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ScienceDirect
Trans. Nonferrous Met. Soc. China 22(2012) 895−900
Transactions of Nonferrous Metals Society of China
www.tnmsc.cn

Impact of anisotropic growth on kinetics of solid-state phase transformation

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Received 9 September 2011; accepted 4 January 2012

Abstract: Based on the statistical analysis of blocking effect arising from anisotropic growth, the anisotropic effect on the kinetics of solid-state transformation was investigated. The result shows that the blocking effect leads to the retardation of transformation and then a regular behavior of varying Avrami exponent. Following previous analytical model, the formulations of Avrami exponent and effective activation energy accounting for blocking effect were obtained. The anisotropic effect on the transformation depends on two factors, non-blocking factor γ and blocking scale k, which directly acts on the dimensionality of growth. The effective activation energy is not affected by the anisotropic effect. The evolution of anisotropic effect with the fraction transformed is taken into account, showing that the anisotropic effect is more severe at the middle stage of transformation.

Key words: transformation kinetics; anisotropic growth; blocking effect; Avrami exponent

1 Introduction

Transformation kinetics involving nucleation and growth of anisotropic particles is a topic of practical importance due to its relevance to a variety of applications which require anisotropic material by their very nature [1]. However, the classical treatments of transformation kinetics by JOHNSON, MEHL, AVRAMI, KOLMOGOROV (JMAK) [2−4] are usually derived for isotropic particles or for aligned anisotropic particles (where no odd blocking effect is found). The calculation of the kinetics of transformation involving anisotropic particles is a much more challenging problem than that for isotropic particles, due to the blocking effect arising from anisotropic growth. SHEPILOV [5] gave one treatment of the blocking effect in one-dimension (1D), which did not employ the mean-field approach usually used for JMAK analyses. Subsequently, SHEPILOV III and BAIK [6] discussed the blocking in a broader context, though only limited numerical results were given. On the basis of the statistic derivation of JMAK theory, BIRINE and WEINBERG [7−9] formulated the growth of anisotropic particle (especially elliptically shaped particle) and the overall kinetics of transformation predominantly for 1D; only pre-existing nuclei were assumed in 2D and the possibility that particles grew around each other was excluded. With Monte Carlo method, PUSZTAI and GRÁNÁSY [10] and KOOI [11,12] studied the mutual blocking of anisotropically growing particles up to all relevant orders, and KOOI [11,12] proposed an analytical model to describe the blocking effect. On the basis of KOOI's model, the deviations from JMAK-like kinetics due to the anisotropic effect were investigated further by LIU and YANG [13]. There is another option to extend the mathematical formulation of the JMAK theory, adding (one or more) new variables that provide freedom to improve the agreement in case that anisotropic growth occurs [14,15].

Following the JMAK statistical consideration, a stochastic treatment accounting for the blocking effect arising from anisotropic growth was proposed, and analytical models for solid-state transformation where a particle undergoes 1-scale blocking, k-scale blocking and infinite-scale blocking were developed [16]. On this basis, the present study is aimed at discussing the effect of anisotropic growth on the solid-state transformation via nucleation and growth. As known, anisotropic effect leads to the retardation of transformation, which can be

Foundation item: Project (2011CB610403) supported by the National Basic Research Program of China; Project (51125002) supported by the National Funds for Distinguished Young Scientists of China; Project (51071127) supported by the National Natural Science Foundation of China; Projects (09-QZ-2008, 24-TZ-2009) supported by the Free Research Fund of State Key Laboratory of Solidification Processing, China; Project (CX201008) supported by the Doctorate Foundation of Northwestern Polytechnical University, China
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DOI: 10.1016/S1003-6326(11)61262-4

evaluated from the varying Avrami exponent. New expressions for Avrami exponent n_{new} and effective activation energy Q_{new} subjected to the anisotropic effect were obtained, the evolution of anisotropic effect during the transformation was taken into account.

