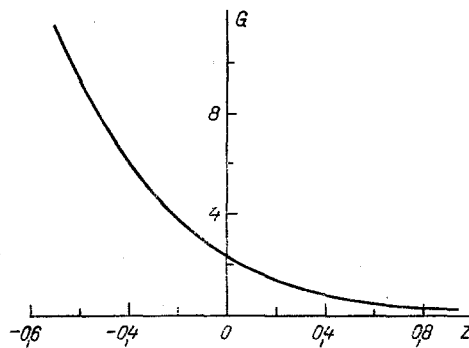present for the fluctuation coefficient of the growth rate of crystals.

Fig. 1. Plot of the function G(z).

during dissolution and, conversely, some crystals will dissolve during crystallization. These effects will be insignificant if the fluctuation coefficient (when made appropriately dimensionless) is a small quantity. This is usually done in practice (see, e.g., [18]). Evidently, a more adequate description of the process will be given by equations of the hyperbolic type or equations with a different structure, whose solutions do not give rise to the disadvantages indicated above. In any case, there are processes such that description of crystallization by means of Eq. (1) becomes undesirable [19].

One of our aims is to map out ways of looking for analytical solutions of the dissolution problem on the basis of Eq. (1), supplemented with relations known from the literature for the dissolution rate and its fluctuations.

Kinetic Regime. In this regime the dissolution rate depends only on the difference between the equilibrium and current concentrations of the dissolved substance and by means of straightforward manipulations this relation can be recast in the form

$$v = k(M - Q) = k\left(\int\limits_0^\infty r^3 f \, dr - Q\right).$$

(2)

The quantity Q is characterized to within $4\pi/3$ by the total volume of the crystals which can dissolve (with allowance for the initial volume) at the given temperature (which we assume is constant). As for the fluctuation coefficient D, its functional relation to the other parameters of the process has not been elucidated completely and several forms of this relation are used in calculations. Following [12, 18], we take the coefficient D to be proportional to v:

$$D = Bv, \quad B = \text{const} > 0,$$

(3)

which was verified in the experiments reported in the aforementioned papers and has shown itself to be fairly good in crystallization problems.

To complete the formulation of the problem it is necessary to give the auxiliary conditions. The initial condition

$$f|_{t=0} = g(r)$$

(4)

expresses the granulometric composition at t = 0. It is also easy to determine the condition as r → ∞: this is the boundedness of the function f as r → ∞. Here in fact we can require that f → 0, since in practical situations the function f, starting from some size r_m, becomes identically equal to zero. By virtue of the existence of a differential operator of the second order in the variable r it is necessary to formulate one more boundary conditions for r. This poses the fundamental complexity in the formulation of the problem. In the case of crystallization we set the condition for r = r_* — the nucelation radius, which is usually assumed to be zero because it is so small in comparison with the characteristic (average) size of crystals. This condition expressed the rate of formation of the new phase. Nor were any difficulties encountered in formulating the dissolution problem for D = 0, when the field of the characteristics of Eq. (1) indicated that no condition at all should

1410

be set for r and the solution of the problem is determined uniquely by the initial condition (4). The natural physical limits on the variable r for Eq. (1) are given by the interval r $\in$ (0, ∞). Mathematically, however, the point r = 0 is in no way isolated under the conditions (2) and (3) and it is scarcely feasible to set a condition at r = 0 which is associated with the rate at which particles disappear since this quantity is one that is sought rather than one that determines the problem, as is clear in the limiting case D = 0. With these circumstances we suggest that the most natural requirement is that the problems be consistent at D $\neq$ 0 and D = 0. First let us make a necessary condition (for further discussion)

$$d\lambda/dt = v, \quad \lambda|_{t=0} = 0, \tag{5}$$

temporarily assuming v to be a known function of t. In this case Eq. (1) becomes

$$\partial f/\partial \lambda = B\partial^2 f/\partial r^2 + \partial f/\partial r, \tag{6}$$

and the parameter λ will have the same sign as the time by virtue of the notation we have adopted for the dissolution equations (1) and (2).

