

## RESEARCH ARTICLE

# Approximate analytical solution of the integro-differential model of bulk crystallization in a metastable liquid with mass supply (heat dissipation) and crystal withdrawal mechanism

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This paper is devoted to an approximate analytical solution of an integro-differential model describing the process of nucleation and growth of particles in crystallizers, taking into account the thermal-mass exchange with the environment and the removal of product crystals from the metastable medium. The method developed in this work for solving model equations (kinetic equation for the particle size distribution function and balance equations for temperature/impurity concentration) is based on using the saddle point method for calculating the Laplace-type integral. It is shown that the degree of metastability of the liquid decreases with time at a fixed value of the mass inflow from the outside (heat flow to the outside). The crystal size distribution function has the form of an irregular bell-shaped curve, which increases with the intensification of heat and mass exchange with the environment.

## KEYWORDS:

integro-differential equations, applied mathematical modeling, particulate assemblages, metastability removal, crystal growth

## 1 | INTRODUCTION

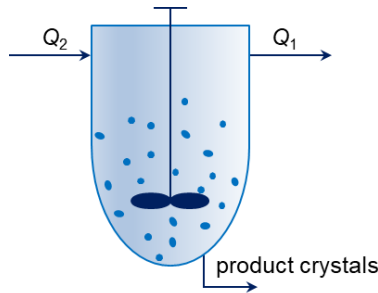
It is well known that in the metastable region of phase transformation, the nucleation and evolution of new phase nuclei and the growth of larger dendrite-like branching structures can occur.<sup>1–12</sup> As this takes place, the bulk formation of a new phase happens on the centers of crystallization (small foreign inclusions) under strong supercooling (supersaturation) of the liquid.<sup>13–16</sup> The evolution of dendritic crystals usually occurs in the presence of a temperature (concentration) gradient and interphase boundary.<sup>17–22</sup> Often in natural and technological processes, there is a simultaneous phase transition in the volume of the metastable medium and in a certain spatial direction, which is determined by the temperature and/or concentration gradient.<sup>23–25</sup> Mathematical modeling of such a process is an extremely difficult task since the model consists of integro-differential equations in the solid and liquid phases, the boundary between which moves with time.<sup>26–28</sup> As is well known, there are no generally accepted methods for solving such problems with moving boundaries and each process requires the development of an individual approximate approach to solving the problem. Here, for example, the differential series and saddle point methods may be mentioned.<sup>29–34</sup>

One of the most important technological processes widely used in metallurgy, chemical, and pharmaceutical industries is crystal growth in metastable melts and solutions of a crystallizer.<sup>35–38</sup> In this process, it is important to remove crystals of the crystallization product from the metastable liquid. Mathematically, this means an additional summand in the Fokker-Planck type kinetic equation.<sup>39–42</sup> On the other hand, to maintain smooth operation of the crystallizer, it is necessary to ensure a constant inflow of mass (impurity), as well as to carry out thermal extraction. Mathematically, this means having mass and heat

fluxes in the balance equations for temperature and impurity concentration in the liquid.<sup>39–42</sup> Accounting for these additional effects greatly complicates the mathematical formulation of the problem of phase transformation in the metastable liquid of a crystallizer. The present paper is devoted to the development of one possible approach to solving this problem.

## 2 | THE MODEL EQUATIONS

Let us consider a mathematical model of the process of nucleation and crystal growth in a metastable liquid, taking into account the withdrawal of product crystals, an external source of mass, or heat sink (figure 1). Let us write down the kinetic equation for the crystal distribution function taking into account the term responsible for the crystal runoff as well as the diffusion term describing the "diffusion" of the distribution function  $f(r, t)$  in the crystal size space<sup>43–47</sup>



**FIGURE 1** A sketch of the crystallization process with mass supply ( $Q_2$ ), heat removal ( $Q_1$ ) and product crystals withdrawal.

$$\frac{\partial f}{\partial \tau} + \frac{\partial}{\partial r}(gf) + \tilde{g}f = \frac{\partial}{\partial r} \left( D \frac{\partial f}{\partial r} \right), \quad r > r_*, \quad \tau > 0, \quad (1)$$

where  $\tau$  and  $r$  are the time and spatial variables,  $g$  is the growth rate of spherical crystals,  $\tilde{g}$  represents the withdrawal rate of product crystals from a crystallizer,  $r_*$  is the size of newly born particles,  $I$  is the nucleation rate, which is the function of melt supercooling (or supersaturation in the case of solutions). The right-hand side of equation (1) is responsible for the "diffusion" mechanism of growing particles in the space of their sizes.

