ISSN:2790-1688 DOI: 10.56028/aetr.4.1.321.2023

Phase Field Simulation of the effect of Aging Precipitates on Grain Growth in Aluminum Alloys

Xin Song*

CSSC intelligent Technology Shanghai CO., LTD, Shanghai 201210, China

*Corresponding author e-mail: 653104900@qq.com

Abstract. Aluminum alloy skin for automobile is the focus of development, application and research of aluminum alloy for automobile. At present, the problems such as aging stability, formability, concavity resistance and bake hardening of anti-baking paint need to be solved urgently. The aging precipitates of baking varnish have the effect of pinning grain boundaries and refining grains. In this study, the topological transformation law of polycrystalline evolution during normal grain growth was analyzed by continuous phase field model, and the influence of different size, morphology and volume fraction of precipitated phase particles on polycrystalline growth were simulated. By verifying the Zener relationship between the particle size of the precipitate and the limit radius of the grain, an exponential function relationship different from the growth law of the ideal spherical particle pinned polycrystals was obtained. This research has an important scientific guiding role in the aging modification design of automobile skin and in improving the service life of aluminum alloy sheets.

Keywords: aluminum alloy; paint aging; precipitated phase particles; grain growth; Zener relationship.

1. Introduction

As the main material of automobile skin, the aging process of aluminum alloy paint has always been the focus and difficulty in the development and application of automobile aluminum alloy. The automobile skin generates precipitated phase particles with different morphologies, sizes and volume fractions during paint aging. These particles embedded in the aluminum matrix form a non-coherent interface with the aluminum matrix in the late aging period. The interaction between the "pinned" particles and the aluminum matrix will significantly affect the evolution of its microstructure, especially the migration of grain boundaries and the movement of dislocations during deformation [1,2,3,4]. It has become an effective method to design high strength aluminum alloy by controlling the grain growth to improve the mechanical properties of the alloy. According to the Hall-Petch polycrystalline strength theory [5], the smaller the grain size within a certain range, the higher the alloy strength. Grain refining has become an important consideration in designing high-strength aluminum alloys. Therefore, it is necessary to study the role of precipitates in grain boundary movement and grain growth, which is helpful to analyze the limit grain size of alloys and obtain quantitative prediction of alloy properties.

Traditional material calculation methods, such as cellular automata [6] and Monte Carlo [7], usually describe the material interface as a numerical interface without thickness. The sharp characteristics of the numerical interface make it necessary to track the interface position in real time during simulation [8]. For phase transformation problems with complex interface structures, such as dendrite growth during solidification [9], it is difficult to adopt the classical sharp interface model. As shown in Fig. 1a, in the diffusion interface field model, the value of the phase field variable undergoes a continuous change process in the interface, at which time the interface has a certain thickness. In the sharp interface model (Figure 1b), the field variable has a sudden change in value at the interface. At this time, the interface has no thickness, and the composition and structure of the solid phase on both sides of the Zero thickness interface will have a sudden change.

ISSN:2790-1688 DOI: 10.56028/aetr.4.1.321.2023

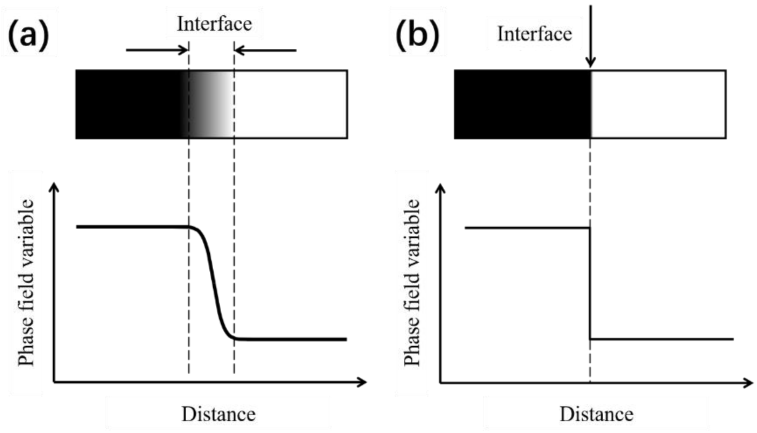


Figure 1. Schematic diagram of phase field variables of different types of interfaces:
(a) diffusion interface
(b) Sharp interface

In this study, the continuous phase field model will be used to simulate the process of normal grain growth and the inhibition of grain growth by "pinning" grain boundaries of second phase particles. Through the phase field simulation of "pinning" grain boundaries of second phase particles with different volume fraction, size and spatial orientation, the influence of different types of second phase particles on grain boundary migration will be studied.