For B = 0 the characteristics of Eq. (6) have the form r + λ = const and the value of the initial function (4) will be transposed along them without distortion. The extreme characteristic, which delineates the initial region r $\geqslant$ 0, is determined by the relation r = $-\lambda$. For B $\neq$ 0 the initial distribution will be "smeared" and, in our opinion, for the consistency of the solutions as B $\to$ 0 it is necessary to set the condition requiring that there be no inflow of crystals from the region r < $-\lambda$, i.e., that this region not affect the region r > $-\lambda$, as is the case when D = 0. This condition is equivalent to conservation

of the number of crystals in the region r $\in$ ($-\lambda$, ∞): $\int_{-\lambda}^{\infty} f dr$ = const. Then, integrating Eq. (6)

over r within the given limits, we easily arrive at the sought condition

$$B\partial f/\partial r|_{r=-\lambda} = 0, \tag{7}$$

which is also satisfied for B = 0. With this the formulation of the problem is completed. We note that relations (2) and (3) make the problem of determining the function f nonlinear.

By the simple change of variables r + λ $\to$ r, λ $\to$ λ, Eq. (6) is transformed into the heat equation and condition (7) is set at a "fixed" point. The solution of this problem is easily found by standard methods,

$$f(r, \lambda) = \frac{1}{2\sqrt{\pi B\lambda}} \int_0^\infty g(\xi) \left\{ \exp\left[-\frac{(r+\lambda-\xi)^2}{4B\lambda} \right] + \exp\left[-\frac{(r+\lambda+\xi)^2}{4B\lambda} \right] \right\} d\xi. \tag{8}$$

From this we determine the third moment M of the distribution function,

$$M = \int_0^\infty r^3 f dr = (B\lambda)^{3/2} \int_0^\infty g(\xi) \left[G\left(\frac{\lambda-\xi}{2\sqrt{B\lambda}} \right) + G\left(\frac{\lambda+\xi}{2\sqrt{B\lambda}} \right) \right] d\xi, \tag{9}$$

where

$$G(z) = 2[2(1+z^2)\exp(-z^2)/\sqrt{\pi} - z(3+2z^2)\text{erfc}(z)];$$

$$\text{erfc}(z) = \frac{2}{\sqrt{\pi}} \int_z^\infty \exp(-t^2) dt. \tag{10}$$

The plot of the function G(z) is shown in Fig. 1. For large values of the argument the function G(z) has the asymptotic form

$$G(z) \sim -4z^3, \ z \to -\infty; \ G(z) \sim (3/z^4\pi^{1/2})\exp(-z^2), \ z \to +\infty.$$

For a specific function g(z) the integral (9) can be calculated in principle and, therefore, the function M = M(λ) can be determined. The solution of the problem in this case is completed by integrating (5):

$$t = \int_0^\lambda d\xi/k\,[M(\xi) - Q],$$

(11)

which associates the parameter λ with time. The functions f and M (the volume of the crystals) also become determined by virtue of (11).

It can be shown that $M(\lambda) \to 0$ as $\lambda \to \infty$ for bounded functions g(r). As a result, the function M at some $\lambda = \lambda_k$ becomes equal to Q (the inequality $M(0) = \int_0^\infty g(r)r^3 dr > Q$, which ensures the necessary driving force of the process, should be satisfied at the initial time). A finite time will correspond to the value $\lambda = \lambda_k$ by virtue of the divergence of the integral (11). Making λ tend to λ_k in Eq. (8), we obtain the equilibrium distribution function f*. We note that the equilibrium function f* cannot be obtained from Eq. (1) for $\partial f/\partial t = 0$ since the other terms of this equation become zero at $t \to \infty$, i.e., the equation becomes completely degenerate.

Passing to the limit $B \to 0$ in Eqs. (8) and (9), we obtain the relations

$$f(r, \lambda) = g(r + \lambda), \quad M = \int_0^\infty r^3 g(r + \lambda)\,dr,$$

(12)

which are easily reduced to the fundamental equation of [9] upon satisfaction of Eqs. (2) and (5).

Power Dependence of the Dissolution Rate on the Crystal Size. Besides the kinetic regime considered above, more complicated laws governing the variation of crystal size are used in dissolution theory and practice, e.g., a diffusion law when $v \sim 1/r$ and a law $v \sim 1/r^{1/2}$ [8, 20]. Both examples come under the general case of the power law

$$v = k(M - Q)/r^\omega,$$

(13)

which has also been used to analyze crystallization processes [3, 21]. Usually, $\omega \in (0, 1)$. We note that, as before in (2), we use a linear dependence of the dissolution rate on the supersaturation, although the calculations in essence are not made more complicated with the more general formula $v \sim \varphi(s)$, $\varphi(0) = 0$, where φ is an arbitrary function (in practice, usually a power function). This is mainly because the linear law is obtained from theoretical considerations and the power formulas indicated above have exponents close to unity.