Equation (1) should be supplemented with the boundary condition, which defines the flux of particles overcoming the nucleation barrier, i.e.

$$gf - D \frac{\partial f}{\partial r} = I, \quad r = r_*, \quad \tau > 0. \quad (2)$$

Here the nucleation rate represents an exponential dependence of the energy barrier level:<sup>15</sup>  $I = I_* \exp(-p\Delta\theta_0^2/\Delta\theta^2)$ ,  $p = 16\pi\gamma_i^3\theta_p/(3L_V^2k_B\Delta\theta_0^2)$  in the case of supercooled melts, and  $I = I_* \exp(-p \ln^{-2}(C_l/C_p))$ ,  $p = 16\pi\gamma_i^3M_s^2/(3\rho_s^2R_g^2k_B\theta_s^3)$  in the case of supersaturated solutions. These nucleation rates are known as the Weber-Volmer-Frenkel-Zel'dovich (WV-FZ) nucleation kinetics. We assume that the pre-exponential factor  $I_*$  is constant,  $\Delta\theta = \theta_p - \theta_l > 0$  and  $\Delta C = C_l - C_p > 0$  stand for the dimensional supercooling and supersaturation of liquid ( $\Delta\theta_0 = \theta_p - \theta_0$  and  $\Delta C_0 = C_0 - C_p$  are their initial levels),  $\theta_p$  represents the constant temperature of crystallization,  $\theta_l$  is the liquid temperature,  $C_l$  is the solute concentration,  $C_p$  is the concentration at saturation,  $\gamma_i$  represents the constant surface tension,  $L_V$  stands for the latent heat of phase transformation,  $k_B$  is the Boltzmann constant,  $\rho_s$  is the density of solid phase,  $R_g$  is the absolute gas constant, and  $\theta_s$  stands for the temperature of solution.

It is important to note that in addition to the exponential expressions mentioned above for the nucleation rate, there are also the following empirical formulas, called Meirs kinetics:  $I = I_*(\Delta\theta)^p$  for supercooled liquids and  $I = I_*(\Delta C)^p$  for supersaturated solutions.

If we are dealing with crystallization from a supercooled melt, then the thermal balance equation taking into account the heat exchange with the environment will look like

$$\rho_m C_m \frac{d\theta_l}{d\tau} = Q_1 + 4\pi L_V \int_{r_*}^{\infty} f(\tau, r) r^2 \frac{dr}{d\tau} dr, \quad r > r_*, \quad \tau > 0, \quad (3)$$

where  $Q_1 < 0$  represents a given heat flux leaving the metastable liquid through the crystallizer walls,  $\rho_m$  and  $C_m$  stand for the density and specific heat of liquid mixture.

When studying the crystallization process from a supercooled solution into which an impurity is added from outside to keep the crystallizer running smoothly, the mass balance equation has the form

$$\frac{dC_l}{d\tau} = Q_2 - 4\pi C_p \int_{r_*}^{\infty} f(\tau, r) r^2 \frac{dr}{d\tau}, \quad r > r_*, \quad \tau > 0, \quad (4)$$

where  $Q_2$  represents the mass flux entering the metastable solution.

To close equations (1)-(4) we need to set additional initial and boundary conditions in the form of

$$f = 0, \quad \Delta\theta = \Delta\theta_0, \quad \Delta C = \Delta C_0, \quad \tau = 0; \quad f = 0, \quad r \rightarrow \infty. \quad (5)$$

It is important to note that it is very difficult to accurately determine the mutual Brownian diffusion coefficient  $D$ . However, a good approximation confirmed by practice is the proportionality of this coefficient to the particle growth rate, i.e.<sup>43</sup>  $D = d_1 g(\tau, r)$ , where  $d_1$  is a constant coefficient.

In this paper, we will use for a certainty the quasi-stationary expressions for the particle growth rate derived earlier by Alexandrov and Malygin.<sup>15</sup> These evolutionary laws look like  $g = \beta_* \Delta\theta / (1 + \beta_* q r)$ ,  $q = L_V / \lambda_l$  for supercooled melts, and  $g = \beta_* \Delta C / (1 + \beta_* q r)$ ,  $q = C_p / D_l$  for supersaturated solutions. Note that the constant  $\beta_*$  stands for the kinetic coefficient,  $\lambda_l$  and  $D_l$  represent the temperature conductivity and diffusivity in the liquid phase.

