ДЕФЕКТОСКОПИЯ, ОБЕСПЕЧЕНИЕ СВОЙСТВ МАТЕРИАЛОВ И ПРОГНОЗИРОВАНИЕ РЕСУРСА ИЗДЕЛИЙ

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Канд. физ.-мат. наук Лебединский Ю.А. ,
Канд. техн. наук Брановицкий А.М., Дементьев В.А.
(Институт технологии металлов НАН Беларуси, Могилев)

Ph. D. Lebedinsky Yu. A., Ph. D Branovitsky A. M., Dement'ev V. A.
(Institute of Technology of Metals of the National Academy of Sciences of Belarus, Mogilev)

Impurity trapping defects within dendrite-rosette morphology transformation during modeling of solidification of binary alloys under stirring

Дефекты в первичных кристаллах для дендритно-розеточной морфологической трансформации при моделировании затвердевания бинарных сплавов с перемешиванием

When primary crystals with a cubic lattice grow from the stirring binary melt, additional branches grow as well as the main dendrite branches. Dendritic morphology is replaced by rosette morphology when sufficiently strong stirring takes place. The morphology changing very influence on all properties of solidifying alloy. In particular, crystal morphology influence on propagation of waves, which used for diagnostic of internal structure of metal. Wave scattering on microstructure non-uniformity is high depend from crystals size and its morphology. So analysis of conditions of dendrite-rosette transition may have great importance on possibility of diagnostic of internal defects.

The behavior of the morphology is investigated depending on the distribution of the solute concentration, both due to stirring and due to the influence of neighboring branches. Stirring increases the solute concentration gradient in front of the phase boundary and promotes the growth of additional branches in this direction. Calculations have also been made for various values of surface tension anisotropy, including for facet crystals growth. Initially, the perturbation is always perpendicular to the surface of phase boundary, it is directed approximately along the solute concentration gradient. Subsequently, the branch bends depending on the ratio of anisotropy and the degree of stirring, which determines the magnitude of the concentration gradient. Moreover, strong temperature noise and washout degree lead to arise defects of impurity trapping.

Когда первичные кристаллы с кубической решеткой растут из перемешиваемого бинарного расплава, кроме основных ветвей дендритов растут ещё и дополнительные ветви. При достаточно сильном перемешивании дендритная морфология сменяется на розеточную. Изменение морфологии заметно влияет на все свойства затвердевающего сплава. В частности, влияние морфологии кристаллов на распространение волн используется для диагностики внутреннего строения металла. Рассеяние волн на неоднородности микроструктуры во многом зависит от размера кристаллов и их морфологии. Таким образом, анализ условий перехода дендрит-розетка может иметь большое значение для возможности диагностики внутренних дефектов.

Поведение морфологии исследуется в зависимости от распределения концентрации растворенного вещества, как из-за перемешивания, так и из-за влияния соседних ветвей. Перемешивание увеличивает градиент концентрации растворенного вещества перед фазовой границей и способствует росту дополнительных ветвей в этом направлении. Также были выполнены расчеты при различных значениях анизотропии поверхностного натяжения, в том числе для роста гранных кристаллов. Первоначально возмущение всегда перпендикулярно поверхности границы раздела фаз, оно направлено приблизительно по градиенту концентрации растворенного вещества. Впоследствии ветвь изгибается в зависимости от соотношения анизотропии и степени перемешивания, которая определяет величину градиента концентрации.

Introduction

Solidification during casting of binary alloys is often associated with the formation of primary crystals in the form of dendrites. Since the dendritic morphology is one of the reasons for the deterioration of the mechanical properties of alloys, casting methods have become widespread, in which the growth of primary crystals occurs when these crystals move in the melt and the dendritic morphology transforms to the rosette one.

Dendrite structure for many alloys also have strong scattering capability for wave propagation, especially for ultrasound waves with wave length value near crystal size. So dendrite-rosette transition can decreasing scattering and improve defects detection.

