Influence of Random Nucleation Condition on Transformation Kinetics in Phase Field Simulations

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Abstract

Influence of nucleation condition in phase field simulation is systematically investigated. Two-dimensional multi-phase-field model for poly-crystalline material was used, and the transformation kinetics was compared with conventional Johnson-Mehl-Avrami-Kolmogorov (JMAK) model. At first, the nucleation was set as the initial condition, and the nuclei arrangement was varied as on regular lattice points or randomly distributed. In such a model, the kinetics did not correspond well to JMAK plot. Time-dependent nucleation was then considered, and it revealed that the kinetic curve agrees well to JMAK plot. Finally, limitation was imposed on the setting of nucleation sites. As a result, it revealed that restriction in the nucleation site interfere the free growth and that the kinetics deviate from the ideal one. It is concluded that proper time-dependent condition with nucleation site set make better correspondence in the transformation kinetics with the JMAK plots.

Keywords: Phase transformation, Phase field model, Microstructure, JMAK model, Nucleation, Uncertainty, Computer simulation

Introduction

Phase field (PF) model has become a useful tool for simulating microstructure formation process of engineering materials, and various complex patterns, such as dendrite, cells, lamella, and poly-crystals, have been regenerated successfully [e.g. Provatas and Elder (2010)]. The model has a basis on the thermodynamics, and free-energy minimization is ensured for growing process of the precipitated phase. Nucleation of the new phase is, however, out of the framework of the PF model, and nuclei are disposed as a computational condition. In solidification process, the melt is usually homogeneous and specific site-dependency in the melt is not found except the wall and surfaces. Randomness is then unavoidably introduced such that the nucleation site is scattered in the model using random numbers, but it brings uncertainty in the obtained results. For re-crystallization during hot-work process, recently, site-dependent nucleation is modeled based on the finite-element analysis using crystal-plastic theorem and sophisticated methods have been in great progress [e.g. Takaki et al.(2009], but the random nucleation model is still a major tool for PF simulations in general. On the contrary, macroscopic kinetics of phase transformation has long been studied. Johnson-Mehl, Avrami and Kolmogorov independently derived kinetic equation, currently known as JMAK equation, in which time evolution of the volume fraction is described using exponential term with empirical parameters. This equation is applied for various processes such as solidification, in-solid phase transformation and re-crystallization processes, and has been built in finite-element codes for engineering use. Phase field simulation exhibits microstructure in detail, while the total or averaged evolution of transformation area is not necessarily accorded with the kinetics [Jou and Lusk (1997), Li et al. (2007), Simmons et al. (2004), Alekseechkin (2011), Uehara (2014)] which is an outstanding problem for bridging the scale. Therefore, in this study, the comparison of the kinetics between PF simulation and JMAK algorithm is systematically demonstrated. A simple two-dimensional model is used to pick out the dominative factors.

Phase Transformation Kinetics

The conventional transformation kinetics known as JMAK form is summarized in this section. Assuming solidification process, solid region is initiated with nucleation, and it grows concentrically at a constant rate G. Then a spherical region of the radius $R = G\,t$ is formed at time t, and the volume is $V = 4\pi R^3/3$. When the frequency of the nucleation is N per unit time, the number of nuclei generated in the duration between t and $t + \mathrm{d}\tau$ is $N\,\mathrm{d}\tau$, and the resulting volume of solid region is $V = 4\pi G^3(t-\tau)^3/3$. Important notice is that every sphere is assumed to continue growing despite they collide to each other in reality. The volume calculated based on this assumption is termed extended volume V_e, which is represented by

$$V_\mathrm{e} = \int_0^t \frac{4}{3}\pi N G^3 (t-\tau)^3 \mathrm{d}\tau = \pi N G^3 t^4 , \tag{1}$$

where the second equality holds when both G and N are constant. Now, denoting the volume fraction of solid at time t by $x(t)$, the liquid fraction is $1-x(t)$, and the solidification actually occurs in this region. Then the increment of the solid fraction is $\mathrm{d}x = (1-x(t))\,\mathrm{d}x_\mathrm{e}$, where x_e is the extended volume fraction. Substituting Eq. (1) into this relation,

$$x = 1-\exp(-At^4), \quad \text{where} \quad A = \frac{\pi}{3} N G^3 \tag{2}$$

is obtained. Generally, this formula is expanded as $x = 1-\exp(-At^n)$, where exponent n is a parameter or named Avrami number, which is one of the fitting parameters. The theoretical value in the two-dimensional model is $n = 3$, which is used as a reference in this paper.