### 3 | ANALYTICAL SOLUTIONS

For the convenience of constructing analytical solutions, we will use the following dimensionless variables

$$t = \frac{\tau}{\tau_0}, \quad s = \frac{r}{l_0}, \quad F = l_0^4 f, \quad g_0 = \frac{\tau_0 g}{l_0}, \quad u_0 = \frac{d_1}{l_0}, \quad z = s - s_*, \quad s_* = \frac{r_*}{l_0}, \quad (6)$$

where

$$w = \frac{\Delta\theta}{\Delta\theta_0}, \quad b_1 = \frac{4\pi L_V}{\rho_m C_m \Delta\theta_0}, \quad Q = -\frac{Q_1 \tau_0}{\rho_m C_m \Delta\theta_0}, \quad I_0 = I(\Delta\theta_0), \quad l_0 \left( \frac{\beta_* \Delta\theta_0}{I_0} \right)^{1/4}, \quad \tau_0 = (\beta_*^3 \Delta\theta_0^3 I_0)^{-1/4}$$

in the case of supercooled melts, and

$$w = \frac{\Delta C}{\Delta C_0}, \quad b_1 = \frac{4\pi C_p}{\Delta C_0}, \quad Q = \frac{Q_2 \tau_0}{\Delta C_0}, \quad I_0 = I(\Delta C_0), \quad l_0 \left( \frac{\beta_* \Delta C_0}{I_0} \right)^{1/4}, \quad \tau_0 = (\beta_*^3 \Delta C_0^3 I_0)^{-1/4}$$

in the case of supersaturated solutions.

The model equations (1)-(5) can be rewritten in dimensionless form using variables (6) as

$$\frac{\partial F}{\partial t} + \frac{\partial}{\partial z} (g_0 F) + \alpha l_0 g_0 F = u_0 \frac{\partial}{\partial z} \left( g_0 \frac{\partial F}{\partial z} \right), \quad z > 0, \quad t > 0, \quad (7)$$

$$\frac{dw}{dt} = Q - b_1 w \int_0^{\infty} (z + s_*)^2 \frac{F dz}{1 + Q_0(z + s_*)}, \quad t > 0, \quad (8)$$

$$F - u_0 \frac{\partial F}{\partial z} = \frac{\exp(-p\varphi(w))}{g_0} = J(w), \quad z = 0; \quad F = 0, \quad w = 1, \quad t = 0; \quad F = 0, \quad z \rightarrow \infty, \quad (9)$$

where  $\tilde{g} = \alpha g$  ( $\alpha$  is a constant coefficient),  $\varphi = 1 - w^{-2}$  and  $\varphi = \ln w$  in cases of WVFZ and Meirs kinetic mechanism for supercooled systems. If we are dealing with supersaturated systems, these functions take the form  $\varphi = \ln^{-2}(1 + w_p^{-1}) - \ln^{-2}(1 + w/w_p)$  (WVFZ) and  $\varphi = \ln w$  (Meirs).

The rate of particle growth in dimensionless variables (6) reads as

$$g_0 = \frac{ds}{dt} = \frac{w}{1 + Q_0 s}.$$

Here  $Q_0 = \beta_* q l_0$ . Integrating this equation, we obtain

$$g_0(t) = w(t) \left( 2Q_0 \int_{t_*}^t w(t_1) dt_1 + (1 + Q_0 s_*)^2 \right)^{-1/2}$$

if  $t \geq t_*$  and  $g_0(t) = 0$  if  $t < t_*$ , where  $t_*$  is the time of appearance of a newly born particle with radius  $s_*$ . This formula demonstrates that  $g_0$  depends on  $t$  only.

Let us rewrite the kinetic equation (7) through the modified variable  $x$  in the form of

$$\frac{\partial F}{\partial x} + \frac{\partial F}{\partial z} + \alpha l_0 F = u_0 \frac{\partial^2 F}{\partial z^2}, \quad x = \int_0^t g_0(t_1) dt_1. \quad (10)$$

Equation (10) supplemented with the boundary and initial conditions (9) can be solved using the Laplace transform method.<sup>16</sup> The final result reads as

$$F = \exp\left(\frac{z}{2u_0}\right) \int_0^x \frac{J(x-y)}{2u_0} \exp(-a_1 y) \left[ \frac{2\sqrt{u_0}}{\sqrt{\pi y}} \exp\left(\frac{-z^2}{4u_0 y}\right) - \exp\left(\frac{z}{2u_0} + \frac{y}{4u_0}\right) \operatorname{erfc}\left(\frac{z}{2\sqrt{u_0 y}} + \frac{\sqrt{y}}{2\sqrt{u_0}}\right) \right] dy, \quad (11)$$

where  $a_1 = \frac{1 + 4u_0 \alpha l_0}{4u_0}$ .