The dendritic morphology of primary crystals during solidification of binary alloys is associated with the anisotropy of the surface tension energy depending on the direction of the normal to the surface. For the case of a cubic lattice with predominant crystal growth in the [100] and analogous directions, primary crystals are dendrite with an angle of π/2 between the main axes.

Usually, the maximum solubility of the second component of the melt (solute impurity) in the solid phase of the primary crystals is less than the concentration of solute in the melt. Therefore, in front of the boundary between the solid phase and the melt, there is a zone with an increased concentration of solute in the liquid. To ensure crystal growth, the excess solute in this zone must have time to diffuse into a zone far from the boundary – the so-called diffusion-controlled growth.

In the case of a motionless melt, a number of additional branches also arise in the zone between the main ones. But since diffusion of solute from this zone is hindered, and besides, the main branches during growth additionally "dump" solute into this zone, the growth of such additional branches is suppressed. Over time, secondary branches begin to grow from the main branches.

In the case of the movement of the melt relative to the growing primary crystal, a part of the melt with a high solute concentration is removed from the zones between the main branches. Additional branches are given the opportunity for growth and the dendritic morphology becomes rosette. Typically, this type of morphology results in improved mechanical properties of the alloy. Solidification within flow is often used in practice in a wide variety of casting methods.

In the general case of the movement of the melt relative to the growing crystal, the branches of the crystal change growth direction in comparison with the stationary case. This applies to all branches: additional, secondary, branches when splitting from the main, etc. Here we will consider the case of bending of additional branches that appear in the case of rosette morphology of a growing crystal.

In this work, we briefly analyze the morphology of such growing additional branches for the two-dimensional case by the phase field method, which is perhaps the only method that can correctly describe the details of the morphology of growing crystals.

The direction of the branches depends on the ratio of surface tension anisotropy and the intensity of removal of solute. Such removal is named washout in this work. The anisotropy of the surface tension energy γ is described as

$$\gamma = \gamma_0 \left(1 + \varepsilon_4 \cos 4\varphi\right), \qquad (1)$$

where φ is the angle between the X-axis and the normal to the crystal surface, and ε4 determines the degree of

anisotropy. In this case, undercooling near such a surface due to its curvature will decrease proportionally expression [1]:

$$\gamma + \frac{\partial^2 \gamma}{\partial \varphi^2} = \gamma_0 \left(1 - 15\varepsilon_4 \cos 4\varphi\right). \quad (2)$$

With a sufficiently high anisotropy in the above expression, negative value of (2) arises for some directions φ, which contradicts the principles of thermodynamics. This means that there are no surfaces in the crystal, which have such an angle between the normal and the coordinate axis. Such crystals are called faceted [2]. That is, not all surface orientations are possible. Angle φ will be denoted φ_m when right hand of the expression (2) for undercooling equals 0.

In view of the possible strong dependence on the degree of anisotropy, calculations were also made for the such high anisotropic case of faceted crystals.

Phase-field model

The used initial phase field model for binary melt is described in detail early [3]. Here we present only the equations themselves for the phase variable ϕ and solute concentration U.

In the case of faceted crystals the phase field method requires regularization of the equation for phase field evolution [1]. So, the equation for the phase variable ϕ has the form [1,4,5] :

$$\tau\left(\varphi\right)\frac{\partial \phi}{\partial t} = \nabla \cdot \left(W\left(\varphi\right)^2 \nabla \phi\right) - \partial_x \left[W\left(\varphi\right) W_\varphi\left(\varphi\right) \partial_y \phi\right] + \partial_y \left[W\left(\varphi\right) W_\varphi\left(\varphi\right) \partial_x \phi\right] + \phi - \phi^3 - \lambda\theta\phi\left(1 - \phi^2\right)^2$$

for the case when the angle ϕ is in the range

$$\frac{\pi}{2} i + \varphi_m \le \varphi \le \frac{\pi}{2}\left(i+1\right) - \varphi_m, \; i = 0 - 3,$$

where now

$$\varphi = \arctan\left(\frac{\partial \phi}{\partial y} \Big/ \frac{\partial \phi}{\partial x}\right),$$

is angle between gradient ϕ and X-axis, W(ϕ) and $\tau(\phi)$ are proportional to 1+ ε4 cos(4φ), t – time, λ – coupling parameter, θ – thermal and concentration undercooling. Here φ is in the range of allowable directions of normal to crystal surface.