Fundamental Equation for Phase Field Simulation

A poly-crystalline microstructure formation is considered in this study, and multi-phase-field model originally proposed by Steinbach et al. [Steinbach et al. (1996)] is used. Only the fundamental form is described here:

$$\dot{\phi}_i = -\frac{2}{n}\sum_{j=1}^{n} m_{ij}\left(f_{ij}\sqrt{\phi_i\phi_j} + \Sigma_k (w_{ik}-w_{jk})\phi_k + \Sigma_k (a_{ik}^2 - a_{jk}^2)\nabla^2\phi_k \right) , \tag{3}$$

where ϕ_i is the multi-phase field which is assigned for every grain of precipitated phase for $i = 1$ to N while $i = 0$ is assigned for the original phase, m_{ij}, f_{ij}, w_{ij} and a_{ij} are the parameters and n is the number of existing phase. The values of these parameters, of course, affect the transformation kinetics and should be discussed in detail. However, in this study, to focus on the fundamental characteristics of the pattern formation, constant values for every combination i and j are assumed, and standard values are used. Instead, the total number N of precipitated phase considered is the focused parameter in this study. In the poly-crystalline model, the number of grains N_g is consistent with the number of multi-phase-field variable, i.e. $N_\mathrm{g} = N$, to identify each grain as a different phase, but it is time-consuming since the number of combination increases. In this paper, N is taken as 16 even for models with N_g larger than 16, since significant difference was not found in the preliminary calculation.

Model and Conditions

Phase field equation (3) is numerically solved using finite difference method. Two-dimensional square domain is divided by 400x400 lattice points, and periodic boundary condition is imposed on every directions. The whole domain is initially homogeneous original phase. Various nucleation conditions are applied, and three cases reported in this paper are listed in Table 1. Commonly the nucleus position $\boldsymbol{r}_i = (x_i, y_i)$ is selected using random number and the phase field value of ϕ_i is changed to 1, if it is still 0. If the position is already in precipitated phase, the nucleation procedure

is skipped. Firstly, in Case 1, nuclei are set as the initial condition; all nuclei are initially disposed on random position, while regular arrangement is also tested for comparison. Time-dependent nucleation is assumed in Case 2; n_c nuclei are generated every t_c time steps. Finally in Case 3, the nucleation site is limited on specific position; the case for square lattice is demonstrated in this paper. Volume fraction, or actually area fraction, is calculated by $x(t) = N_p / N_T$, where N_p is the number of grids where the phase field value ϕ_0 representing the original phase is 0 and N_T is the total number of grids (N_T=160000). Time-evolution curve of the volume fraction is compared with JMAK plot according to equation (4). The exponent is set as $n = 3$ as the two-dimensional ideal model, and the parameter A is determined so that the time for $x = 0.5$ coincides; i.e. $A = -\ln(1-x)/t^3 = -\ln(0.5)/t_h^3$, where t_h is the time when x reaches 0.5 in the PF simulation.

