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TRANSFORMATION KINETICS FOR INSTANTANEOUS NUCLEATION IN THE FINITE VOLUME – APPLICATION OF STATISTICAL THEORY OF SHIELDING

KINETYKA PRZEMIANY PRZY NATYCHMIASTOWYM ZARODKOWANIU W MAŁEJ OBJĘTOŚCI – ZASTOSOWANIE STATYSTYCZNEJ TEORII EKRANOWANIA

The framework of the Kolmogorov-Johnson-Mehl-Avrami (KJMA) theory is applied usually for the study of transition kinetics when the processes are ruled by nucleation and growth. This theory accurately describes only the transitions with the identical convex shape of new nuclei with the identical growth velocity distribution at an interface of the growing grains. The infinite initial volume of the mother phase is one of the indispensable conditions for the above theory. The proposed earlier extension of KJMA theory (statistical theory of the shielding growth) enlarges the scope of its application and eliminates the above limitation. The model of the transformation kinetics in the space of finite volume has been analyzed and discussed.

Keywords: KJMA theory, instantaneous nucleation, finite volume, shielding

W badaniach kinetyki procesów kontrolowanych zarodkowaniem i wzrostem skutecznie wykorzystuje się statystyczną teorię krystalizacji, podwaliny której zostały założone przez Kołmogorova, Jonhsona, Mehla i Avramiego (teoria KJMA). Powyższa teoria opisuje badane zjawiska precyzyjnie w przypadku, gdy wszystkie nowopowstające obiekty mają identyczny rozkład prędkości wzrostu na powierzchni (podobny kształt geometryczny), są wypukłe, a w przypadku anizotropii kształtu są identycznie zorientowane w przestrzeni. Jednym z założeń powyższej teorii jest nieskończona objętość zanikającego materiału. Zaproponowane wcześniej rozszerzenie teorii KJMA (statystyczna teoria ekranowanego wzrostu) zwiększa zakres zastosowania klasycznych równań, pokonując niektóre ograniczenia. W artykule przedstawiono badania kinetyki wzrostu ziaren nowej fazy w przypadku małej objętości fazy macierzystej.

1. Introduction

The mechanism that drives numerous transition processes is the nucleation and growth of elementary objects of a new substance within the substance subject to transformations. The phenomena of this type are numerous and include crystal physics, metallurgy, polymer physics, ferroelectric domain switching, magnetization and metastability in statistical physics models, phase transitions in particle physics as well as biology or ecological landscapes [1]. No matter how much these phenomena may differ, changes in the volume of the transformed fractions are described by the same statistical theory.

The theoretical fundamentals of the mathematical formulae used nowadays were developed by Kolmogorov [2]. Comprehensive case studies of the transformations are described in publications written by Johnson and Mehl [3] and Avrami [4]. This article discusses the case disregarded by the classical statistical theory. This is the case of particles growth in the spacemen of size comparable with a final grain size.

The general equation of the statistical theory of solidification, called the Kolmogorov equation, enables us to predict the real transformed fraction volume from the, so called, extended specific volume (Ω):

$$V(t) = 1 - \exp(-\Omega(t)), \tag{1}$$

where t – the time.

The value of Ω is calculated from some geometrical rules, taking into account the space dimension, the shape, size and quantity of particles in a unit volume but disregarding certain limitations resulting from their interaction [2]. Some of the conditions indispensable to satisfy the above mentioned equation are: an equal velocities of growth of all the grains of a single phase in a given direction and at a given time instant, randomly

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distributed spatial nucleation of grains, concave shape of the grains, infinitive initial volume of the primary phase. In reality, these conditions are not always satisfied, and actual kinetics of the process differs from that determined by equation (1). Anisotropic particle formation, finite size of initial substance and non-uniform spatial distribution result in a reduction of the transformation rate [5]. The similar effect gives a concave shape of growing particles [6]. Different physical phenomena that provide to deviations from the preconditions given by [2] lead to the modification of the equation (1) [7].

Numerous attempts are known which aim at an improvement of the statistical theory of phase transformations. The purpose of this improvement is the correct prediction of transition kinetics in situations when the above mentioned preconditions are not satisfied. For instance, KJMA model is used successfully for the description of the crystallization processes in bulk glasses the, however, this model cannot be applied and must be substituted by another empirical equation in the case of powered samples [8].