For the fluctuations of the crystal growth rate we use another relation

$$D = rv/E, \quad E = \text{const},$$

(14)

which was proposed in [22]. Introducing a new variable by analogy with (5)

$$d\tau/dt = k(M - Q)/E, \quad \tau|_{t=0} = 0,$$

(15)

we rewrite Eq. (1) as

$$r^{\omega-1}\frac{\partial f}{\partial \tau} = -\frac{\partial^2 f}{\partial r^2} + \frac{(1 - \omega + E)}{r}\frac{\partial f}{\partial r} - \frac{\omega E}{r^2}f.$$

(16)

In contrast to Eq. (6) the point r = 0 here is a singular point and this circumstance imposes the condition that the solution of Eq. (16) that is bounded as $r \to 0$ (in fact, $f \to 0$ is obtained) be chosen. The other auxiliary conditions for this equation remain the same as before.

Equation (16) admits separation of variables and thus allows a more general solution to be constructed by superposition of the partial solutions: $A(\beta) \exp(-\beta^2 \tau) r^\alpha J_{\omega-\alpha}(\beta r^\gamma)$, where β is the separation parameter, $\alpha = (\omega - E)/2$, $\gamma = (\omega + 1)/2$, $J_{\omega-\alpha}(z)$ is a Bessel function of the first kind [23], which ensures that the solution is bounded (with allowance for the factor r^α) as $r \to 0$, and $A(\beta)$ is a function which is determined by the initial condition (4). Upon executing the Fourier method while taking the known integrals into account [23], we obtain the following expression for the function f

$$f(r, \tau) = \frac{r^\alpha}{2\gamma\tau} \int\limits_0^\infty g(\xi) \xi^{\omega-\alpha} I_\nu \left[\frac{(\xi r)^\gamma}{2\tau\gamma^2} \right] \exp\left[-\frac{r^{2\gamma} + \xi^{2\gamma}}{4\tau\gamma^2} \right] d\xi, \tag{17}$$

where $\nu = (E + \omega)/(\omega + 1)$, and $I_\nu(z)$ is a modified Bessel function. Calculation of the third moment of the function f gives

$$M = \frac{\Gamma\left(\dfrac{\omega+4}{\omega+1}\right)}{\Gamma(\nu+1)} (4\gamma^2\tau)^{\frac{3}{\omega+1}} \int\limits_0^\infty g(\xi) \xi^{2(\omega+E)} \; F\left(\frac{E+\omega-3}{\omega+1},\; 1+\nu,\; -\frac{\xi^{2\gamma}}{4\tau\gamma^2}\right) d\xi, \tag{18}$$

where $F(\alpha, \gamma, z)$ is a degenerate hypergeometric function of the arguments given above, and $\Gamma(z)$ is the gamma function. This relation reduces the solution of the entire problem to the integration of (15).

Functionally Invariant Solutions of the Dissolution Equation. Equations (5), (8), (9) and (15), (17), (18) obtained above in principle solve the respective dissolution problems, by reducing them to the successive execution of several integrations. This cannot be completed in the general case, however, by analytical means. The greatest inconvenience is presented by the numerical calculation of the integrals (9) and (18), which are then used in the secondary integrations of (11) and (15). More detailed information about the properties of the fundamental dissolution equations can be obtained in some limiting cases, although even then it is often necessary to determine the characteristics of the initial function $g(r)$ [7-9].

Any exact solutions, which sometimes permit general conclusions to be made about the properties of the solutions and can also serve as test variants for checking numerical calculations, are of great assistance in the analysis of processes described by nonlinear equations. For problems of dissolution and other similar processes in the case when the particle growth rate does not fluctuate Buevich [8, 9] constructed a family of functionally invariant exact solutions of the fundamental equation which, with a successful approximation of the initial function, make it possible to analyze real processes fairly reliably and to make general conclusions. It proved possible to use such methods also in related problems of crystallization from solutions with allowance for the fluctuations of the crystal growth rate [16, 17]. In the simplest variant the essence of the method is reduced to separation of variables in the nonlinear equation and subsequent construction of a set of exact solutions by superposition of the simplest solutions. The validity of this scheme, which is usually applied to linear equations, stems mainly from the fact that the nonlinear part of the differential operator is due to a factor which depends only on time and appears in the expression for the dissolution rate after calculation of the third moment of the distribution function. In the analytically more involved situation with fluctuations of the crystal growth rate at a constant fluctuation coefficient, direct separation of variables in the fundamental equation may prove to be impossible and a solution must then be constructed on the basis of other considerations [17, 24]. Separation of variables is possible for Eq. (1) with the laws we have used for the dissolution rate fluctuations, i.e., Eqs. (3) and (14), but with the $1/r^\omega$ power law for the dissolution rate we arrive at Bessel functions. Such a set of solutions will be somewhat inconvenient for the purposes of approximating the given auxiliary conditions; accordingly, we use functionally simpler expressions which involve an exponential function.