Table 1. Simulation condition

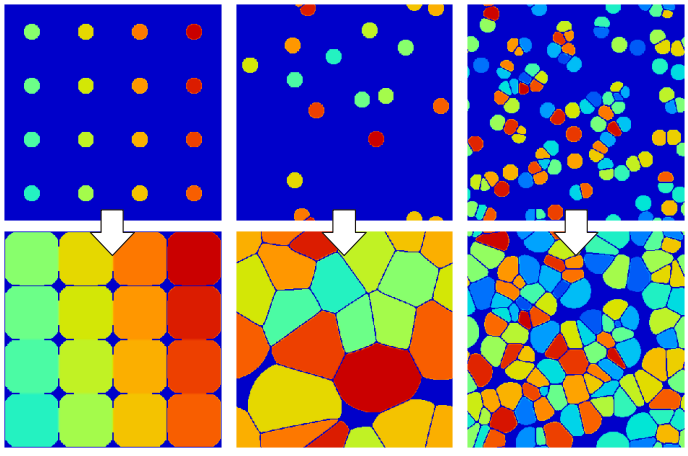
Case No.	Arrange	Timing
1	Regular / Random	Initial set
2	Random	1 nucleus per 10 steps / 1 nucleus per 2 steps / 2 nuclei every step
3	R2 lattice / R3 lattice	Initial set / 1 nucleus per 10 steps / 1 nucleus per 2 steps

Results and Discussion

Case 1 --- Nucleation as Initial Condition

Results for Case 1 are shown in Figs 1 and 2. Figure 1 shows the phase-field distributions at the early stage which represents the initial arrangement of nuclei, and the resultant polycrystalline structure just before the phase transformation completes in the whole model. The cases for total number of nuclei n = 16 and 100 are presented. The color indicates the identifying number of the multi-phase-field variable, where ϕ_0, i.e. the original phase, is shown in blue. As a matter of course, the growing domains collide to each other, and grain boundaries are formed; regularity of the obtained structure is dependent on the initial nuclei arrangement.

Figures 2 (a) and (b) represent variation of the volume fraction for regular and random arrangement, respectively. In addition to n = 16 and 100, the cases for n= 36 and 64 are also plotted. Since the results for random arrangement are dependent on the random number generated on the computer, two trials for every condition are plotted. Fig. 2 (c) is comparison between the PF result and JMAK model which is fitted according to the above-mentioned procedure, where the average values of two trials for random case are exhibited. Overall, faster growth is observed for regular arrangement, because the distance between the nuclei is uniformly large, and hence free growth duration lasts long. Compared to JMAK plots, PF result



(a) n=16: regular (b) n=16: random (c) n=100: random

Figure 1. Variation of the phase-field distribution for Case 1.

shows faster growth in the case of regular arrangement for both n=16 and 100. For the random arrangement, on the contrary, PF result shows remarkable delay from JMAK plot, and the delay is significant in the latter stage. This tendency is more apparent in Fig. 2(d); all plots are re-drawn against normalized time t^*, in which time is divided by the time when the volume fraction reached 0.5. Note that JMAK plot is identical independent of the fitting parameter A.