2. Theoretical models and methods

2.1 Phase field model of grain growth

In order to describe grains with different orientations in polycrystalline materials, the diffusion interface field model uses a series of orientation field variables η_i as the order parameter of the phase field model [10,11]:

$$\eta_1(r,t),\eta_2(r,t),\eta_3(r,t),\cdots,\eta_i(r,t)\cdots,\eta_p(r,t) \tag{1}$$

Where $i = 1,\cdots,p$. $\eta_i(r,t)$ is the non-conservative phase field order parameter， and p is the number of possible orientations in the polycrystalline material. For polycrystalline systems considering only grain growth, the characteristic order parameter η_i and the order parameter gradient $\nabla\eta_i$ to construct the total free energy functional of the system [12] F_{total}:

$$F_{total} = \int f\left(\eta_1,\eta_2,\cdots,\eta_i,\cdots\eta_p,\nabla\eta_1,\nabla\eta_2,\cdots,\nabla\eta_i,\cdots,\nabla\eta_p\right) dV \tag{2}$$

In equation (2), f_0 is the local free energy density function; k is the interface gradient energy coefficient. In the interface isotropic system, k is a constant, which is related to the isotropic interface energy parameter; In the system of interfacial anisotropy, k is a function of the orientation difference angle of adjacent grains [13]. The change process of the non-conservative order parameter in the phase field model in time and space must meet the Ginzburg Landau relaxation equation:

$$\frac{\partial\eta_i(r,t)}{\partial t} =- L\frac{\delta F_{total}}{\delta\eta_i(r,t)}, i = 1\cdots\cdots p \tag{3}$$

Where L is the interface dynamics parameter (related to grain boundary mobility).

ISSN:2790-1688 DOI: 10.56028/aetr.4.1.321.2023

2.2 Phase field model of grain growth coupled with "pinning" of the second phase

The following expression can be introduced into the free energy function of the phase field model to study the pinning effect of the second phase particles on the grain boundary [14]:

$$\varepsilon_{sp}\lambda\left(\sum_{i=1}^{sp}\eta_i^2\right) \tag{4}$$

ε_{sp} the characterization coefficient of the second phase; λ is a function related to spatial position. In combination with Ginzburg Landau dynamic equation and the free energy function of coupling the second phase, the dynamic evolution equations of particle pinning with the second phase are derived:

$$\frac{\partial\eta_i(r,t)}{\partial t} = -L\left(\left(-\eta_i+\eta_i^3+2\gamma\eta_i\sum_{j\neq i}^{p}\eta_j^2+2\varepsilon_{sp}\lambda\eta_i\right)-k\nabla^2\eta_i\right) \tag{5}$$

3. Simulation results and analysis

3.1 Normal grain growth process

In this simulation, 512 Δx×512 Δx square grids are selected, and the initial value of the orientation field variable η is a random number between -0.001 and 0.001. Figure 2 shows the schematic diagram of the topological transformation of grains and the corresponding phase field simulation results. G5 grain is a quadrilateral grain, while G1, G2, G3 and G4 are all grains with more than 6 grain edges. According to Mullins theory, the area of G5 grain will gradually decrease to disappear in the evolution process, which is also verified by the results of phase field simulation in Figure 2b.