For cases of regularized values, when surface with such angle does not exist, the equations for ϕ are

$$\tau\left(\varphi_m\right)\left(\frac{\cos\varphi}{\cos\varphi_m}\right)^2 \frac{\partial \phi}{\partial t} = \tau_0 \left[\frac{W\left(\varphi_m\right)}{\cos\varphi_m}\right]^2 \phi_{xx} + \phi - \phi^3 - \lambda\theta\phi\left(1 - \phi^2\right)^2,$$

for angles

$$\left(\frac{\pi}{2} i - \varphi_m\right) \le \varphi \le \left(\frac{\pi}{2} i + \varphi_m\right), \; i = 0,2,$$

and

$$\tau\left(\varphi_m\right)\left(\frac{\sin\varphi}{\cos\varphi_m}\right)^2 \frac{\partial \phi}{\partial t} = \tau_0 \left[\frac{W\left(\varphi_m\right)}{\cos\varphi_m}\right]^2 \phi_{yy} + \phi - \phi^3 - \lambda\theta\phi\left(1 - \phi^2\right)^2,$$

for angles

$$\left(\frac{\pi}{2}i-\varphi_m\right)\le\varphi\le\left(\frac{\pi}{2}i+\varphi_m\right),\ I=1,3.$$

And the equation generalizing the diffusion equation to the case of solute propagation in the layer, which is transitional between phases:

$$\frac{(1+k)}{2}\frac{\partial U}{dt}=\nabla\cdot\left(\tilde{D}\frac{1-\phi}{2}\nabla U+j_{at}\right)+\frac{1}{2}\frac{\partial}{\partial t}\left[\phi\left[1+(1-k)U\right]\right],$$

where $\tilde{D}$ – dimensionless diffusion coefficient, k – partition coefficient.

This work also uses a model of the effect of stirring during solidification, similar to previous one [1], characterized by the parameter S – the intensity of stirring. The value S determines the zone in melt, within which stirring periodically "resets" the current solute concentration to the initial concentration in the melt. So, a value of 5% means that wherever the solute concentration is less than 1.05 relatively to initial one, it is periodically equated to unity. Forced reset of the concentration value to the initial concentration in the melt simulates a washout of solute.

At the initial stage of growth of such branches, for the case when the growth is not yet influenced by neighboring branches, the direction of their growth is determined by the ratio of the degree of anisotropy and the degree of washout. The latter may be determined as the thickness of the transition zone between the solid phase boundary and the region of space where the "excessive" concentration of solute is reset (white zone in further pictures).

Further the calculations are presented for a binary melt with a dimensionless initial undercooling of 0.1, $\tilde{D}=3$, a spatial step of 0.8 relative to the characteristic interface width, and a time step of 0.01 relative to the relaxation time.

Results

A number of calculations were performed for different cases of generation of additional branches. In the first case, calculations were made for different degrees of anisotropy for the same washout 7% (Fig. 1). For this case a small level of thermal noise was added with its rapid change in time and space relative to the characteristic growth time and crystal size. The main crystal axes are rotated by an angle π/4 relative to the coordinate axes – this improves the accuracy for the case of a regularized solution [1]. Note that for the numerical implementation of the regularized solution, there may exist small sections of the crystal surface with a "forbidden" orientation, when the right-hand side of (2) is negative. This is due to the numerical approximation of the derivatives near the points where the surface changes orientation in a jump [2]. It is also seen that for all high degrees of anisotropy, the effect of stirring is practically the same.