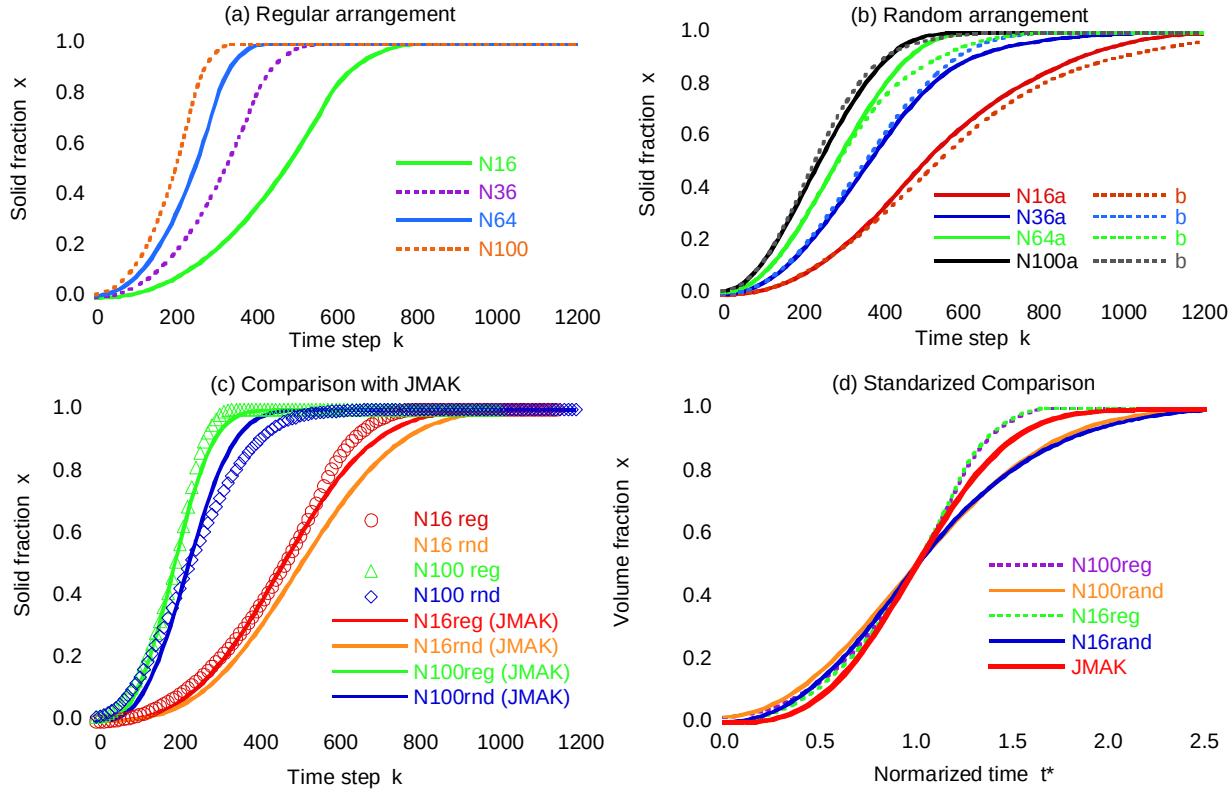


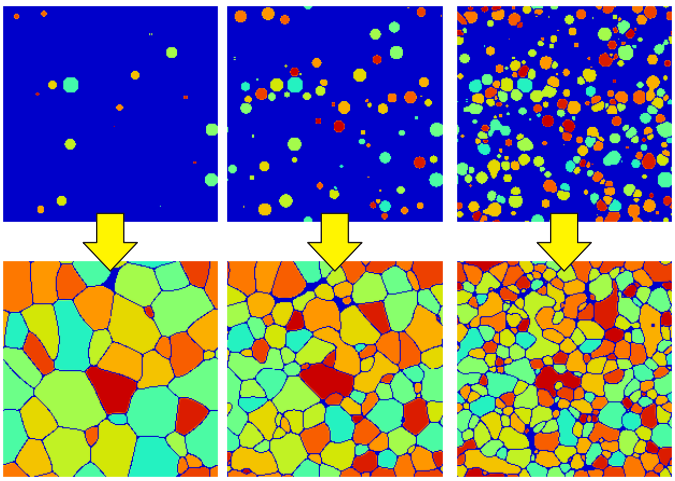
Figure 2. Variation of the volume fraction for Case 1.

Case 2 --- Time-dependent Nucleation

Results for time-dependent nucleation condition in Case 2 are shown in Figs 3 and 4. Here, three conditions are considered: a) one nucleus is generated every 10 steps, (b) one nucleus per 2 steps, and (c) 2 nuclei are generated every time step. Upper figures in Fig. 3 are the phase-field distributions at the 200th time step, and lower figures are those at the time step just before the transformation completes. Naturally, fine grains are formed when nucleation is more frequent, and coarse structure is formed when the nucleation rate is slower. Despite of this obvious difference in the microstructure, the kinetic curves correspond well to each other and also fit the JMAK plot. As shown in the magnified view in Fig. 4, a little discrepancy is found, but they are thought to be in the negligible range. Therefore, it is concluded that the time-dependency of the nucleation plays dominant role for transformation kinetics.