2 Theoretical background

In the JMAK description, the nucleation and growth are modeled as two statistical processes. The original derivation of JMAK equation rests on calculating the probability that a randomly chosen point in space (e.g. the origin point O) remains untransformed in a given time t. The probability that a particle nucleated at time τ grows to the origin point O at time t is expressed as [3]:

$$\mathrm{d}x_{\mathrm{e}} = \dot{N}(\tau)Y(\tau,t)\mathrm{d}\tau \tag{1}$$

where $\mathrm{d}x_{\mathrm{e}}$ is the differential form of the extended fraction x_{e}; $\dot{N}$ is the steady-state nucleation rate per unit volume; $\dot{N}(\tau)\mathrm{d}\tau$ is the probability for a particle nucleated in the time interval [τ, τ+dτ] per unit volume; $Y(\tau, t)$ is the volume of a particle at time t when it is nucleated at time τ. Accordingly, $q(t)$, the probability of the random point O untransformed at time t is obtained as [3,6]:

$$q(t) = \exp[-\int_0^t \dot{N}(\tau)Y(\tau,t)\mathrm{d}\tau] \tag{2}$$

Thus, the JMAK equation describing the temporal evolution of transformed fraction is expressed as:

$$f(t) = 1 - \exp[-x_{\mathrm{e}}(t)] \tag{3}$$

and the extended fraction x_{e} is described as:

$$x_{\mathrm{e}} = \int_0^t \dot{N}(\tau)Y(\tau,t)\mathrm{d}\tau \tag{4}$$

Recently, a modular model for transformation kinetics [15,17] has been proposed that includes, but is not restricted to, the classical JMAK description. This modular model expands the JMAK theory with time- or temperature-dependent kinetic parameters, and the model recognizes three mechanisms, nucleation, growth and impingement of growing new-phase particles, and it is applicable to both isothermal and non-isothermal transformations. A detailed description for the modes of nucleation, growth and impingement was reported in Ref. [15].

Regarding to Ref. [15], the extended fraction x_{e} for different combinations of nucleation and growth mechanisms can be expressed in the following general analytical form as:

$$x_{\mathrm{e}} = K_0^n \alpha^n \exp\left(-\frac{nQ}{RT}\right) \tag{5}$$

where α is identified with either the annealing time t for isothermal transformation or RT^2/Φ for isochronal (heating) transformation with constant heating rate Φ. In general, the kinetic parameters n (growth exponent), Q (effective activation energy) and K_0 (rate constant) are functions of time t (isothermal transformation) or temperature T (isochronal transformation) and depend on the corresponding model parameters of nucleation and growth modes. Explicit expressions for n, Q and K_0 in terms of general nucleation and growth modes for isothermal and isochronal annealing (heating) are given in Ref. [15].

In this modular model, the effect of anisotropic growth is also considered an impingement mode [15,17]. One phenomenological approach accounting for impingement in this case can be given as:

$$f = 1 - [1 + (\xi - 1)x_{\mathrm{e}}]^{-\frac{1}{\xi - 1}} \tag{6}$$

where $\xi \geq 1$. Equation (6) merely modifies the relationship between the transformed fraction f and extended fraction x_{e}, applying the phenomenological factor ξ for impingement.

3 Transformation kinetics involving anisotropic effect

3.1 Model description

For randomly oriented anisotropic particles neglecting the blocking effect, the JMAK theory still holds and the transform fraction depends only on the particle volume but not the particle orientation [18]. However, mutual interference of anisotropic particles is certainly inevitable [6−10]. Accordingly, the anisotropic growth just becomes the problem of the blocking effect (anisotropic effect).

As illustrated schematically in Fig. 1, the dashed circle indicates that an anisotropically growing particle (aggressor A) is equivalently considered an isotropic particle with invariable volume, the aggressor would encounter the successive interferences of other particles (blockers). Assume that aggressor A nucleates at $t=\tau$ and grows towards the origin at a rate dY. As the transformation proceeds, the aggressor is progressively interfered by the first blocker ($t=t_1$), the second blocker ($t=t_2$) and the Nth blocker until it arrives at the origin at time $t=t$ ($\tau<t_1<t_2\cdots<t$). Accordingly, the aggressor grows at a rate of γdY after $t=t_1$, γ^2dY after $t=t_2$, and finally, γ^NdY after $t=t_N$. Physically, dY and γ are defined as the averaged volume increment and the non-blocking factor (i.e. the unblocked part of the averaged volume increment), for a single particle, respectively.

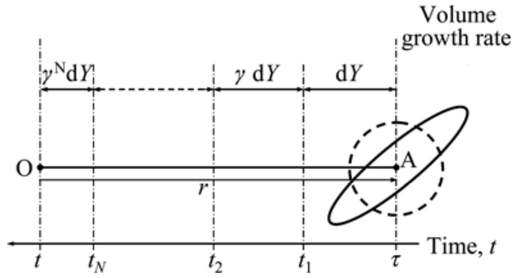

Fig. 1 Schematic diagram depicting statistical treatment of blocking effect arising from anisotropic grow

Considering anisotropic effect, only unblocked particles can grow to the origin. Therefore, a time-dependent function $S(t)$ to represent the non-blocking probability of aggressors is introduced [9]. Thus, an aggressor encountering successive interferences of blockers can still transform the origin probability as [8,16]

$$\mathrm{d}x_{\mathrm{en}} = S(t)\mathrm{d}x_{\mathrm{e}} \tag{7}$$

where $S(t)$ represents all the orientation-, time- and position-averaged value of the non-blocking probability factors.