With an increase in anisotropy, the main branches are pulled out and, accordingly, additional ones are suppressed. Note that the growth of additional branches is observed only for the case of anisotropy of 0.03, which is characteristic for the crystallization of metals (Fig. 2). It is seen that secondary branches in the region of the neck of the dendrite, usually suppressed by the growth of the main branches, can form additional branches.

Because influence of neighboring additional branches both of them begin grow in parallel directions.

The morphology of additional branches significantly depends on the noise level, that is, fluctuations during crystal growth. Noise arises due to temperature and concentration fluctuations of the melt, due to crystal growth defects, due to irregular washout, and many other reasons. Figure 3 shows the evolution with low-frequency noise in time and space, with medium and strong thermal noise. As the noise increases, the crystal grows faster. Additional branches appear in large numbers, which begin to split after thickening.

The morphology also strongly depends on the degree (intensity) of the washout (Fig. 4). Calculations were made at a constant value of anisotropy 0.03 and for different degrees of washout. With an increase of the washout intensity, the growth of crystals accelerates, the number of additional branches and their splitting increases. In case of 20% the growth is very fast, so the solute may be trapped inside primary crystal.

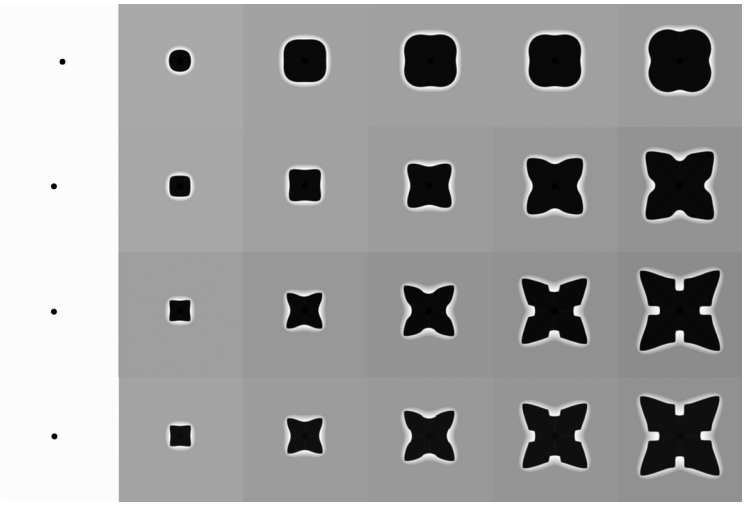

Fig. 1. Evolution of a crystal during stirring at different degrees of surface tension anisotropy. The rows of drawings correspond to different values of ε4 : 0.01, 0.03, 0.09, 0.17(the regularized solution for this case) from top to down. The black area is the solid phase of the primary crystal, the gray area is the melt zone with stirring – where the reset of concentration of solute takes place, the white area is the melt without stirring (without resetting) in the zone near the crystal boundary. The picture in the leftmost column show an initial round crystal nucleus.

Fig. 2. Evolution of a crystal with anisotropy of 0.03. Washout is 7%.

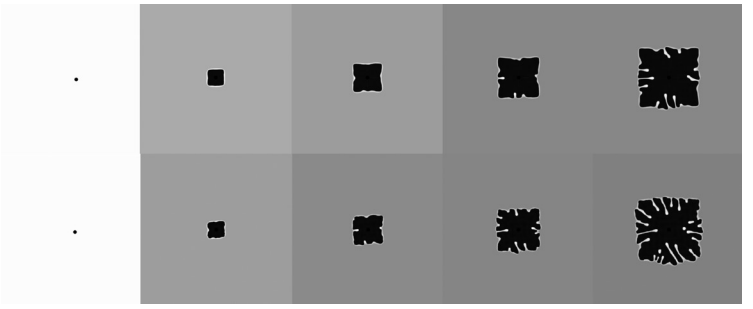

Fig. 3. Evolution of growth with medium (upper row) and strong (lower row) temperature noise. The washout and anisotropy parameters are the same as in Fig. 2.

