Cell Dynamics Simulation of Kolmogorov-Johnson-Mehl-Avrami Kinetics of Phase Transformation -one and two dimensional cases-

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Abstract

In this study, we use the cell dynamics method to test the validity of the Kolmogorov-Johnson-Mehl-Avrami (KJMA) theory of phase transformation. This cell dynamics method is similar to the well-known phase-field model, but it is a more simple and efficient numerical method for studying various scenarios of phase transformation in a unified manner. We find that the cell dynamics method reproduces the time evolution of the volume fraction of the transformed phase predicted by the KJMA theory. Specifically, the cell dynamics simulation reproduces a double-logarithmic linear KJMA plot and confirms the integral Avrami exponents n predicted from the KJMA theory. Our study clearly demonstrates that the cell dynamics approach is not only useful for studying the pattern formation but also for simulating the most basic properties of phase transformation.

Key words: phase transformation, cell dynamics, KJMA theory
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1 Introduction

Phase transformation occurs by the nucleation and subsequent growth of a nucleus in a system where the first-order phase transformation takes place. It has attracted much attention for more than a half century [1–3] from a fundamental point of view as well as from technological interests.

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The nucleation and growth processes are often described in terms of the old standard theory called the KJMA theory developed by Kolmogorov [1], Johnson and Mehl [2], and Avrami [3]. According to this theory, the time evolution of the volume fraction of a new transformed phase follows the linear KJMA plot with the integral Avrami exponent n that is given by the slope. However, it is recognized that this theory often fails to explain experimental results [5,6]; neither the KJMA plot becomes linear nor the Avrami exponent becomes an integer.

In this study, we use the cell dynamics method [13,14] to study the validity of the KJMA theory. This method is computationally efficient, and yet it keeps the connection to the phase diagram through the Landau-type free energy.

2 Cell Dynamics Method for Nucleation and Growth

To study the phase transformation, it is customary to study the partial differential equation called the phase-field model [9,10] which is equivalent to the time-dependent Ginzburg-Landau (TDGL) [8] or Cahn-Hilliard model [7]:

$$\frac{\partial \psi}{\partial t} = -\frac{\delta \mathcal{F}}{\delta \psi}, \tag{1}$$

where δ denotes the functional differentiation, ψ is the *nonconserved* order parameter, and $\mathcal{F}$ is the free energy functional. This free energy is usually written as the square-gradient form

$$\mathcal{F}[\psi] = \frac{1}{2} \int \left[D(\nabla \psi)^2 + h(\psi) \right] \mathrm{d}\mathbf{r}. \tag{2}$$

where the local part $h(\psi)$ of the free energy $\mathcal{F}(\psi)$ we used is [9]

$$h(\psi) = \frac{1}{4} \eta \psi^2 (1 - \psi)^2 + \frac{3}{2} \epsilon \left(\frac{\psi^3}{3} - \frac{\psi^2}{2} \right). \tag{3}$$

which determines the bulk phase diagram and the value of the order parameter ψ in equilibrium phases.

This TDGL equation (1) for the nonconserved order parameter ψ was loosely transformed into a space-time discrete cell dynamics equation by Puri and Oono [14]. In their cell dynamics method, the partial differential equation (1) is replaced by a finite difference equation in space and time in the form

$$\psi(t+1, n) = F[\psi(t, n)], \tag{4}$$

where the time t is discrete and an integer, and the space is also discrete and is expressed by the integral site index n. Numerical efficiency of this cell dynamics method is obvious because it is essentially the cellular automaton with time and space being discrete integer. The mapping F is given by

$$F[\psi(t,n)] = -f(\psi(t,n)) + D\left[\ll \psi(t,n) \gg -\psi(t,n)\right],\qquad(5)$$

where $f(\psi) = dh(\psi)/d\psi$, and the definition of $\ll \cdots \gg$ for a two-dimensional square grid is given by [13,14]

$$\ll \psi(t,n) \gg = \frac{1}{6}\sum_{i=\mathrm{nn}}\psi(t,i) + \frac{1}{12}\sum_{i=\mathrm{nnn}}\psi(t,i),\qquad(6)$$

where "nn" denotes nearest neighbors and "nnn" next-nearest neighbors.