Let us take Eq. (1) with laws (13) and (14) for the dissolution rate and its fluctuations. As the "fundamental" solutions we use

$$f_i(r,\ t) = r^\omega \theta_i(t) \exp\left[-\frac{\varkappa_i(t)\, r^{\omega+1}}{\omega+1}\right]. \tag{19}$$

A similar relation, but with a constant $\varkappa_i$, was used in [8] in an analysis of the problem for D = 0. Looking for the solution in the form of the sum of n terms of the form (19), upon substitution of this sum in Eq. (1) we obtain the system ($\varphi = k[M - Q]$)

$$d\theta_i/dt + \varphi\theta_i\varkappa_i\,[1 + (1 + 2\omega)/E] = 0, \tag{20}$$

$$d\varkappa_i/dt + \varphi(\omega + 1)\,\varkappa_i^2\,/E = 0,\ i = 1,\ 2,\ ...,\ n, \tag{21}$$

such that Eq. (1) will be satisfied if the system is satisfied. Dividing Eq. (20) by Eq. (21) and integrating the resulting expression, we find the relation between the functions θ_i and $\varkappa_i$:

$$\theta_i(t) = c_i\,[\varkappa_i(t)]^\mu,\ \mu = (E + 1 + 2\omega)/(1 + \omega),\ i = 1,\ 2,\ ...,\ n. \tag{22}$$

In exactly the same way, if we separate, e.g., the first equation from the system (21) and divide it by the i-th, upon integration we obtain

$$\varkappa_i(t) = \varkappa_1(t)/[1 - \beta_i\varkappa_1(t)],\ \beta_1 = 0,\ i = 1,\ 2,\ ...,\ n. \tag{23}$$

The system of 2n equations (20), (21) thus in fact reduces to one equation for the function $\varkappa_1(t)$. It is necessary beforehand to calculate the third moment of the distribution function of the solution sought. We have

$$M = \int_0^\infty r^3 f\,dr = \Gamma\left(\frac{\omega + 4}{\omega + 1}\right)(\omega + 1)^{3/(\omega+1)} \sum_{i=1}^n c_i\,[\varkappa_i(t)]^\delta = \tag{24}$$

$$= \Gamma\left(\frac{\omega + 4}{\omega + 1}\right)(\omega + 1)^{3/(\omega+1)}\,[\varkappa_1(t)]^\delta \sum_{i=1}^n c_i\,[1 - \beta_i\varkappa_1(t)]^{-\delta},$$

where $\delta = (\omega + E - 3)/(\omega + 1)$. The function $\varkappa_1(t)$ is now found by simple integration from the equation

$$d\varkappa_1/dt + k\,[M(\varkappa_1) - Q]\,\varkappa_1^2\,(1 + \omega)/E = 0. \tag{25}$$

The initial condition $\varkappa_1(0)$ must be proven for Eq. (25). Taking this into account, we have at our disposal 2n arbitrary constants c_i, β_i, $\varkappa_i(0)$, the appropriate choice of which can result is a good approximation of the given auxiliary conditions. The functions $\varkappa_i$ are best arranged according to value: $\varkappa_i(t) > \varkappa_j(t)$ for $i > j$. Moreover, in order for the third moment to exist it is necessary that all $\varkappa_i$ be positive. Form this and from Eq. (23) it follows that $\beta_i > \beta_j$ for $i > j$ and $\beta_n < 1/\varkappa_1(0)$. Since dissolution begins when M > Q, it follows from Eq. (25) that the function $\varkappa_1$ and hence all $\varkappa_i$, i = 1, 2, ..., n, will decrease monotonically and, therefore, in accordance with Eq. (22) the functions θ_i will also decrease (c_i are assumed to be positive quantities). As is seen from formula (24), the volume of the crystals will decrease (i.e., dissolution will occur) only when E > 3 $-\omega$, which is consistent with the condition that the fluctuation coefficient be small, as indicated above.

