

A relaxation time of secondary dendritic branches to their steady-state growth

E A Titova¹, D V Alexandrov¹ and P K Galenko²

¹ Department of Theoretical and Mathematical Physics, Laboratory of Multi-Scale Mathematical Modeling, Ural Federal University, Ekaterinburg 620000, Russian Federation

² Friedrich-Schiller-Universität-Jena, Physikalisches-Astronomische Fakultät, 07743 Jena, Germany

E-mail: ekatitova@mail.ru

Abstract. In the present article, we use the selection theory to estimate the non-stationarity time of evolving dendrites to their steady-state growth.

1. Introduction

The microstructure predictions in solidification processes have the deep scientific and practical roots (see [1–5] and references therein). A development of experimental methods in solidification processing made possible to reach a much broader range of measurable crystal growth velocities, temperature gradients and cooling rates [6]. For instance, splats and films can be quenched with the cooling rate of the order of 10^7 K/s, the temperature gradients can have the order of 10^8 K/s in laser annealing of sample surfaces, and containerless methods of droplets processing provide the deep undercoolings having the values of 200–400 K prior to the primary crystallization. A large driving force for transformation, arising in such methods, leads to fast solidification from a liquid metastable state as well as the high-speed solid-state transformations of metastable crystalline phases [7]. For instance, the experimentally measured solidification velocity has the order of $10^{-1} - 10^2$ m/s in droplets processed by the electromagnetic levitation facility [7, 8]. As such, the total duration of primary solidification in small droplets is rather short and estimated as [8]: $10^{-5} - 10^{-3}$ s.

For the analytical calculations of rapid solidification regimes and theoretical estimations of microstructural parameters of dendritic and eutectic crystals, different models of non-equilibrium crystallization are used, as a rule, formally developed for the steady state scenario [9–11]. However, the steady state approximation for conditions of rapid solidification of small samples (films, splats, droplets) is questionable and such approach is often criticized. A very common view is that a steady state scenario may be expected to exist but not in rapidly solidifying small samples [12]: the stationary regime of solidification can be reached at long times that is possible in usual experimental circumstances (directional solidification) or well-known classic technologies (continuous casting and low intensive brazing). In other words, during the short periods of time, the steady state is not achieved and, consequently, the quasi-stationary approximation in modeling of rapid solidification does fail. The detailed calculations of solidification regimes lead, however, to the remarkable behavior of the transient time between the non-stationary and stationary regimes of solidification: the non-stationary time sharply depends on the interface



Table 1. Material parameters for succinonitrile (SCN) and nickel (Ni)

Parameter		SCN	Ni
Melting temperature	T_A (K)	331.233 [27]	1728 [28]
Latent heat	L (J/(m ³))	$4.781 \cdot 10^7$ [27]	$2.67 \cdot 10^6$ [30]
Heat capacity	c_p (J/(m ³ K))	$2.08 \cdot 10^6$ [27]	$6.39 \cdot 10^3$ [30]
Solid-liquid interface energy	σ (J/m ²)	$8.9 \cdot 10^{-3}$ [27]	0.275 [30]
Capillary length	d_0 (m)	$2.821 \cdot 10^{-9}$ [27]	$4.92 \cdot 10^{-10}$ [29]
Surface energy stiffness	α_d (-)	0.0825 [27]	0.27 [30]
Thermal diffusivity	D_T (m ² /s)	$1.13 \cdot 10^{-7}$ [27]	$1.2 \cdot 10^{-5}$ [30]
Selection constant	σ_0 (-)	0.1 [22]	0.191 [29]
Relaxation time for $\partial\phi/\partial t$ (*)	τ_ϕ (s)	$2.5 \cdot 10^{-7}$	$5.5 \cdot 10^{-7}$
Maximum speed of ϕ -field(*)	V_ϕ^B (m/s)	11.5	25.25
Diffusion coefficient of ϕ (*)	ν_ϕ (m ² /s)	$5.5 \cdot 10^{-5}$	$3.5 \cdot 10^{-4}$

(*)The numerical value of this parameter is chosen in the present work.

velocity and undercooling in solidifying samples [13]. In particular, for the planar solid-liquid interface, it has been shown that the non-stationary time rapidly decreases as the crystal growth velocity increases taking the values of the order of milli- or even micro-seconds with the velocity of the order of centimeters per second [13].