Case 3 --- Restriction in Nucleation Site

Finally, the effect of site-dependent nucleation is investigated. As shown in the result for Case 1, initial nuclei arrangement influences the transformation kinetics. In this section, the time-dependency is also considered. Nucleation site is limited on the square lattice, while the actual



(a) 1 per 10 steps (b) 1 per 2 steps (c) 2 per 1 step

Figure 3. Distribution of phase field for Case2.

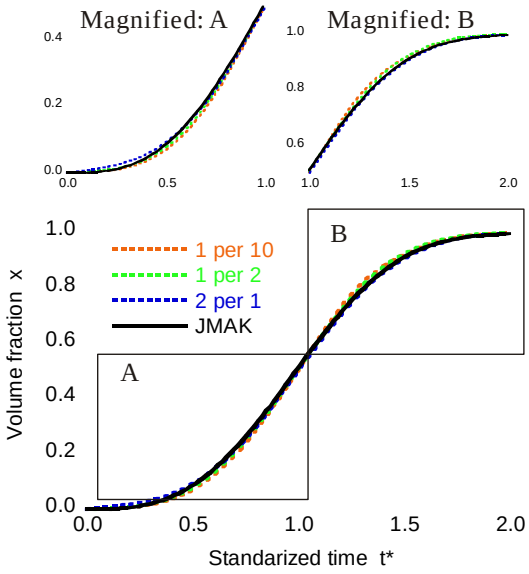
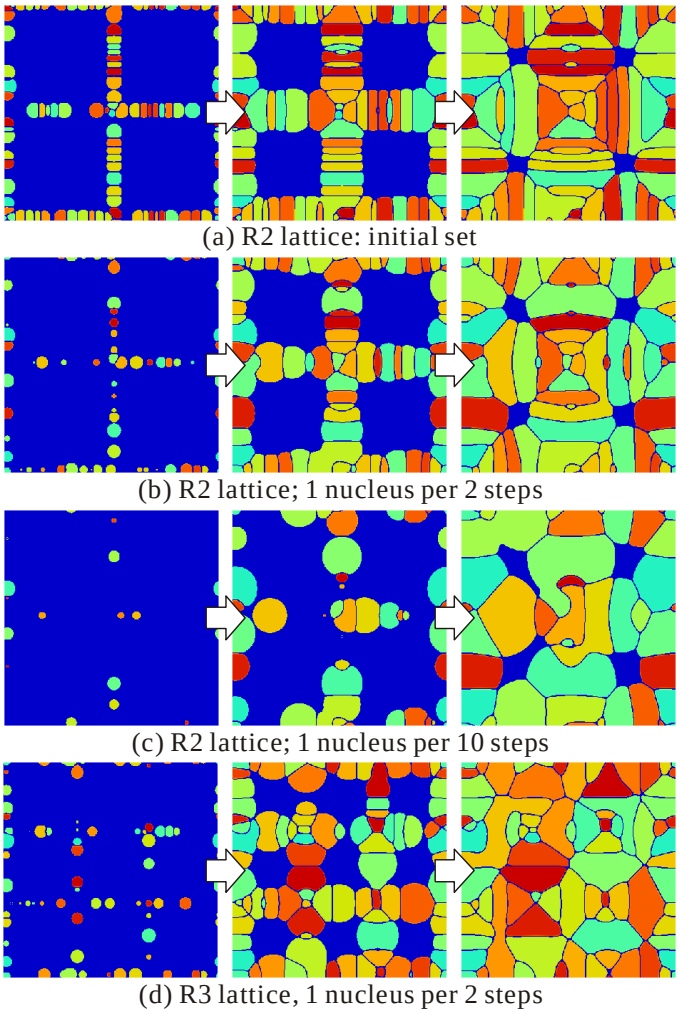


Figure 4. Time evolution for Case 2.

position on the lattice is set at random. Two types of lattice (2x2 (denoted as R2) and 3x3 (R3)) are used. The nucleation rates are assumed as same as those in the previous section, while the case for nucleation at the initial condition is also used for comparison.