Denote the probability function for a particle to encounter only N-scale blocking as $p_N(t)$. Particularly, $p_0(t)$ indicates the probability of the particle unblocked. And then the probability that the aggressor after undergoing the N-scale blocking without any higher-scale blocking transforms the origin at time t can be expressed as:

$$\mathrm{d}x_{\mathrm{en}} = p_0(t)\mathrm{d}x_{\mathrm{e}} + p_1(t)\gamma\mathrm{d}x_{\mathrm{e}} + p_2(t)\gamma^2\mathrm{d}x_{\mathrm{e}} + \cdots + p_N(t)\gamma^N\mathrm{d}x_{\mathrm{e}} \tag{8}$$

It should be mentioned that Eq. (8) represents the statistical contributions of different degrees of anisotropic effect to $\mathrm{d}x_{\mathrm{en}}$. This strongly implies that the anisotropic effect on the transformation depends on not only γ, but also N; the coexistence of multiple blocking prevails. Comparing Eqs. (8) and (7), one can obtain a time-dependent, averaged and non-blocking probability function $S(t)$,

$$S(t) = \sum_{i=0}^{N} \gamma^i p_i(t) \tag{9}$$

Regarding to Ref. [16], the probability function for each particle undergoing the Nth blocking until the origin is transformed at t, namely, $p_N(t)$, is concluded to obey Poisson distribution as:

$$p_N(t) = \frac{1}{N!}\left[\int_0^{x_{\mathrm{e}}} S(x'_{\mathrm{e}})\mathrm{d}x'_{\mathrm{e}}\right]^N \exp\left[-\int_0^{x_{\mathrm{e}}} S(x'_{\mathrm{e}})\mathrm{d}x'_{\mathrm{e}}\right] \tag{10}$$

3.2 Formal expression for fraction transformed

3.2.1 One-scale blocking

If an aggressor encounters only one-scale blocking without any higher-scale blocking (see Fig. 1), the probability of the aggressor unblocked $p_0(t)$ and the probability of the aggressor once blocked p_1(t) occur and follow

$$p_0(t) + p_1(t) = 1 \tag{11}$$

$p_0(t)$ can be given in Eq. (10) and $p_1(t)$ can be given as $1-p_0(t)$. According to Eq. (9), the function of non-blocking probability $S(x_{\mathrm{e}})$ can be obtained as:

$$S(x_{\mathrm{e}}) = \exp\left[-\int_0^{x_{\mathrm{e}}} S(x'_{\mathrm{e}})\mathrm{d}x'_{\mathrm{e}}\right] + \gamma\left\{1-\exp\left[-\int_0^{x_{\mathrm{e}}} S(x'_{\mathrm{e}})\mathrm{d}x'_{\mathrm{e}}\right]\right\} \tag{12}$$

Equation (12) is a differential equation for x_{en} and x_{e}, with $S(x_{\mathrm{e}})=\mathrm{d}x_{\mathrm{en}}/\mathrm{d}x_{\mathrm{e}}$ and $x_{\mathrm{en}} = \int_0^{x_{\mathrm{e}}} S(x'_{\mathrm{e}})\mathrm{d}x'_{\mathrm{e}}$, and the analytical solution follows

$$x_{\mathrm{en}} = \ln\frac{\gamma - 1 + \exp(\gamma x_{\mathrm{e}})}{\gamma} \tag{13}$$

Meanwhile, $S(x_{\mathrm{e}})$ can be obtained as:

$$S(x_{\mathrm{e}}) = \frac{\gamma\exp(\gamma x_{\mathrm{e}})}{\gamma - 1 + \exp(\gamma x_{\mathrm{e}})} \tag{14}$$

The transformed fraction f accounting for the one-scale blocking arising from the anisotropic growth can be expressed as:

$$f = 1 - \exp(-x_{\mathrm{en}}) = 1 - \frac{\gamma}{\gamma - 1 + \exp(\gamma x_{\mathrm{e}})} \tag{15}$$