Availability of the framework of the KJMA model has been analyzed in [9] in order to study the restricted growth of a solid phase from a fluid for different conversion degrees. The analytical theory for continuous nucleation and one-dimensional (1D) growth in two-dimensional (2D) system, and 2D growth in three-dimensional (3D) system has been developed in [10] that is based on the Monte Carlo simulations. This type of growth leads to hard impingement and obtains strong deviations from the traditional KJMA theory. An analytical method for calculating volume fractions of phases in a system with several simultaneously growing different phases was proposed in [11]. In this paper growth of the grains for more than one phase in 2D and 3D has been treated. The kinetics for the growth of more than one phase with different velocities has been investigated analytically for 1D systems and numerically for 2D systems in [12].

Another example of high difference between the KJ-MA model prediction and real product fraction is a diffusion limited growth with a continuous nucleation of new phase. Velocity of each grain borders migration in this case decreases with time from the nucleation instant. The tasks of diffusion growth have been investigated in papers [6, 13].

The study of the non-random nucleation's influence on the deviation of the KJMA kinetics has been presented in [14] that is based on the Monte Carlo method for 3D systems.

The peculiar kind of transition when each nucleus, once nucleated, grows instantaneously to a fixed size and than stops growing was a subject of paper [15, 16]. Transformation kinetics of anisotropic particles growing in thin films has been analyzed in [17]. The anisotropic growth of randomly oriented grains and finite volume of the space (grains radii are comparable with the overall volume) were discussed in [18]. The phenomenological renormalization of the KJMA function has been proposed.

Special scaling low has been introduced in [1] in order to exploit the KJMA theory in finite volumes.

Yet, as reported by Kooi [10], all improvements mostly are not of a general character but refer to some specific cases only.

It has been proved [6] that the most frequent cause of deviations from the traditional KJMA theory is the effect of shielding (sometimes the *screening* term is used). The example of shielding of the growing grain with a faster migration of its outer border by the slower ones was discussed in [19] by means of the developed mean-field theory.

The influence of shielding effect for the varied interface movement velocity has been discussed in [20]. Computer simulations of the shielding effect in the case of anisotropic growth were the subject of the papers [21, 22]. It was shown that higher anisotropy of randomly oriented elliptical grows provides bigger overestimation of the equation (1) results. In [23] simple analytical expressions have been obtained for the blocking and phantom effects due to shielding of 1D growth with distribution of growth velocities. Taking into account shielding effects the three stages of the anisotropic particle growth have been shown in [24] : early stage of growth practically without any interaction between the grains, freeze of the fastest growth direction on the next stage and the growth controlled by the interfaces with the lowest migration rate at last stage.

The solution presented in further part of this study is based on the statistical theory of the screened growth [25-26]. This theory has been used for the transition kinetics prediction in the cases of anisotropy and concave particles and for growth of grains with a vary velocity. Goodness of prediction of shielded growth has been checked by cellular automata simulation for the 2D radial growth case of two kinds of grains with different velocities [27].

This study presents the ability of this theory in the case of growth of the instantaneously nucleated and uniformly distributed isotropic grains in space of finite volume.

2. Shielding rate of the growth in the finite volume

When the particles growth is not limited by the spacemen border the extended specific volume Ω may be

calculated as a sum of extended volume of each grain, taking into account they multiple overlapping (see Fig. 1). The fraction of the non-transformed volume in this case is equal to $\exp(-\Omega(t))$. When the grains are growing in the space of finite volume (eg. bounded by the spacemen border) the equation (1) gives us the overestimated result, even through all another precondition of

[2] were satisfied. Near the shield (specimen boundary) the extended volume calculated according to [2] will include the outer extended volume of the grains nucleated in the sample near the border (gray field at the fig. 1) and extended volume of the phantom grains nucleated outside (dashed lines at the fig. 1).



Fig. 1. Grain nucleated in the specimen volume (solid circle lines) and phantoms nucleated on the outside (dashed lines), vertical line - specimen boundary

It has been assumed [25] that we know the function S(u, t) determining the field of an external boundary of the extended grains the migration of which at a given time instant t takes place at a velocity not greater than u. For the faceted and spherical grains this function is continuous in intervals while for other non-faceted grains it is of a continuous character. In its continuous intervals S(u, t) the function S'(u, t) is equal to its partial derivative $\partial S(u, t)/\partial u$ and at the points of discontinuity where S(u, t) has a jump equal to the size of the respective crystal facets $\Delta S(u_F, t)$, therefore, this function assumes the value :

$$S'(u_F, t) = \Delta S(u_F, t)/u_F,$$
(2)

where u_F – the respective velocity on the surface.

