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Solute redistribution around crystal shapes growing under hyperbolic mass transport

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ABSTRACT

Using a model of local non-equilibrium diffusion during rapid solidification of a binary system, the isosolutal shapes of growing crystals in steady-state approximation are obtained. It is found that for crystals growing with constant velocity along a selected coordinate direction, two isosolutal growth shapes can occur. These are: the parabolic platelet in two-dimensional case and the paraboloid of revolution in three-dimensional case. In the isothermal case of diffusionless solidification, when the velocity of solidification is equal to or greater than the solute diffusive speed in the bulk system, these interfaces can have an arbitrary configuration. Special attention is given to mathematical transformations from parabolic (paraboloidal) coordinates to usual Cartesian coordinates for Ivantsov solutions extended to the case of rapid dendritic growth in which the solidification velocity V is comparable with the solute diffusion speed V_D in bulk liquid.

1. Introduction

Analytical solutions of Ivantsov [1–3] about shapes of crystals growing in concentration and heat fields play an exceptional role in solidification theory and have various practical applications. Indeed, Ivantsov solutions present zero order approximation of the stability theory of growing crystals [4,5], they are main equations for the development of dendritic growth models [6], they are the basis for the development of the theory of anisotropic growth of dendritic crystals [7] and crystals under forced convective transport [8], they present basic solutions for numerical tests [9] and they play an extraordinary role in interpretation of experimental data [10,11].

Ivantsov [1–3] and also Horvay and Cahn [12] have found solutions for seven main shapes of crystals which satisfy the balance conditions at the phase interface under diffusional transfer of heat or mass in the bulk system. These solutions were found for quasi-equilibrium conditions of growth of isotropic crystals in a non-stationary regime (the growth velocity is inversely proportional to the square root of time) or in a steady-state regime of motion with a constant velocity along a selected coordinate direction which is well-known as "dendritic problem". The latter has been intensively investigated within the non-isothermal form of crystal shapes [13] and computations of binary dendrites growing under non-isothermal conditions [6].

One of the remarkable features of the solution of Horvay and Cahn is that they directly applied curvilinear coordinates for dendritic problem as described by Ivantsov solutions. Having obtained an elegant method of analytical solution, they have found an elliptical paraboloid which is usually used in analysis of crystal growth with non-symmetric forms [14,15]. Further advancement of Ivantsov solutions was made in their application to rapid solidification problems for qualitative interpretation and quantitative description of non-linearity in the "dendrite growth velocity-undercooling" relationship [16,17]. These non-linearities with steep changes of the crystal growth velocity were, at first, obtained in kinetics of droplets solidification [11] and they required special introducing a local non-equilibrium in bulk phases which lead to an extended description of the mass transport problem. Indeed, high level of undercooling reached in small droplets provides fast propagation of the crystal-liquid interface of velocity V which can be of the order or even larger than a characteristic diffusion speed V_D in bulk phases. Therefore, using methods of extended thermodynamics [18], the dendritic problem has been reformulated so that the method suggested by Ivantsov for solution of the partial differential equations of the parabolic type for heat or solute diffusion [1–3] has been extended to obtain a solution of partial differential equation of a hyperbolic type for

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solute diffusion [19]. The hyperbolic transport equation takes into account a finite speed V_D of the atomic diffusion and its solution, applied to the isothermal dendritic problem, again results in the steady-state isosolutal shapes: the paraboloid of revolution and the parabolic platelet. These forms, however, may transform into arbitrary shapes due to degeneration of the concentration fields into homogeneous distribution if the growth velocity overcomes the diffusion speed, i.e. at $V \geqslant V_D$.

The main focus of the present article is to apply the method of Horvay and Cahn to the description based on curvilinear coordinates for the rapidly growing dendrite. This application requires special consideration by several reasons. *First*, because the non-planar parabolic interface moves with the constant velocity $V \approx V_D$ only in one-direction, a specific coordinate transformation with the scale $(1 - V^2/V_D^2)^{1/2}$ should be made for the parabolic (paraboloidal) coordinates that requires a non-trivial treatment. *Second*, presentations of the Ivantsov solutions in various forms for practical computations [6,20,21] should also be generalized to their rapid solidification case which uses a description of mass transport by the hyperbolic equation. *Third*, a suggested method for equations of the hyperbolic type can be further useful, for example, in analogous problems of gas dynamics where obtaining solutions of supersonic regimes of gas flow around the isobaric surfaces is necessary. As a consequence, the article is devoted to the analytical treatments of the rapidly growing parabolic crystals to obtain the results useful for their further application in analytical and practical computations.