3.2.2 k-scale blocking

Analogously, if an aggressor encounters the k-scale blocking without any higher-scale blocking, then k+1 events, namely, the probability of the particle unblocked $p_0(t)$ and the probabilities of the particle blocked once $p_1(t)$, twice $p_2(t)$,⋯ and k-scale $p_k(t)$ occur and follow

$$\sum_{i=0}^{k} p_i(t) = 1 \tag{16}$$

The probability of the aggressor encountering the previous (k−1) scales blocking satisfies Eq. (10), and the $p_k(t)$ can be given as $1-\sum_{i=0}^{k-1} p_i(t)$. Applying $S(x_{\mathrm{e}})=\mathrm{d}x_{\mathrm{en}}/\mathrm{d}x_{\mathrm{e}}$, Eq. (9) can be rewritten as:

$$\frac{\mathrm{d}x_{\mathrm{en}}}{\mathrm{d}x_{\mathrm{e}}} = \gamma^k + \sum_{i=0}^{k-1}\frac{1}{i!}(x_{\mathrm{en}})^i\exp(-x_{\mathrm{en}})(\gamma^i - \gamma^k) \tag{17}$$

Since Eq. (17) is a non-linear ordinary differential equation, the exact analytical solution to this equation is not feasible. Nevertheless, numerical calculations show that the transformed fraction can be still obtained through f=1−exp[$-x_{\mathrm{en}}(t)$] [16].

3.2.3 Infinite-scale blocking

According to Eq. (10), an aggressor encountering infinite-scale blocking, $p_N(t)$, satisfies

$$\sum_{0}^{\infty} p_N(t)=1 \tag{18}$$

Inserting Eq. (10) into Eq. (9) with N approaching infinity yields

$$S(x_e(t))=\sum_{N=0}^{\infty}\gamma^N p_N(t)=\sum_{N=0}^{\infty}\frac{1}{N!}\left[\gamma\int_0^t S(x_e(\tau))\mathrm{d}x_e(\tau)\right]^N\cdot \exp\left(-\int_0^t S(x_e(\tau))\mathrm{d}x_e(\tau)\right) \tag{19}$$

Eq. (19) can be rewritten as:

$$S(x_e(t))=\exp\left\{-\int_0^t (1-\gamma)S[x_e(\tau)]\mathrm{d}x_e(\tau)\right\} \tag{20}$$

Applying $\mathrm{d}x_{en}=S(x_e(t))\mathrm{d}x_e$, the differential equation Eq. (20) can be solved as:

$$x_{en}=\frac{1}{1-\gamma}\ln[1+(1-\gamma)x_e] \tag{21}$$

and

$$S(x_e)=\frac{1}{1+(1-\gamma)x_e} \tag{22}$$

At last, the transformed fraction f can be expressed as:

$$f=1-\exp(-x_{en})=1-[1+(1-\gamma)x_e]^{-\frac{1}{1-\gamma}} \tag{23}$$

Note that Eq. (23) is completely the same as the phenomenological Eq. (6) with $\xi=2-\gamma$ $(0<\gamma<1)$, which shows that the phenomenological treatment (see Eq. (6)) corresponds just to the extreme case where the particle encounters the infinite-scale blocking.

3.3 Avrami exponent and effective activation energy

Avrami exponent is commonly used as a tracer of the mechanisms underlying the transformation. However, anisotropic growth of neighboring particles leads to mutual blocking and then the retardation of transformation. Therefore, the anisotropic effect can be evaluated from the varying Avrami exponent.

Generally, for isothermal transformation, the Avrami exponent is often evaluated by plotting $\ln(-\ln(1-f))$ versus $\ln t$, whereas the effective activation energy is evaluated by plotting $\ln t$ versus $1/T$ [15,17]. Applying $f=1-\exp[-x_{en}(x_e)]$, in combination with Eq. (7), a new expression for Avrami exponent subjected to anisotropic effect, n_{new}, is deduced as:

$$n_{new}=\frac{\mathrm{d}\ln[-\ln(1-f)]}{\mathrm{d}\ln t}=\frac{\mathrm{d}\ln x_{en}}{\mathrm{d}\ln x_e}\frac{\mathrm{d}\ln x_e}{\mathrm{d}\ln t}=\frac{x_e}{x_{en}}S(x_e)n \tag{24}$$

where n corresponds to the analytical expression obtained from the modular model for transformation assuming different nucleation and isotropic growth modes (see Ref. [15]).