Moreover, using the theoretical model developed in [13], it is straightforward to show that the non-stationary time sharply increases with the decrease of crystal growth velocity taking the values of the order of seconds or even minutes for the interface velocity of the order of micrometer per second. Such remarkable dependence should, obviously, exist not only for the planar interface but also for many other crystal growth shapes. As one of the main crystal growth forms is the dendritic crystal shape (for slow and rapid regimes of rapid solidification [1–3]), we provide analytical treatments for obtaining the non-stationary time which could be quantitatively estimated in comparison with the time for primary solidification of experimentally investigated samples. Our studies are limited to two dimensional dendritic growth. Many of properties of metallic samples (for instance, the time for homogenization) strictly depend on the interdendritic spaces between secondary branches of dendrite. Applicability of the steady-state models to the growth of secondary branches is an actual task. Therefore, the present analysis gives some estimations of a non-stationary period of growing secondary branches in the slow and rapid solidification conditions.

2. Relaxation time to a steady-state regime

The relaxation time τ_T characterizes how fast the dendrite tip velocity,

$$v(t) \propto A_v(t) \exp(-A_m t / \tau_T) \quad (1)$$

relaxes to the steady-state regime of dendritic growth [14]. Here $A_v(t)$ is the time-dependent amplitude of relaxation, A_m is the factor which depends on the model parameters, t is the time and the relaxation time τ_T represents a relaxation parameter to the steady state regime of thermal dendrite. Let us consider the symmetric model of dendritic growth in the positive z direction with a constant velocity v . Let $\zeta = \zeta(x, t)$ and ρ designate the instantaneous coordinate of the crystallization front and the radius of curvature of the dendrite tip, respectively. It is convenient to measure lengths in units of ρ and times in units of ρ/v . In the case of zero surface

tension, the dendrite shape represents the Ivantsov parabola [14–17]

$$\zeta_{iv} = -\frac{x^2}{2}.$$

In the case of nonzero surface tension, this solution should be modified by a shape correction $\zeta_0(x)$. The steady-state solution in the form of $\zeta_{iv} + \zeta_0(x)$ exists only if the so-called selection parameter

$$\sigma^* = \frac{d_0}{\rho P_g} = \frac{2d_0 D_T}{\rho^2 v} = \frac{d_0 v}{2D_T P_g^2} \quad (2)$$

takes a special value determined from the microscopic solvability theory [18–25]. Here

$$P_g = \frac{\rho v}{2D_T} \quad (3)$$

is the growth Péclet number, d_0 is the capillary length, and D_T is the thermal diffusivity.

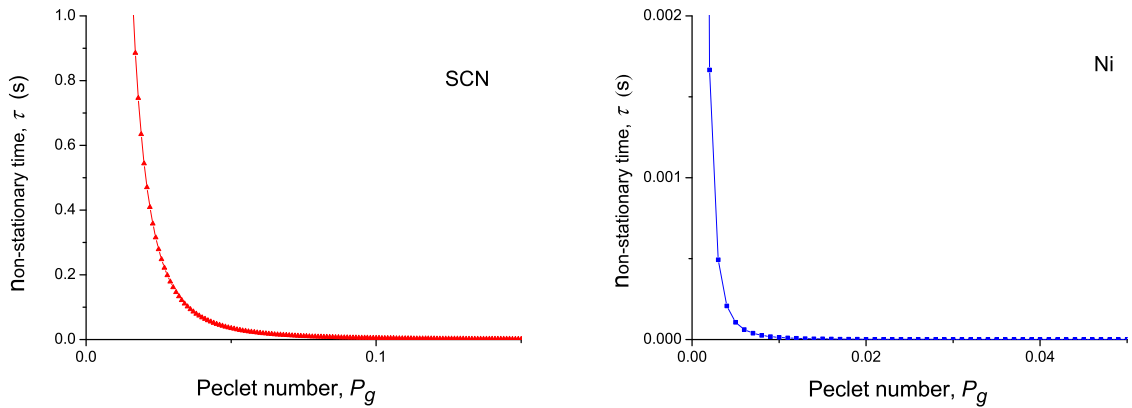


Figure 1. The thermal dendrite growth from the undercooled nickel and SCN melts. The relaxation time τ_T is a function of the growth Péclet number $P_g = \rho v / 2D_T$. Calculations are made using Eqs. (9) and (10).