In constructing solutions by means of the superposition of the terms of (19), we can use either an infinite number of these terms or a continuous distribution in the form of an integral over some set. Here we confine ourselves to an analysis of the continuous distribution over the entire interval $r \in (0, \infty)$. In the given case we can use the formulas for the direct and inverse Laplace (Riemann–Mellin) transformations for the relation between the initial function (4) and the "spectral" density of the distribution. Operations similar to those above lead to the relation

$$\frac{f}{r^\omega} = \int_0^\infty N(y)\,y^{-\mu}\varepsilon\,[\varkappa(t),\ y]^\mu \exp\left\{-\frac{r^{\omega+1}\varepsilon\,[\varkappa(t),\ y]}{\omega + 1}\right\}dy, \tag{26}$$

where

$$\varepsilon\,[\varkappa\,(t),\ y]^{-1} = [\varkappa\,(t)]^{-1} - [\varkappa\,(0)]^{-1} + y^{-1}. \tag{27}$$

Substituting $t = 0$ into this and using Eq. (4), we see that the function $N(y)$ has as its Laplace transform the function $g(r)/r^\omega$ which can be expressed in terms of the variable $\xi = r^{\omega+1}/(\omega + 1)$ in order to determine $N(y)$ from the Riemann—Mellin formula. The properties of pairs of functions which effect a direct and an inverse Laplace transformation are well known and we assume that the function $g(r)$ is such that $N(y)$ exists. Thus, the function $N(y)$ can be assumed to be known. For the complete solution of the problem we must still determine the time variation of the function $\varkappa(t)$, for which purpose we calculate the third moment of the solution (26):

$$M = \Gamma\left(\frac{\omega + 4}{\omega + 1}\right)(\omega + 1)^{3/(\omega+1)} \int\limits_0^\infty N\,(y)\,y^{-\mu}\varepsilon\,[\varkappa\,(t),\ y]^\delta\,dy. \tag{28}$$

The functions ε and $\varkappa$ satisfy the same equation

$$d\varkappa^{-1}/dt = k\,(\omega + 1)\,\{M\,[\varkappa\,(t),\ \varkappa^0] - Q\}, \tag{29}$$

but different initial conditions: $\varkappa|_{t=0} = \varkappa^\circ$, $\varepsilon|_{t=0} = y$. The use of just one of these functions (ε) without a relation between (27) and $\varkappa(t)$ would make it difficult to find the latter because the integration in (28) is performed over the variable y and y would be performing two functions, that of a variable of integration and that of the initial condition for Eq. (29), written for the function ε. Now the dependence on y in the integral (28) is explicit and in some cases it can be calculated analytically and the entire expression for M depends on one free parameter, $\varkappa^\circ$. Thus, Eq. (27) makes it possible to "unlink" the problem of finding the variable defined by Eq. (29), which is a great advantage even when the integral in (28) has to be found numerically.

Having integrated (29) for the initial condition $\varkappa_{t=0} = \varkappa^\circ$, we obtain the complete solution of the problem, which depends on one parameter $\varkappa^0$ and satisfies the initial condition (4). Such arbitrariness in finding the solution is apparently attributable to the existence of a differential operator of the second order in the variable r. This parameter can be arranged so as to satisfy other requirements imposed on the solution.

In conclusion, we point out that a set of "self-similar" solutions can also be constructed for the kinetic regime with conditions (2) and (3) in the following form:

$$f = \sum_{i=1}^n \theta_i\,(t)\exp\,(-\varkappa_i r), \tag{30}$$

where the numbers $\varkappa_i$ are constant. The calculations are similar in many ways to those carried out in [8, 9, 16] and, therefore, are not given here. We note, however, that as before when the family (19) was used, the coefficient of dissolution rate fluctuations should be small enough, $B < 1/\max\{\varkappa_i\}$, for the volume of the crystals to decrease with time.

NOTATION

B, E, k, Q, and ω, physical constants in the laws of dissolution and dissolution rate fluctuations; c_i and β_i, integration constants; D, coefficient of dissolution rate fluctuations; $f(r, t)$ and $g(r)$, crystal size distribution function and its initial value; f_i, set of "self-similarity" solutions; M, third moment of the distribution function; $N(y)$, spectral density of self-similar solutions; r, r_m, and r_*, crystal size, its maximum value, and the size of new-phase nuclei; s, undersaturation; t, time; v, dissolution rate; and θ_i and $\varkappa_i$, functions of time in the self-similar solutions.

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