Analogously, a new expression for effective activation energy of transformation subjected to anisotropic effect can be obtained as:

$$Q_{new}=\frac{\mathrm{d}\ln t}{\mathrm{d}(1/T)}R=\frac{\mathrm{d}\ln[-\ln(1-f)]/\mathrm{d}(1/T)}{\mathrm{d}\ln[-\ln(1-f)]/\mathrm{d}\ln t}R=Q \tag{25}$$

It should be noted that, in Eq. (25), Q is according to the modular model expressed as $(d/mQ_G+(n-d/m)Q_N)/n$ [15,17]. Then the relationship between Q_{new} and n_{new} can be deduced as:

$$Q_{new}=\frac{d}{m}\frac{x_e}{x_{en}}S(x_e)Q_G+\left[n-\frac{d}{m}\frac{x_e}{x_{en}}S(x_e)\right]Q_N \Big/ n_{new} \tag{26}$$

where d is the growth dimensionality; m is the growth mode parameter (m=1 for interface-controlled growth and m=2 for volume diffusion-controlled growth); Q_G is the growth activation energy and Q_N is the nucleation activation energy. Equation (26) clearly shows that the anisotropic effect directly acts on the dimensionality of growth. This makes sense since the particle morphology changes after encountering interference. However, Q_G and Q_N hold unaffected, and then the overall effective activation energy is not affected by the anisotropic effect.

4 Evolution of anisotropic effect with transformation

On the basis of JMAK theory, if all the particles anisotropically grow in an infinite matrix without the influence of other particles, each exhibits the same rate distribution and shape but different orientations. However, the impingement of anisotropic particles certainly exists, which must change the rate distribution, the shape and even the particle orientation. From the statistic viewpoint, with transformation proceeding, the evolution of anisotropic effect is bound to happen.

From Section 3.3, it is necessary to introduce a parameter, μ, which can reflect the evolution of anisotropic effect as:

$$\mu=\frac{x_e}{x_{en}}S \tag{27}$$

According to Section 3.1, when $t \to 0$, $x_{en} \to x_e$ and $S \to 1$, then $\mu \to 1$; when $t \to \infty$, $x_{en} \to x_e S$, then μ also approaches 1. It means that isotropic growth has dominance at the initial stage and last stage of the transformation. In other words, at the end of transformation, all particles have a tendency to grow isotropically because the rate distributions again tend to the same and/or particles orientations tend to parallel after adequate impingement. However, at the middle stage, the dominance of isotropic growth is undermined, owing to a severe blocking effect arising from anisotropic growth. The conclusion was also found by SHEPILOV and BAIK [6] and BIRNIE III and WEINBERG III [7], but they did not provide a valid interpretation.

An example is taken to expound the analysis. Assuming mixed nucleation and three-dimensional (3D) interface-controlled growth with model parameters as $N_0=5\times10^{15}$ $m^{-3+}\cdot s^{-1}$, $N^*=1\times10^{10}$ m^{-3}, $v_0=1\times10^9$ m/s, Q_N=100 kJ/mol and Q_G=200 kJ/mol (as described in Ref. [15], N_0 is the temperature-independent nucleation rate; N^* is the number of pre-existing nuclei per unit volume; v_0 is the pre-exponential factor for growth; Q_N is the nucleation activation energy and Q_G is the growth activation energy), for isothermal transformation at T=680 K, the evolution of n_{new} with f is obtained, subjected to the same-scale blocking but different γ (e.g. only 1-scale blocking occurring but γ=0.2, 0.4, 0.6, 0.8, see Fig. 2(a)), and subjected to different- scale blocking but the same γ (e.g. γ=0.4 and k-scale blocking with k=0, 1, 2, 3, see Fig. 2(b)). It is shown clearly that the transformation accounting for the blocking effect arising from anisotropic growth depends on not only γ but also k.

From Eq. (27), x_e/x_{en} and S act as the two key factors to evaluate the anisotropic effect. Applying the same model parameters as that used for Fig. 2, the evolution of x_e/x_{en}, S and μ with f, as well as the evolution of n_{new} and n with f, subjected to γ=0.4 and 1-scale blocking, is calculated and shown in Fig. 3. Clearly, x_e/x_{en} increases monotonously with f, but S declines with f. Consequently, μ declines first, reaches a minimum value, and then increases. This implies that three basic regimes of transformation could be identified. At the initial stage of transformation, x_e/x_{en} and S both approach to unity, then $\mu \to 1$; the equivalent isotropic particles each grow independently without the blocking effect. At the middle stage of transformation, severe blocking effect occurs; μ does have a minimum value at some time. At the final stage of transformation, however, the blocking effect is alleviated, so that μ tends to the value where the blocking has not occurred, i.e. $\mu \to 1$ as seen in Fig. 3.