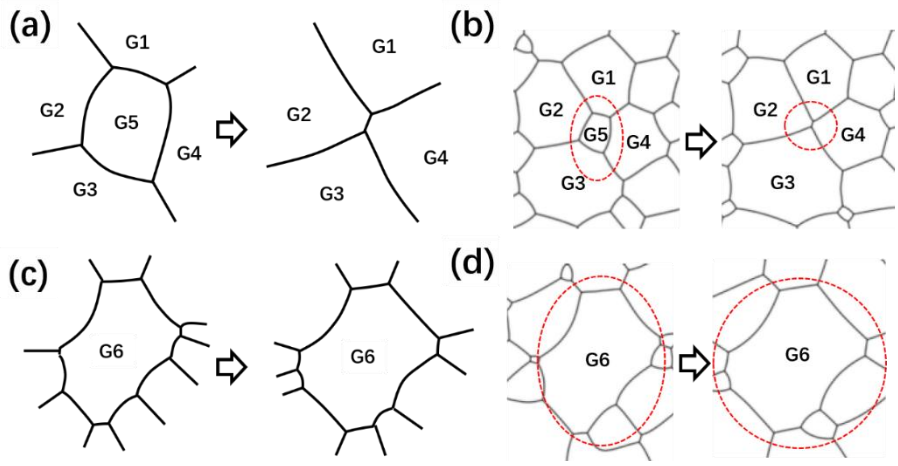


Figure 2. Topological transformation of several grain boundaries.
(a, c) is the crystal topology diagram, and (b, d) is the phase field simulation result

Figure 2 shows the topological transformation of grain boundary before and after the evolution of grain G6. It is obvious that the grain growth process simulated by phase field is consistent with the (6-f) principle of Mullins [15] grain edge number and the stability conditions that the grain boundary needs to meet.

ISSN:2790-1688 DOI: 10.56028/aetr.4.1.321.2023

3.2 Phase field simulation of grain boundary pinned by second phase particles

3.2.1 Effect of volume fraction of second phase particles on polycrystalline growth

In this phase field simulation, five groups of second phase particles with different volume fractions（$f_a = 0.005$，$f_b = 0.015$，$f_c = 0.03$，$f_d = 0.06$，$f_e = 0.09$）are randomly distributed in the polycrystalline simulation area in Fig. 3b as the initial samples of grain growth.

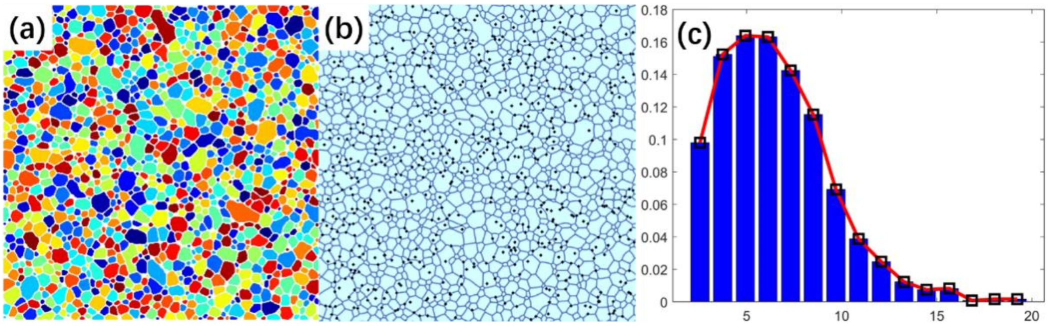


Figure 3. Initial Grain Structure of Random Distribution of Second Phase Particles

In the process of normal grain growth, the average area of grains has a linear relationship with the evolution time, which is consistent with the stability condition of Lifshitz Lyozov growth discussed previously. However, this stability relationship is no longer applicable to the process of pinned polycrystalline growth of the second phase particles, and the trend of average grain growth is weakened by the second phase particles (Figure 4).

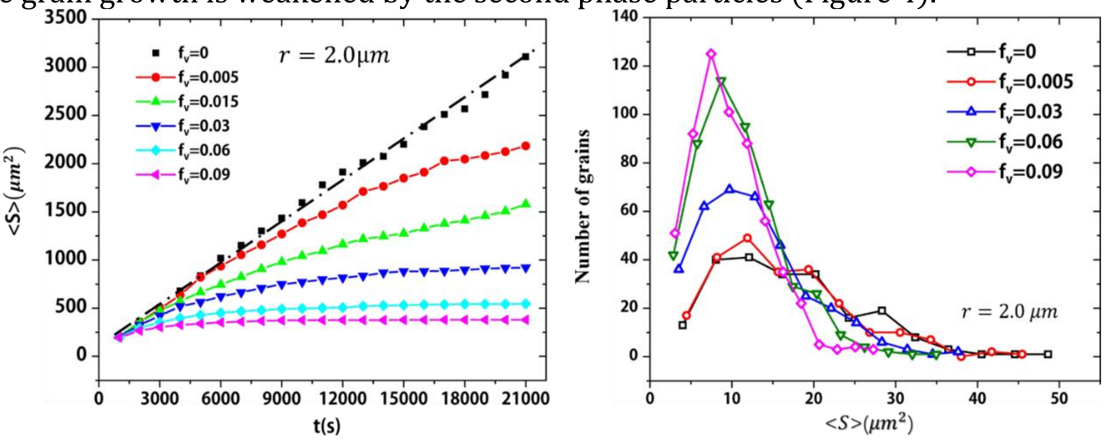


Figure 4. (a) Effect of second phase particles with different volume fractions on average grain area during polycrystalline evolution;
(b) Size frequency distribution curve of grains after pinning of second phase particles with different volume fractions