2. The model

Consider the isothermal and isobaric case of solidification in which crystals grow from a chemically binary liquid. We shall neglect the slower diffusion in the solid crystal phase in comparison with the much faster diffusion in liquid and assume that the crystal-liquid interface velocity V may reach values of the order of the solute diffusion speed V_D in the bulk liquid, such that $V \sim V_D \approx 0.1 - 10$ m/s [11,16,17]. In this case, we use the model of local non-equilibrium solidification in which the transport of atoms is described by the hyperbolic equation

$$\tau_D \frac{\partial^2 C}{\partial t^2} + \frac{\partial C}{\partial t} = D\nabla^2 C. \tag{1}$$

together with the boundary condition

$$-D\nabla_n C = (C - C_S)V_n + \tau_D \frac{\partial}{\partial t}((C - C_S)V_n), \tag{2}$$

$$C_S = kC, \tag{3}$$

and with the far-field condition for the solute concentration C in liquid,

$$C|_\infty = C_0. \tag{4}$$

Here $\tau_D = D/V_D^2$ is the relaxation time of diffusion flux to its steady state, D is the coefficient of solute diffusion in liquid, V_D is the solute diffusion speed in the bulk liquid (it is considered also as speed of the front of solute diffusion profile), $\nabla_n C = \vec{n} \cdot \vec{\nabla} C$ is the normal gradient of solute concentration to the interface, $\vec{n} = (n_x, n_y, n_z)$ is the normal vector to the interface with the components n_x, n_y, n_z, $V_n = \vec{V} \cdot \vec{n}$ is the projection of the vector velocity $\vec{V}$ on the vector $\vec{n}$ and $k(V_n)$ is the coefficient of solute partitioning at the interface dependent of the normal velocity V_n.

Equation (1) is the simplest mathematical model combining the diffusive (dissipative) mode and the propagative (wave) mode of mass transport under local non-equilibrium conditions [18]. In addition to the usual "Fickian diffusion" with which a pure dissipative process can be described, Eq. (1) may predict propagative and diffusive features of the diffusion process and, therefore, terminologically, it can be characterized as "non-Fickian diffusion". Together with the conditions (2)–(4), the problem of dendrite growth has been analysed numerically for isothermal approximation [22] and under non-isothermal conditions of crystal growth [23]. Verifications of such descriptions have been made in atomistic simulations [24] and derived from the coarse-grained approach for fast transformations [25].

Further, we shall consider the dendritic problem [1–3] in which growth of the isosolutal needle-like shape proceeds with a constant value of velocity V along the selected space direction. Introducing the Cartesian coordinate system in which the z-axis coincides with the direction of the velocity vector $\vec{V}$, we analyze the problem in new Cartesian coordinates fixed to the crystal. Then, Eq. (1) takes the form

$$\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \left(1 - \frac{V^2}{V_D^2}\right)\frac{\partial^2 C}{\partial z^2} + \frac{V}{D}\frac{\partial C}{\partial z} = 0. \tag{5}$$

Using Eq. (2), the interface boundary condition leads to

$$-D\left(\nabla_n C - \frac{VV_n}{V_D^2}\frac{\partial C}{\partial z}\right) = (C - C_S)V_n.$$

Finally, taking into account the condition $V_n = n_z V$, Eq. (2) is rewritten as

$$n_x \frac{\partial C}{\partial x} + n_y \frac{\partial C}{\partial y} + n_z\left(1 - \frac{V^2}{V_D^2}\right)\frac{\partial C}{\partial z} = -(C - C_S)\frac{Vn_z}{D}. \tag{6}$$

3. Growth regimes

As experimentally found [11], drastic changes in the kinetics of rapid solidification occur around the growth velocity V comparable with the diffusion speed V_D. Therefore, considering Eq. (5) one should distinguish three qualitatively different cases of relationship between V and V_D. These describe the regimes at which the interface moves with: (*i*) the velocity smaller than the solute diffusion speed in the bulk liquid ($V < V_D$), (*ii*) the velocity equal to the solute diffusion speed ($V = V_D$) and (*iii*) the velocity greater than the solute diffusion speed ($V > V_D$).

A general solution of Eq. (5) can be expressed in terms of the eigenfunctions $e^{imx}/\sqrt{2\pi}$ and $e^{iny}/\sqrt{2\pi}$ of the Laplace operator as

$$C(x, y, z) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} e^{imx}e^{iny}f_{mn}(z)dmdn + C_0, \tag{7}$$

where m and n are the corresponding eigenvectors. As the concentration C far from the interface is equal to C_0, the functions $f_{mn}(z)$ have to be zero at $z \to \infty$. Substitution of Eq. (7) into Eq. (5) leads to the ordinary differential equation for $f_{mn}(z)$:

$$\left(1 - \frac{V^2}{V_D^2}\right)\frac{d^2 f_{mn}}{dz^2} + \frac{V}{D}\frac{df_{mn}}{dz} - (m^2 + n^2)f_{mn} = 0. \tag{8}$$

The nontrivial solutions of Eq. (8) have the form $f_{mn} \sim e^{\lambda z}$ with factors λ subject to equation

$$\left(1 - \frac{V^2}{V_D^2}\right)\lambda^2 + \frac{V}{D}\lambda - (m^2 + n^2) = 0. \tag{9}$$

3.1. Case $V < V_D$

Using Vieta's formulas which give the relation between the roots λ_1 and λ_2 and the coefficients of the quadratic Eq. (9), we have