It is important to know what is the real relaxation time when the parabolic dendrite shape can be considered as steady-state. In order to answer this question let us introduce a small time-dependent correction $\zeta_1(x, t)$ to the steady-state solution and present the secondary branches $\zeta(x, t)$ as:

$$\zeta(x, t) = \zeta_{iv} + \zeta_0(x) + \zeta_1(x, t). \quad (4)$$

The time-dependent addition $\zeta_1(x, t)$ has been found in [26] on the basis of WKB theory. The result is

$$\zeta_1(x, t) = f(2z)^{1/8} \sigma^{*1/4} \text{Im} \left\{ \exp \left[\frac{(2z)^{1/4}}{c^3 \sqrt{\sigma^*}} \left(1 - i \frac{z-t}{\sqrt{2z}} \right)^{3/2} \right] \right\}, \quad (5)$$

where $c = \sqrt{3/2}$, $z = x^2/2$, and f is a constant of unity order. Raising the complex number $1 - i(z-t)/\sqrt{2z}$ to the power $3/2$, separating the real and imaginary parts of expression (5) and introducing the dimensional variables

$$\tau = \frac{\rho t}{v}, \quad \xi = x\rho, \quad (6)$$

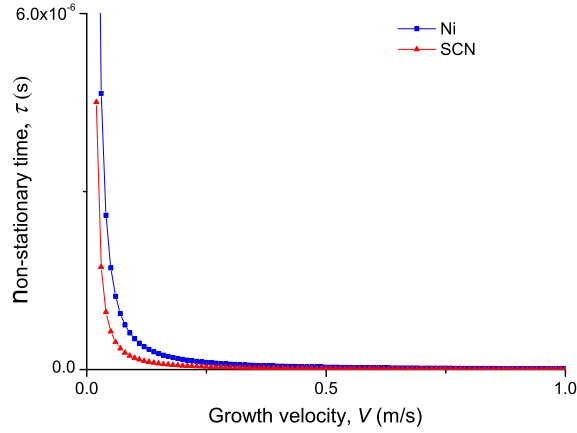


Figure 2. Growth of the thermal dendrite from the undercooled nickel and SCN melts. The relaxation time τ_T is a function of the growth velocity. Calculations are made by Eqs. (3) and (10).

one can represent the final result as

$$\zeta_1(x, t) = A \exp \left\{ - \left[(\beta\delta)^{4/3} + \left(\frac{(\beta\delta)^{2/3}}{2} - \frac{\tau}{\tau_T} \right)^2 \right]^{3/4} \right\}, \quad (7)$$

where the following designations are introduced

$$\tau_T = \frac{\rho}{v(\beta\delta)^{2/3}}, \quad \beta = \frac{\sqrt{\xi}}{c^3 \sqrt{\rho\sigma^*}}, \quad \alpha = \frac{\xi}{2\rho} - \frac{v\tau}{\xi}, \quad \delta = |\cos(\varphi/2)|,$$

$$A = f \left(\frac{\xi\sigma^*}{\rho} \right)^{1/4} \sin[\beta \sin(\varphi/2)], \quad \varphi = \arctan \left(\frac{\alpha^2 - 3}{1 - 3\alpha^2} \right). \quad (8)$$

To obtain a dependence of the thermal relaxation time τ_T of secondary dendritic branches on the growth Péclet number P_g let us express the dendrite tip velocity v from the solvability condition derived in Refs. [21–23] for arbitrary Péclet numbers

$$v(P_g) = \frac{2D_T\alpha_d^{7/4}\sigma_0 P_g^2}{d_0(1 + a_1\sqrt{\alpha_d}P_g)^2}, \quad (9)$$

where α_d is the surface energy stiffness, σ_0 is the selection constant and $a_1 = \sqrt{8\sigma_0/7}(3/56)^{3/8}$. Taking into account expression (8) and considering a vicinity of dendritic tip ($\xi \sim \rho$) one can get

$$\tau_T \approx \frac{2D_T P_g c^2}{\delta^{2/3} v^2(P_g)} \left(\frac{d_0 v(P_g)}{2D_T P_g^2} \right)^{1/3}, \quad (10)$$

with the following estimated constants: $\alpha \approx 1/2$ and $\delta \approx 0.738$. These estimations were made for the thermal dendrite with the following values of the parameters: the relaxation time $\tau_T \approx 10^{-10} \dots 10^{-7}$ (s), the scaled coordinate $\xi \approx \rho \approx 10^{-6}$ (m) and the growth velocity $v \leq 10^{-2}$

(m/s). Then, using equation (8), one gets $\alpha \approx 0.5 - v\tau/\rho$ with $v\tau/\rho \leq 10^{-3} \ll 1/2$ and obvious estimation for the parameter δ . Expression (10) determines the thermal relaxation time as a function of the growth Péclet number P_g . Together with the dendrite tip velocity v given by the selection criterion (9) this makes possible to quantitatively estimate the relaxation time to the steady-state regime of dendrite growth.