When the interface velocity tends to zero the S'(u, t) function goes to infinity, but for motionless border product of these values bounded :

$$\Delta S(u_F, t) = \lim_{u_F \to 0} (S'(u_F, t) \cdot u_F).$$
(3)

The velocity of the extended grain growth can be expressed with Stieltjes integral :

$$\frac{\partial\Omega}{\partial t} = \int_{0}^{u_m} uS'(u,t)du + \sum_i u_{Fi}^2 S'(u_{Fi},t), \qquad (4)$$

where the second term allows for the faceted or spherical growth velocity, while the first term allows for the non-faceted growth velocity.

The extended volume is determined by the integration of equation (4) after the time :

$$\Omega(t) = \int_{0}^{t} \left(\int_{0}^{u_{m}} uS'(u,\tau) \, du + \sum_{i} u_{F_{i}}^{2} S'(u_{F_{i}},\tau) \right) d\tau, \quad (5)$$

where u_m – the maximum migration velocity of the boundary.

The shielding rate in the case of one-, two-, and three-dimensional growth is given in papers [25, 26]. If,

within the integration range, there are points of discontinuity of the function S(u), Stieltjes integral applies, and the shielding rate for velocity u_2 is :

- for 1D :

$$\frac{\partial \ln S'(u_2,t)}{\partial t} = -\left(\int_{u_{\min}}^{u_2} (u_2 - u_1) S'(u_1,t) \, du_1 + \sum_i (u_2 - u_{F_i}) S'(u_{F_i},t) \, u_{F_i}\right) \tag{6}$$

– for 2D :

$$\frac{\partial \ln S'(u_{2,t})}{\partial t} = -\frac{u_{2}}{\pi} \left(\int_{u_{\min}}^{u_{2}} \left(\sqrt{1 - \left(\frac{u_{1}}{u_{2}}\right)^{2}} - \left(\frac{u_{1}}{u_{2}}\right) \arccos\left(\frac{u_{1}}{u_{2}}\right) \right) S'(u_{1}, t) \, \mathrm{d} \, u_{1} + \sum_{i} \left(\sqrt{1 - \left(\frac{u_{F_{i}}}{u_{2}}\right)^{2}} - \left(\frac{u_{F_{i}}}{u_{2}}\right) \arccos\left(\frac{u_{F_{i}}}{u_{2}}\right) \right) S'(u_{F_{i}}, t) \, u_{F_{i}} \right)$$
(7)

- for 3D :

$$\frac{\partial \ln S'(u_2,t)}{\partial t} = -\frac{1}{4u_2} \left(\int_{u_{\min}}^{u_2} (u_2 - u_1)^2 S'(u_1) \, \mathrm{d} \, u_1 + \sum_i (u_2 - u_{F_i})^2 S'(u_{F_i},t) \, u_{F_i} \right),\tag{8}$$

where : u_2 – the velocity of growth of the screened surface, u_1 – the integration variable and summation was performed for all surfaces F_i with the growth velocity \hat{U}_{Fi} less than u_2 .

Next we will analyze the case of instantaneous nucleation with an isotropic grain growth with a constant velocity in the sample of finite volume. Let the ratio of the outer border of specimen and of its volume is equal to F_0 . Hence from (3) the product $S'(u_F, t)u_F$ is equal to F_0 . In this case the shielding effect takes place on the outer border of sample where $u_F = 0$. Equations (6)-(8) may by written now as

$$\frac{\partial \ln S'(u_2, t)}{\partial \tau} = -u_2 F_0 \tag{9}$$

$$\frac{\partial \ln S'(u_2, t)}{\partial t} = -\frac{u_2}{\pi} F_0 \tag{10}$$

$$\frac{\partial \ln S'(u_2, t)}{\partial \tau} = -\frac{u_2}{4} F_0 \tag{11}$$

thus, the general view of shielding rate equation for the 1D, 2D, and 3D growth in finite volume bordered by motionless surface is :

$$\frac{\partial \ln S'(u_2, t)}{\partial \tau} = -k_{S,D} u_2 F_0, \qquad (12)$$

where : shielding coefficient values are : $k_{S1} = 1$, $k_{S2} = 1/\pi$ and $k_{S3} = 1/4$.

3. Prediction of the Transition Kinetic in the Finite Volume

The subjects of the next case studies are the instantaneous nucleation and isotropic growth in 1D, 2D and 3D samples with a thickness equal to 2d (for 2D and 3D other dimensions of the sample are infinitively large). The specific outer surface value for this sample is equal to

$$F_0 = 1/d.$$
 (13)

All extensive parameters of growing grains are reduced to volumetric (3D), areal (2D) or linear (1D). Consequently, the extended specific volume (Ω) is a dimensionless quantity. Units of the S(u, t) and F_i is 1/L, and units of the grain numbers (*n*) is L^{-D}.