Now substituting the dimensionless growth rate  $g_0$  into the second expression (10), we arrive at

$$g_0(x) = \frac{w(x)}{1 + Q_0(x + s_*)}, \quad t(x) = \int_0^x \frac{1 + Q_0(x_1 + s_*)}{w(x_1)} dx_1. \quad (12)$$

Equations (8), (11) and (12) imply that

$$g_0 \frac{dw}{dx} = Q - b_1 w \int_0^x J(x-y) h(y) dy = Q - b_1 w \int_0^x J(y) h(x-y) dy, \quad (13)$$

$$h(y) = \int_0^\infty \frac{(z + s_*)^2}{2u_0 [1 + Q_0(z + s_*)]} \exp\left(\frac{z}{2u_0} - a_1 y\right) \left[ \frac{2\sqrt{u_0}}{\sqrt{\pi y}} \exp\left(\frac{-z^2}{4u_0 y}\right) - \exp\left(\frac{2z + y}{4u_0}\right) \operatorname{erfc}\left(\frac{z}{2\sqrt{u_0 y}} + \frac{\sqrt{y}}{2\sqrt{u_0}}\right) \right] dz.$$

The integral contribution in equation (13) can be evaluated using the saddle-point technique.<sup>31,32</sup> Assuming that  $p \gg 1$ , we come to<sup>31,32</sup>

$$\int_0^x \bar{h}(x, y) \exp[p\varphi(w(y))] dy \approx p^{-1/2} \exp[p\varphi(w(y_0))] \sum_{k=0}^\infty a_k(y_0, x) p^{-k}, \quad \bar{h}(x, y) = \frac{h(x-y)}{g_0(y)}, \quad (14)$$

$$a_k(x, y_0) = -\frac{2^{2k+1}}{(2k)!} \Gamma\left(\frac{2k+1}{2}\right) \left[ H(y_0, y) \frac{\partial}{\partial y} \right]^{2k} (\bar{h}(x, y) H(y_0, y))_{y=y_0}, \quad H(y_0, y) = \frac{\sqrt{\varphi(w(y_0)) - \varphi(w(y))}}{\varphi'(w(y))},$$

where  $\Gamma$  stands for the gamma function, and  $y_0$  represents a point where  $\varphi(w(y_0))$  has a maximum value. Taking into consideration that  $dw/dx = 0$  at this point, we obtain from (13):

$$\frac{Q}{w_0} = b_1 \int_0^{x_0} J(x_0 - y) h(y) dy, \quad w_0 = w(x_0). \quad (15)$$

Let us now change the variable in equation (15) using the inverse function  $y(w_0)$ :

$$\frac{Q}{w_0} = b_1 \int_1^{w_0} \exp(p\varphi(w_1)) (1 + Q_0 s_*) w_1^{-1} (dy/dw_1) h[y(w_1)] dw_1. \quad (16)$$

Now differentiation of equation (16) with respect to the variable supercooling (supersaturation)  $w$  gives the differential equation with separable variables. Integrating this equation, we get  $w_0(x_0)$

$$\int_1^{w_0} \left( \frac{Q}{w_1} \right)' w_1 \exp(-p\varphi(w_1)) dw_1 = b_1 \int_1^{x_0} (1 + Q_0 s_*) h(y) dy = \tilde{H}(x_0). \quad (17)$$

Considering the Meirs kinetic mechanism, we obtain from (17) the following supercooling  $w_0 = (1 + p\tilde{H}(x_0)/Q)^{-1/p}$ .