3.2.2 Effect of particle size of second phase on polycrystalline growth

Figure 5 shows the microstructure morphology of the influence of second phase particles with different sizes on the growth of polycrystalline grains at a certain time step. The volume fraction of second phase particles in the three groups of phase field simulations is $f_v = 0.05$, the particle radius of the second phase is $r = 3.0$um，5.0um，8.0um. From the three groups of simulation results, it can be concluded that the ability of grain boundary "de pinning" is closely related to the size of the second phase pinning particles. The pinning particles are large in size, and almost no "de pinning" phenomenon occurs.

ISSN:2790-1688 DOI: 10.56028/aetr.4.1.321.2023

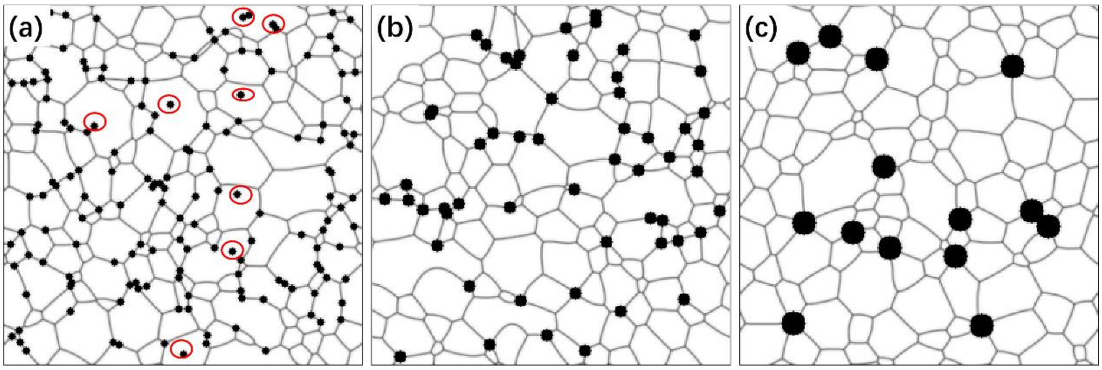


Figure 5. Effect of second phase particles with different sizes on grain growth (t=15000). Three groups of second phase particles with different sizes are $r = 3\mu m$, $r = 5\mu m$, $r = 8\mu m$

3.2.3 Effect of the morphology of second phase particles on polycrystalline growth

The pinned particles in polycrystalline materials are often not limited to spherical particles. At present, for aluminum alloys of different grades, the morphology of the second phase precipitated by aging is different, mainly in the form of lath（GPII Zone， θ'phase）、needle like（β'', β'）、horseshoe, shuttle and sphere.

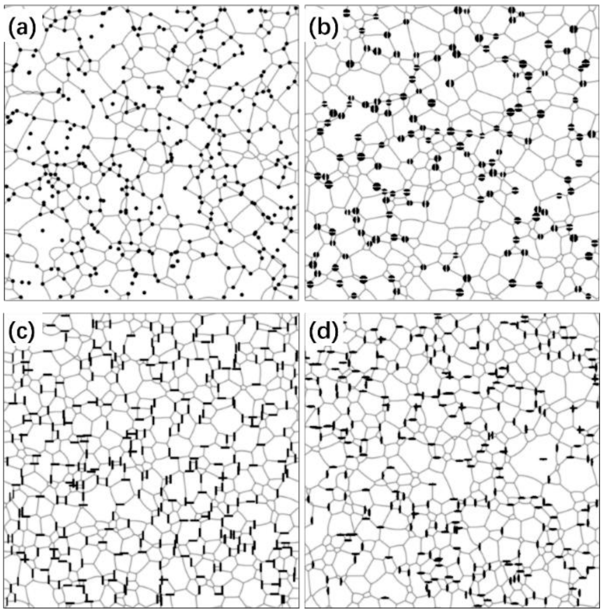


Figure 6. Effect of second phase particles with different morphologies (spherical, horseshoe shaped, strip shaped, shuttle shaped) on the growth of polycrystalline particles (t=10000).