Growth rate of the extended grains with the u_2 velocity may be written according to eq. (4) as :

$$\frac{\partial\Omega}{\partial t} = u_2^2 S'(u_2, t). \tag{14}$$

The extended volume is determined by the integration of this equation (4) after the time:

$$\Omega(t) = \int_{0}^{t} u_2^2 S'(u_2, \tau) d\tau.$$
 (15)

Growth of grains is screened on the extended boundary of specimen with a shielding rate from eq. (12).

For the 2D and 3D isotropic growth the boundary of the extended grains is changed with the growing radius

of these grains and from geometrical relations for the partially screened grains with instant nucleation follows:

$$\frac{\partial \Delta S\left(u_{2},t\right)}{\partial R_{2}} = k_{F,D} \frac{\Delta S\left(u_{2},t\right)}{u_{2}t},$$
(16)

where coefficients are equal to $k_{F1} = 0$, $k_{F2} = 1$ and $k_{F3} = 2$.

Using definition (2) the above equation after the division by u_2 can be written as :

$$\frac{\partial S'(u_2,t)}{\partial R_2} = k_{F,D} \frac{S'(u_2,t)}{u_2 t}.$$
 (17)

Since in the examined case the growth velocity $u_2 = dR_2/dt$ is time-independent, equation (15) for anisotropic growth is reduced to the following form :

$$\Omega(t) = u_2^2 \int_0^t S'(u_2, \tau) d\tau.$$
 (18)

The rate of changes in $S'(u_2, t)$ depends on the two competitive processes, i.e. an increase of the grain dimensions and shielding of the surface :

$$\frac{\mathrm{d}S'(u_2,t)}{\mathrm{d}t} = \frac{\partial S'(u_2,t)}{\partial R_2} \frac{\mathrm{d}R_2}{\mathrm{d}t} + S'(u_2,t) \frac{\partial \ln S'(u_2,t)}{\partial t}.$$
 (19)

On substituting to this equation the derivatives (12) and (17) we obtain :

$$\frac{\mathrm{d}S'(u_2,t)}{\mathrm{d}t} = S'(u_2,t) \left(\frac{k_{F,D}}{t} - k_{S,D} u_2 F_0\right). \tag{20}$$

The separation of variables and integration of this equation enables calculation of the non-screened boundary of grains :

$$\ln S'(u_2, t) = \ln t^{k_{F,D}} - k_{S,D} u_2 F_0 t + C$$
(21)

Since at the instant of growth due to a small size of the grain boundaries (equal to $S'(u_2, t)u_2$)) shielding can be neglected, we have :

$$\lim_{t \to 0} S'(u_2, t) = \begin{cases} 2nu_2^{-1} & \text{for } 1D\\ 2\pi nt & \text{for } 2D\\ 4\pi nu_2 t^2 & \text{for } 3D \end{cases}$$
(22)

and it is possible to determine the integration constant C:

$$C = \begin{cases} \ln(2nu_2^{-1}) & \text{for } 1D\\ \ln(2\pi n) & \text{for } 2D\\ \ln(4\pi nu_2) & \text{for } 3D \end{cases}$$
(23)

Finally, from equation (2)

$$S'(u_2,t) = \begin{cases} \frac{2n}{u_2} \cdot \exp(-u_2F_0t) & \text{for } 1D\\ 2\pi nt \cdot \exp\left(-\frac{u_2F_0t}{\pi}\right) & \text{for } 2D \\ 4\pi nu_2t^2 \cdot \exp\left(-\frac{u_2F_0t}{4}\right) & \text{for } 3D \end{cases}$$
(24)

According to equation (18) now we can calculate the history of the grains' extended volume when growing in the finite volume :

$$\Omega(t) = \begin{cases} 2nu_2 \int_{0}^{t} \exp(-u_2 F_0 \tau) d\tau & \text{for } 1D \\ 2\pi nu_2^2 \int_{0}^{t} \tau \exp\left(-\frac{u_2 F_0 \tau}{\pi}\right) d\tau & \text{for } 2D \quad (25) \\ 4\pi nu_2^3 \int_{0}^{t} \tau^2 \exp\left(-\frac{u_2 F_0 \tau}{4}\right) d\tau & \text{for } 3D \end{cases}$$