Now expanding (13) in Taylor's series, we arrive at the expression for  $w_0$  and  $x_0$  in the form of  $dw/dx = Q/g_0$ , where  $w = 1$  at  $x = 0$ . It means that  $w_0$  and  $x_0$  in (14) are known. Now considering only the main term of the asymptotic solution in (14), we arrive at the one-point Cauchy problem

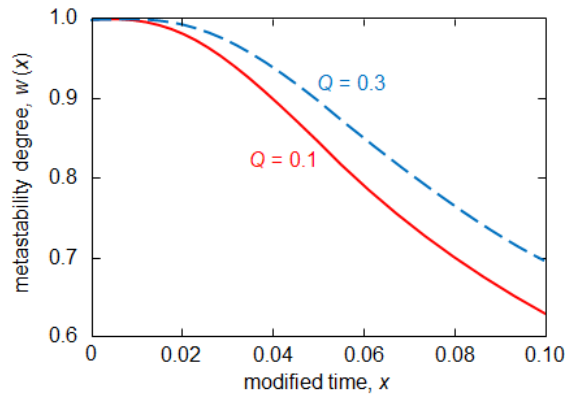
$$\frac{dw}{dx} = \frac{1}{g_0} [Q + 2b_1 w p^{-1/2} \exp(p\varphi(w(y_0))) \Gamma(1/2) \bar{h}(x, y_0) H(y_0, y_0)]. \quad (18)$$

where  $w = 1$  at  $x = 0$  and  $y_0 = x_0$ .

At the end of this section, we note specifically that formulas (11), (12), and (18) define an approximate analytical solution of the problem in the parametric form ( $x$  is a parameter).

## 4 | DISCUSSION AND CONCLUSION

Figures 2 and 3 illustrate our analytical solution (11), (12) and (18). As is easily seen, the melt supercooling (supersaturation) decreases with increasing the modified time  $x$  (or the dimensionless time  $t$ ). As this takes place, the stronger the heat sink or mass influx (higher values of  $Q$ ), the higher the metastability  $w$  of the liquid. Let us especially note that if we choose the case  $Q = 0$  (turn off the heat sink or mass influx), the metastability degree  $w$  will quickly reach zero (or nearly zero) values. At such

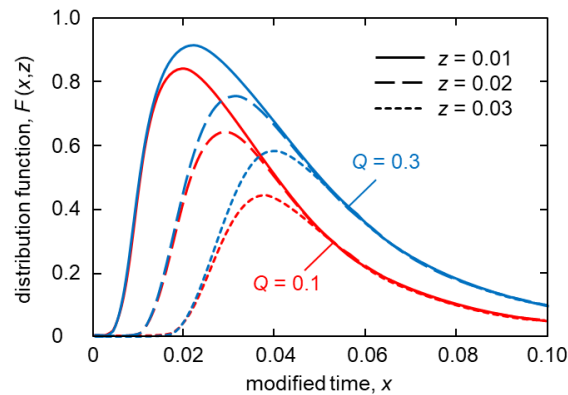


**FIGURE 2** The melt supercooling (supersaturation)  $w$  as a function of the modified time variable  $x$ . Material parameters typical for metallic melts correspond to the previous studies.<sup>15,48</sup>

low supercoolings (supersaturations) it is no longer correct to use the model of the intermediate stage of phase transformation, since the interaction between particles leads to the final stage of phase transition, when Ostwald ripening, coagulation, and agglomeration of particles take place.<sup>49–55</sup>

The crystal size distribution function illustrated in figure 3 is a bell-shaped distribution with an irregularly shaped dome. The distribution of smaller crystals at different time moments (compare the curves at different values of spatial variable  $z$ ) is higher than the distribution of larger crystals, due to the intense nucleation of new particles. As this takes place, a more intense mass inflow or stronger cooling of the liquid leads to an increase of crystals in the crystallizer and consequently of the distribution function (compare the curves at different values of  $Q$ ).

In conclusion, it seems important to note the following circumstance. The analytic theory developed in this paper, which takes into account only the main term of the asymptotic expansion by the saddle point method, can be refined by taking into



**FIGURE 3** The dimensionless distribution function  $F(x, z)$  versus the modified time variable  $x$ . Material parameters typical for metallic melts correspond to the previous studies.<sup>15,48</sup>

account more terms in the approximate calculation of the integral (14). In this case, the metastability degree  $w$  of the liquid and the crystal size distribution function  $F$  will change slightly, although the fundamental character of the behavior of these curves will remain the same. The method of approximate solution of the integro-differential model of nucleation and crystal growth developed in the present paper can be applied to more general laws of crystal growth<sup>12</sup> as well as to the phase transformation temperature shift due to the Gibbs-Thomson effect and kinetics of particle attachment to the interphase boundary.<sup>46,47</sup>

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## Author contributions

The authors contributed equally to the present research article.

## Conflict of interest

The authors declare no potential conflict of interests.

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