3. Conclusions

In summary, the growth of secondary dendritic branches represents a complex phenomenon with many effects and parameters for a wide range of undercoolings. Therefore, we plan to advance the present formalism for estimation of non-stationary time to include the kinetic effects [23], non-equilibrium effects (such as solute trapping, solute drag, trapping of disorder [16]) and three dimensional dendritic structure.

4. References

- [1] Trivedi R and Kurz W 1994 *Int. Mater. Rev.* **39** 49–74
- [2] Herlach D M 1994 *Mater. Sci. Eng. R* **12** 177–272
- [3] Glicksman M E, Koss M B and Winsa E A 1994 *Phys. Rev. Lett.* **73** 573–576
- [4] Alexandrova I V, Alexandrov D V, Aseev D L and Bulitcheva S V 2009 *Acta Physica Polonica A* **115** 791–794
- [5] Herlach D 2015 *Crystals* **5** 355–375
- [6] Herlach D M, Galenko P and Holland-Moritz D 2007 *Metastable solids from undercooled melts* (Amsterdam: Elsevier)
- [7] Hartmann H, Galenko P K, Holland-Moritz D, Kolbe M, Herlach D M and Shuleshova O 2008 *Journal of Applied Physics* **103** 073509
- [8] Eckler K 1992 (Ph.D. thesis), (Ruhr-Universität)
- [9] Galenko P K and Danilov D A 1997 *Phys. Lett. A* **235** 271–280
- [10] Galenko P K and Herlach D M 2006 *Phys. Rev. Lett.* **96** 150602
- [11] Gao J, Han M, Kao A, Pericleous K, Alexandrov D V and Galenko P K 2016 *Acta Mater.* **103** 184–191
- [12] Das A K 1998 *Defect and Diffusion Forum* **162** 97–118
- [13] Galenko P K and Danilov D A 2000 *J. Cryst. Growth* **216** 512–526
- [14] Langer J S 1980 *Rev. Mod. Phys.* **52** 1–28
- [15] Peclé P and Pomeau Y 1986 *Stud. Appl. Math.* **74** 245–258
- [16] Galenko P K, Danilov D A and Alexandrov D V 2015 *Int. J. Heat Mass Trans.* **89** 1054–1060
- [17] Alexandrov D V and Galenko P K 2017 *Physica A* **469** 420–428
- [18] Peclé P and Bensimon D 1987 *Nucl. Phys. B* **2** 259–270
- [19] Alexandrov D V and Pinigin D A 2013 *Technical Physics* **58** 309–315
- [20] Alexandrov D V and Galenko P K 2013 *J. Phys. A: Math. Theor.* **46** 195101
- [21] Alexandrov D V and Galenko P K 2013 *Phys. Rev. E* **87** 062403
- [22] Alexandrov D V and Galenko P K 2014 *Phys. Usp.* **57** 771–786
- [23] Alexandrov D V and Galenko P K 2015 *Phys. Chem. Chem. Phys.* **17** 19149–19161
- [24] Alexandrov D V, Danilov D A and Galenko P K 2016 *Int. J. Heat Mass Trans.* **101** 789–799
- [25] Galenko P K, Danilov D A, Reuther K, Alexandrov D V, Rettenmayr M and Herlach D M 2017 *J. Cryst. Growth* **457** 349–355
- [26] Barber M N, Barbieri A and Langer J S 1987 *Phys. Rev. A* **36** 3340–3349
- [27] Koss M B, LaCombe J C, Tennenhouse L A, Glicksman M E and Winsa E A 1999 *Metall. Mater. Trans. A* **30** 3177–3190
- [28] Hultgren R, Desai P D, Hawkins D T, Gleiser M, Kelley K K and Wagman D D 1973 *Selected values of the thermodynamic properties of the elements* (Ohio: ASM International, Metals Park)
- [29] Gao J, Kao A, Bojarevics V, Pericleous K, Galenko P K and Alexandrov D V 2016 *J. Cryst. Growth* <http://dx.doi.org/10.1016/j.jcrysgro.2016.11.069>
- [30] Galenko P K, Phanikumar G, Funke O, Chernova L, Reutzel S, Kolbe M and Herlach D M 2007 *Materials Science and Engineering A* **448–451** 649–653

Acknowledgments

This work was supported by the Russian Science Foundation (grant no. 16-11-10095).