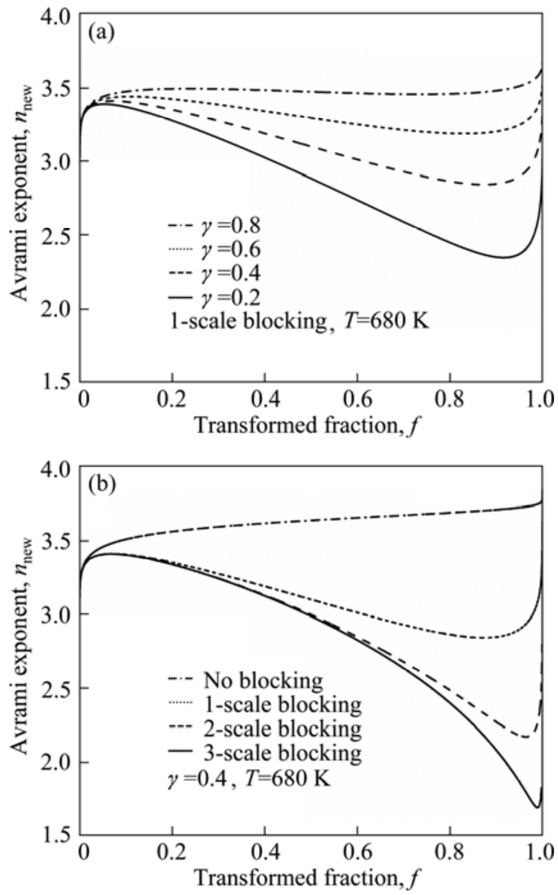

Fig. 2 Evolution of n_{new} with f for transformation assuming mixed nucleation and 3D interface controlled growth: (a) Same-scale blocking but different γ; (b) Different-scale blocking but same γ

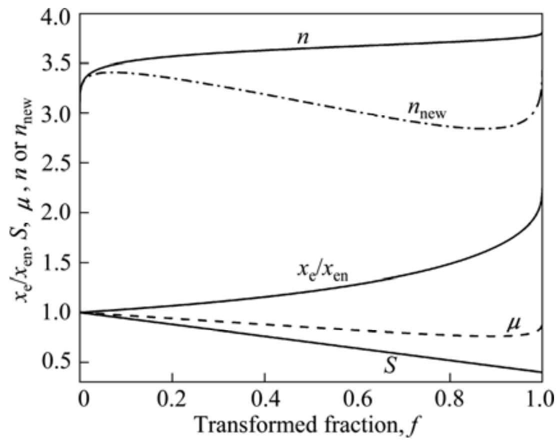

Fig. 3 Evolution of x_e/x_{en}, S and μ with f, as well as evolution of n and n_{new} with f, for transformation subjected to 1-scale blocking and γ=0.4 (Values of model parameters are same as those in Fig. 2)

5 Conclusions

1) The effect of anisotropic growth on the kinetics

of solid-state transformation is described. Anisotropic effect on the transformation depends on not only the non-blocking factor γ but also the blocking scale k.

2) Blocking effect arising from anisotropic growth leads to a reduction of Avrami exponent, but the unblocked value in the end; whereas effective activation energy is not affected by the anisotropic effect.

3) Anisotropic effect directly acts on the dimensionality of growth.

4) Evolution of anisotropic effect with the fraction transformed is taken into account, which can be reflected by the behavior of μ, and the anisotropic effect is more severe at the middle stage of transformation and is alleviated at both initial and last stage.

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颗粒各向异性生长对固态相变动力学的影响

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摘　要：基于对颗粒各向异性生长引起阻碍效应的几何概率分析，研究生长各向异性效应对固态相变动力学的影响。结果表明：阻碍效应导致生长速率降低，进而导致 Avrami 指数的规律变化。根据先前的解析模型，推导动力学参数 Avrami 指数和总有效激活能的解析表达。各向异性效应对相变的影响不仅取决于非阻碍因子γ，还取决于阻碍级数 k。各向异性效应直接作用在颗粒生长维度上，却不影响转变过程的总有效激活能。考虑到各向异性生长所引起的阻碍效应与转变分数的演化，发现各向异性效应在相变过程中期最为剧烈。

关键词：转变动力学；各向异性生长；阻碍效应；Avrami 指数

(Edited by FANG Jing-hua)