The volume fraction of second phase particles is 5%

ISSN:2790-1688
DOI: 10.56028/aetr.4.1.321.2023

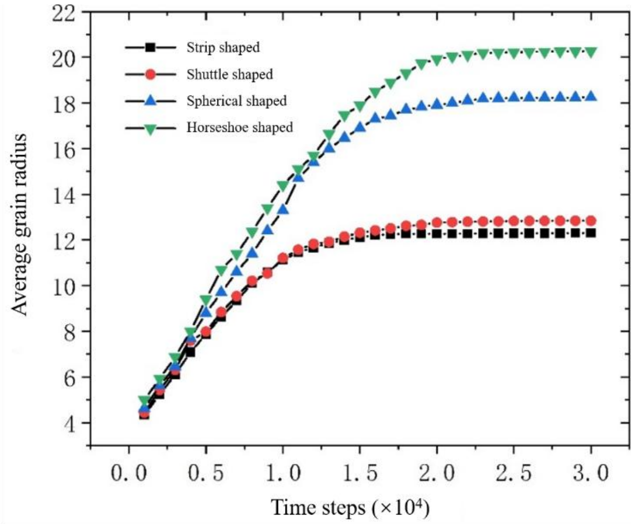


Figure 7. Size frequency distribution curve of second phase particles (spherical, horseshoe, strip, shuttle) with different morphologies after pinning

3.2.4 Verification of the relationship between the second phase particle size and the grain limit radius by Zener equation

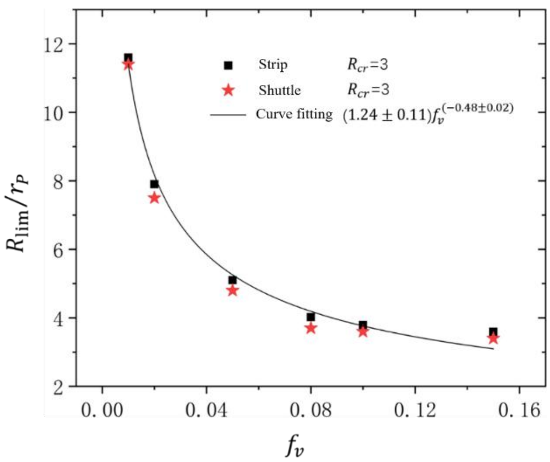
Different volume fractions of second phase particles will form polycrystalline structures with different limit grain sizes. Zener [16] proposed the relationship between the volume fraction f_a of the second phase、the limiting grain radius R_{lim} of polycrystalline materials and the average radius r_{sp} of the second phase particles through the pinning effect of spherical particles on the grain boundary:

$$\frac{R_{lim}}{r_{sp}} = \frac{4}{3} f_a^{-1} \quad (6)$$

More and more experimental results show that there is a simple exponential relationship between the volume fraction of the second phase, the limit grain radius and the average grain radius

$$\frac{R_{lim}}{r_{sp}} = w f_a^{-k} \quad (7)$$

Based on the phase field simulation results obtained in this study, Figure 8 shows the relationship curve between the ratio of the grain limit radius R_{lim} and the equivalent radius r_P of the second phase particles and the volume fraction f_v of the second phase particles.



ISSN:2790-1688 DOI: 10.56028/aetr.4.1.321.2023

Figure 8. Relation curve between the ratio of grain limit radius R_{lim} and the equivalent radius r_P of the second phase particles and the volume fraction f_v of the second phase particles

4. Conclusion

Based on the diffusion interface field theory, the order parameter describing the grain growth and the corresponding phase field model are described in detail, and the relevant terms coupling the second phase particles are introduced into the dynamic equation of the model, and the phase field model with particle pinning effect is derived. Then, through the discussion and optimization of the model parameters, a dynamic equation that can reasonably describe the grain growth is obtained. Through the simulation of grain growth, the topological structure transformation of grain growth is analyzed, and the effects of different volume fractions, different sizes and different shapes of second phase particles on grain growth are studied. The following conclusions are obtained:

1.The phase field simulation of grain growth verified the（$f - 6$）theory of Neumann Mullins grain topological transformation, that is, when the grain edge number f is greater than 6, the grain will grow up, otherwise, the grain will slowly disappear. At the same time, the simulation process of two-dimensional dynamic grain growth also follows the Lifshitz-Lyozov stability condition, that is, the average size of polycrystalline grains $R^2 \sim t$。

2.By discussing the volume fraction, size and morphology of the second phase particles, the basic rule of the second phase particle pinning on the polycrystalline growth was obtained: With the increase of the volume fraction of the second phase, the number of the second phase particles increases, leading to the decrease of the grain limit size. The smaller pinned particles are usually distributed in the whole grain boundary, while the larger pinned particles are usually located at the junction of trigeminal crystals. The more pinned particles with small size, the more obvious the overall grain boundary migration is. However, for a single pinned particle, its pinning effect is reduced, and the phenomenon of "off pinning" often occurs, which will cause abnormal grain growth. The strip and shuttle shaped pinning particles with sharper morphology have more obvious pinning effect, while the spherical and horseshoe shaped particles with more smooth and gentle shape have less ability to pin the grain boundary.

3.By verifying the Zener relationship between the second phase pinning particle size and the limit radius of the grain, an exponential relationship different from the ideal spherical pinning was obtained. In this chapter, it is simulated that the relationship between the equivalent size of the second phase and the grain limit radius is $(1.24 \pm 0.11)f_v^{(-0.48 \pm 0.02)}$.

Acknowledgments

This work was financially supported by Excellent Youth Fund Project of Hunan Provincial Education Department (22B0684).

References

[1] Orowan Egon. Fracture and strength of solids [J]. Reports on Progress In Physics, 1949, 12(1): 185.

[2] Kim Seong Gyoon, Park YB. Grain boundary segregation, solute drag and abnormal grain growth [J]. Acta Materialia, 2008.

[3] Kirchheim Reiner. Grain coarsening inhibited by solute segregation [J]. Acta Materialia, 2002, 50(2): 413-419.

[4] Cahn John W. The impurity-drag effect in grain boundary motion [J]. Acta Metallurgica, 1962, 10(9): 789-798.

ISSN:2790-1688 DOI: 10.56028/aetr.4.1.321.2023
[5] Hall E O. The deformation and ageing of mild steel: III discussion of results [J]. Proceedings of the Physical Society. Section B, 1951, 64(9): 747.

[6] Chopard Bastien， MichelDroz， Chopard. Cellular Automata Simulation of Physical Systems [M] Tsinghua University Press, 2003

[7] Xu Zhongji Monte Carlo method [M] Shanghai Science and Technology Press, 1985

[8] Wheeler Adam A, Murray Bruce T, Schaefer Robert J. Computation of dendrites using a phase field model [J]. Physica D: Nonlinear Phenomena, 1993, 66(1-2): 243-262.

[9] Warren James A, Boettinger William J. Prediction of dendritic growth and microsegregation patterns in a binary alloy using the phase-field method [J]. Acta Metallurgica Et Materialia, 1995, 43(2): 689-703.

[10] Chen Long Qing, Yang Wei. Computer simulation of the domain dynamics of a quenched system with a large number of nonconserved order parameters: The grain-growth kinetics [J]. Physical Review B, 1994, 50(21): 15752.

[11] Cai Yun, Sun Chaoyang, Wan Li, Yang Daijun, Zhou Qingjun, Su Zexing Study on Dynamic Recrystallization Softening Behavior of AZ80 Magnesium Alloy [J] Journal of Metals, 2016, 52 (9): 1123-1132

[12] Fan Danan, Chen Long Qing. Diffusion-controlled grain growth in two-phase solids [J]. Acta Materialia, 1997, 45(8): 3297-3310.

[13] Zaeem M Asle, El Kadiri Haitham, Wang Paul T. Investigating the effects of grain boundary energy anisotropy and second-phase particles on grain growth using a phase-field model [J]. Computational Materials Science, 2011, 50(8): 2488-2492.

[14] Moelans Nele, Blanpain Bart, Wollants Patrick. A phase field model for the simulation of grain growth in materials containing finely dispersed incoherent second-phase particles [J]. Acta Materialia, 2005, 53(6): 1771-1781.

[15] Mullins William W. Two-dimensional motion of idealized grain boundaries [J]. Journal of Applied Physics, 1956, 27(8): 900-904.

[16] Zener Clarence. Elasticity and anelasticity of metals [M]. University of Chicago press, 1948.