When the grains grow in the infinite space $(F_0 \rightarrow 0)$ the results of above equations correspond to the given by classical KJMA equation for instant nucleation :

$$\Omega_{KJMA}(t) = \begin{cases} 2nu_2^t & \text{for } 1D \\ \pi nu_2^2 t^2 & \text{for } 2D \\ 4/3\pi nu_2^3 t^3 & \text{for } 3D \end{cases}$$
(26)

For the growth in the finite volume bounded by outer spacemen walls ($F_0 > 0$) integration of eqn. (25) with taking into consideration the relation between F_0 and spacemen width *d* gives :

$$\Omega(t) = \begin{cases} 2nd\left(1 - \exp\left(-\frac{u_{2}t}{d}\right)\right) & \text{for } 1D\\ 2\pi^{3}nd^{2}\left(1 - \exp\left(-\frac{u_{2}t}{d\pi}\right)\left(\frac{u_{2}t}{d\pi} + 1\right)\right) & \text{for } 2D\\ 256\pi nd^{3}\left(2 - \exp\left(-\frac{u_{2}t}{4d}\right)\left(\left(\frac{u_{2}t}{4d}\right)^{2} + 2\left(\frac{u_{2}t}{4d}\right) + 2\right)\right) & \text{for } 3D \end{cases}$$
(27)

Let *R* is a "dimensionless size of the extended grains" : $R = u_2 t/d$ for 1D, $R = u_2 t/\pi d$ for 2D, and $R = u_2 t/4d$ for 3D. The lag between the extended vol-

ume in the case of non-limited growth and growth in the finite volume is equal to :

$$\frac{\Omega(t)}{\Omega_{KJMA}(t)} = \begin{cases} R^{-1}(1 - \exp(-R)) & \text{for } 1D\\ 2R^{-2}(1 - \exp(-R)(R+1)) & \text{for } 2D\\ 3R^{-3}(2 - \exp(-R)(R^2 + 2R + 2)) & \text{for } 3D \end{cases}$$

The predicted results for the extended volume lags between unbounded growth and bounded one for 1D, 2D and 3D are shown in fig. 2. The lag of extended volume bounded growth comes significant when the dimensionless size of the extended grains amount to more than 0,1. Transition time (and respective dimensionless grain size) depends on the grain density.



(28)

Fig. 2. The lag between the extended volume in the case of non-limited growth and growth in the finite volume

The influence of the outer border of spacemen on the kinetics of transition may be shown using the simulation of bounded and unbounded growth by eqn. (1) where extended volume will calculated by eqns. (26) for free growth and (28) for bounded one. The calculation results for 1D, 2D and 3D are shown in fig. 3-5. Dashed lines present non-restricted growth in the infinite space and solid lines obtain for finite size spacemen for the same grain densities.

As it can be seen on the diagrams, the maximal lag between the transformed volume fraction between unbounded and bounded growth is not too large (about 10 %). Cases of the 1D growth with a law grain density

looks strange at first glance because of transition "freezing" when the volume fraction of initial substance is far from zero value. The reason of these arrests (shown in fig. 3) is the high probability of zero nuclei in the 1D sample of 2*d* length. According to probability density of Poisson distribution these values are equal to p=0,368 for mean value 1, p=0,135 for mean value 2 and 4 p=0,018for 4.

The results of figures 3-5 may be used for the study of transition kinetics when the processes are ruled by nucleation and growth in the case of thin wall and in the surface layers.

4. Results and comments



Fig. 3. The lag between unbounded 1D growth and growth bounded by finite size of spacemen; number – mean grain number for the spacemen length 2d



Fig. 4. The lag between unbounded 2D growth and growth bounded by finite size of spacemen; number – mean grain number for the spacemen area $(2d)^2$



Fig. 5. The lag between unbounded 3D growth and growth bounded by finite size of spacemen; numbers – mean grain number in the volume $(2d)^3$

Because of common nature of preconditions, the obtained results may be used for the kinetics of the nucleation and growth ruled transition in non-homogenous materials (composites, powder, porous or cellular materials, nano-materials) when transition takes place only in one of the components and grains can't go across the border.

5. Conclusions

The statistical theory of the screened growth eliminates some limitations of the KJMA theory of phase transformation and extends the field of KJMA theory application. An example of the application of this theory has been presented in description of the bounded growth of the instantaneously nucleated isotropic grains for the 1D, 2D and 3D growth in finite volume.

Proposed relations (28) enable to simulate analytically the kinetics of processes controlled by nucleation and growth in finite volume bounded by outer border in the framework of the Kolmogorov-Johnson-Mehl-Avrami theory.

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