Simulation results are shown in Figs 5 and 6. In Fig. 5, variation of the phase field represents the grain growth from the nucleation sites on the R2 or R3 lattice. Before two grains on the lattice collide on the lattice, they grow freely, but after the collision, the growth direction is limited inside the lattice. This causes one-dimensional grain growth, and hence deviation from JMAK plot becomes significant, as shown in the kinetic curve in Fig. 6. Especially when many nuclei are set as the initial condition, (see "R2 init"), there is no duration of free growth, and the resultant kinetics appears to be rather linear. As the nucleation rate becomes slower, the kinetics becomes closer to the ideal curve, which is apparent in Fig. 6(b). Also, the case for R3 lattice shows better correspondence to JMAK. This is because relative duration of free growth to the entire transformation is larger for R3. In other words, as the nucleation sites are strictly limited relative to the whole



(a) R2 lattice: initial set
(b) R2 lattice; 1 nucleus per 2 steps
(c) R2 lattice; 1 nucleus per 10 steps
(d) R3 lattice, 1 nucleus per 2 steps

Figure 5. Distribution of phase field for Case 3.

domain, the kinetics deviates from the ideal kinetics.

Conclusions

Nucleation condition is determinative for microstructure formation, and the influence in the phase field simulation was systematically investigated. When the nuclei are set as the initial condition, the kinetics, i.e. the evolution of volume fraction of the transformed region, does not well corresponds to JMAK plot. When time-dependent nucleation condition was introduced, the kinetic curve revealed to agree well to JMAK curve. Restriction on the nucleation site was also determinative for deviation from ideal state. It is then concluded, in this paper, that proper time-dependent nucleation condition makes better correspondence in the transformation kinetics with JMAK theory.

The model considered here is, of course, too simple to be compared with realistic model or experimental results. Three-dimensional model may have additional effects in the kinetics. Physical understanding and its modeling on nucleation process, as well as environmental conditions such as temperature and material composition, are also to be discussed. Nevertheless, fundamental feature of the transformation kinetics presented in this paper is considered to be valuable for further modeling and utilization of the phase field model.

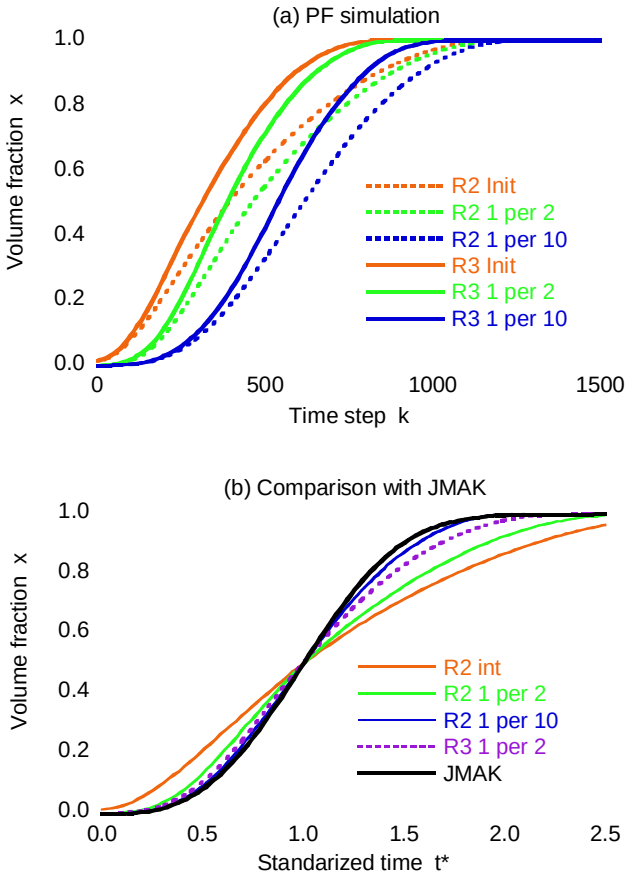


Figure 6. Evolution of volume fraction for Case 3.

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