3 Numerical Results

3.1 KJMA kinetics by cell dynamics simulation

3.1.1 Site-saturation nucleation

In site-saturation nucleation, a fixed number of nuclei are prepared initially, and subsequent growth is monitored. The KJMA theory gives an analytical expression for the volume fraction f of the stable phase as a function of time t. In two dimensions, the formula leads to [9]

$$f = 1 - \exp\left(-\pi n_0 v^2 (t+t_0)^2\right),\qquad(7)$$

where n_0 is the number density (number per unit area) of the randomly distributed initial nuclei. v is the growth rate of the radius of each nucleus discussed in the previous section. t_0 is the origin of time which can hopefully take the incubation time of nucleation into account [10].

From eq. (7), we have

$$\log\left(-\ln(1-f)\right) = 2\log\left(t+t_0\right) + \text{constant}.\qquad(8)$$

Therefore, the KJMA theory predicts that a double logarithms $\log\left(-\ln(1-f)\right)$ versus $\log\left(t+t_0\right)$ is a straight line that is known as the KJMA plot with the integral tangent $n = 2$, which is called the "Avrami exponent".

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We have simulated the site-saturation nucleation using the cell dynamics method. Now a finite number of nuclei of the stable phase is distributed over the area we considered. The initial nuclei are circular and have the diameter $d = 8$, which is larger than the critical radius. Then, the evolution of the transformed volume is monitored as a function of time. We have considered the 100×100 system and introduced a finite number ($N_0 = 20$) of nuclei as the initial condition. Therefore, the number density of the initial nucleus is $n_0 = 20/10000 = 0.002$.
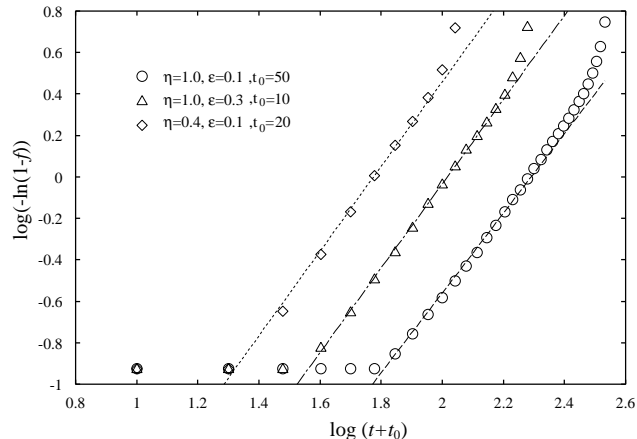

Fig. 1. Evolution of volume fraction f calculated by cell dynamics simulation for site-saturation nucleation as a function of time when incubation time t_0 is taken into consideration. All data follows an almost straight line and confirm the prediction of the KJMA theory given by eq. (8) with all the Avrami exponents n deduced from the straight lines in the figure being close to the theoretically predicted $n = 2$.

Figure 1 shows the KJMA plot when the incubation time t_0 is considered. Now, the time evolutions for several combinations of the potential parameters η and ϵ all fit the straight lines with almost the same Avrami exponent $n \simeq 2$, which is very close to the theoretical prediction, as shown in Table 1. The results in Figs. 1 clearly suggest that the incubation time t_0 should be carefully taken into account to deduce the Avrami exponent n when we analyze experimental as well as simulation data.

Table 1
Avrami exponent n for site-saturation nucleation and for continuous nucleation estimated by cell dynamics simulation for various potential parameters η and ϵ. The value theoretically predicted from the KJMA theory is $n = 2$ for site-saturation and $n = 3$ for continuous nucleation.

η	1	1	0.4
ϵ	0.1	0.3	0.1
n (site saturation)	1.92	2.04	2.03
n (continuous)	2.60	2.35	2.73