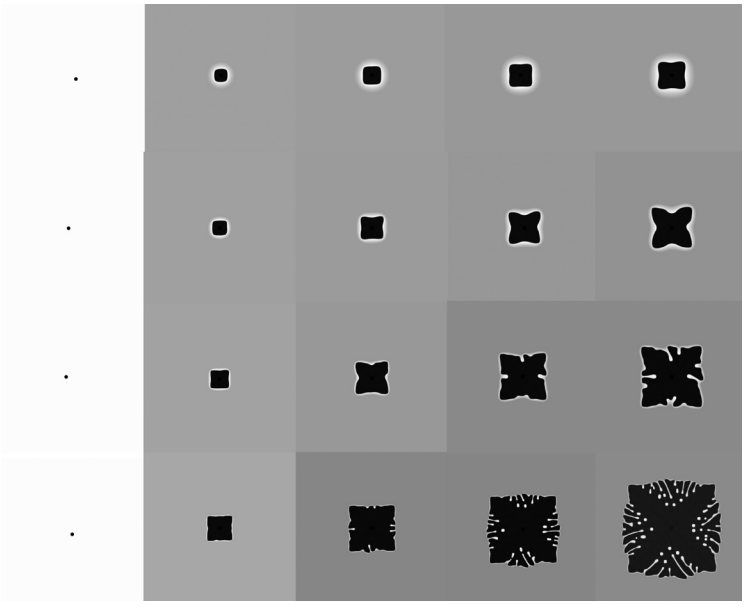

Fig. 4. Evolution of morphology for different washout degree: 2%, 5%, 10%, 20% from top row to down one.

In figure 4 it is clearly seen that at the initial stage, with a sufficiently high washout intensity, the perturbation of the crystal boundary is perpendicular to this boundary. Further, leaving the border region with a high concentration gradient solute, the crystal begins to bend in accordance with the anisotropy of surface tension, trying to minimize the surface energy.

Besides, strong temperature noise (Fig.3) and/or washout degree (20%, Fig. 4) may lead to formation of closed impurity areas inside growing crystal.

Conclusions

1. The appearance of additional branches is characteristic only for the values of the anisotropy coefficient of surface energy, ε4 =0.02-0.03, which is characteristic for metallic crystals.

2. With increasing of temperature fluctuations, the growth of primary crystals is accelerated and the number of additional branches increases. Subsequently, upon expansion, the additional branches can split into two.

3. Strong temperature noise and washout degree are factors of arising defects of impurity trapping.

References

1. Eggleston, J. J., McFadden, G., Voorhees P. W. A phase-field model for highly anisotropic interfacial energy. Physica D 150, 2001, P. 91вЂ“103.

2. Debierre J., Karma A., Celestini F., Guerin R. Phase-field approach for faceted solidification. https://doi.org/10.1103/PhysRevE.68.041604

3. Lebedinsky Yu.A., Branovitsky A.M., Dement'ev V.A. An approximate model of the process of non-dendritic morphology formation in solidification of binary melt under stirring. Nonlinear phenomena in complex systems. Volume 22, Issue4, 2019, P. 367-374. DOI: 10.33581/1561-4085-2019-22-4-367-374.

4. Kasajimaa H., Naganoa E., Suzuki T., Kim S., Kim W. Phase-field modeling for facet dendrite growth of silicon. Science and Technology of Advanced Materials 4, 2003, P. 553-557.

5. Chen Z., Wang Z., Gu X., Chen Y., Hao L., Jos de Wit, Jin K. Phase-field crystal simulation facet and branch crystal growth. Applied Physics A (2018) 124:385