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Figure 2 shows the evolution of the morphology of the two-dimensional system for the site-saturation nucleation when $\eta = 0.4$ and $\epsilon = 0.1$. We observe the almost isotropic growth of every nucleus of the stable phase. At the time step ~ 100, almost all cells are transformed into the stable phase.
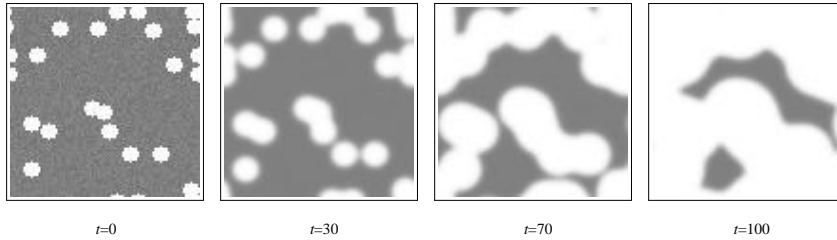

| $t=0$ | $t=30$ | $t=70$ | $t=100$ |

Fig. 2. Typical evolution pattern of site-saturation nucleation calculated by cell dynamics simulation when $\eta = 0.4$ and $\epsilon = 0.1$. The white area indicates the stable phase. Note that it takes only 100 steps to simulate the evolution from $t = 0$ to $t = 100$ because the cell dynamics is discrete cellular automaton.

3.1.2 Continuous nucleation

In the continuous nucleation, a new nuclear embryo is continuously introduced. The KJMA theory of continuous nucleation gives the analytical expression for the volume fraction f of the growing stable phase similar to (7). In two dimensions, we have

$$\log\left(-\ln(1-f)\right) = 3\log\left(t + t_0\right) + \text{constant}. \tag{9}$$

for continuous nucleation instead of (8). Therefore, a double logarithmic KJMA plot should give the "Avrami exponent" $n = 3$ instead of $n = 2$ of the site-saturation nucleation.

In our simulation, a constant nucleation rate $\dot{n}$ is achieved by introducing a new nucleus every $1/\dot{n}$ time step (nucleation time). At each nucleation time step, a position within the two-dimensional area is randomly selected. If the position is already occupied by the stable phase, no new nucleus is placed. If the position is not occupied by the stable phase, a new nucleus is placed and allowed to grow there. In this simulation, we have used a larger 200×200 system to avoid the finite-size effect as much as possible. The steady nucleation rate $\dot{n} = 0.1/40000$ is used. Therefore, a single nucleus is produced at every 10 time steps in the area 200×200.

The time evolution of the transformed volume f is plotted as a function of time t in Fig. 3 as the double logarithmic KJMA plot. Now, the time evolutions for several combinations of the potential parameters η and ϵ all fit the straight lines with almost the same Avrami exponent $n \simeq 3$, which is very close to the theoretical prediction, as shown in Table 1.
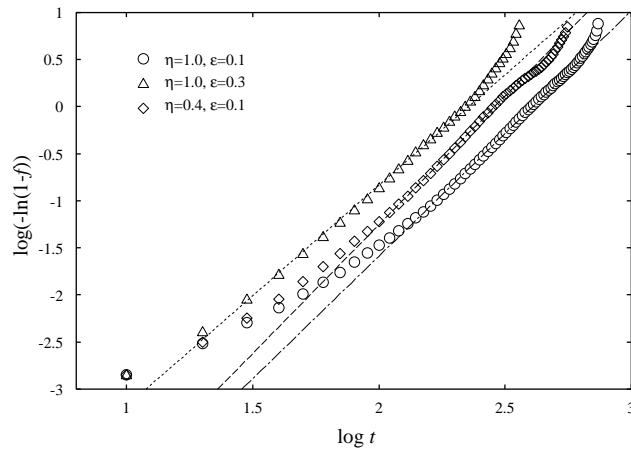
Fig. 3. Evolution of volume fraction f calculated by cell dynamics simulation for continuous nucleation as a function of time. The double logarithmic KJMA plot is used. All curves fit the straight lines predicted from the KJMA theory (9). The Avrami exponents deduced from the straight lines in the figure are all close to the theoretically predicted $n = 3$, as summarized in Table 1.

In this continuous nucleation case, because a nucleus is continuously produced, almost all cells are occupied at the later stage, and then the production of a nucleus stops. The situation becomes closer to the site-saturation nucleation.

Figure 4 shows the time evolution of the transformed volume f as a function of time when the nucleation occurs along a one-dimensional line. The Avrami exponent changes from $n = 2.48$ of the early stage to $n = 2.01$ of the late stage. Since the nucleation occurs along a one-dimensional line, the exponent $n = 2.48$ is smaller than theoretical $n = 3$ for two dimensional continuous nucleation. It becomes $n = 2.01$ at later stage because no new nucleus is produced and the site-saturation starts to occur.

Figure 5 shows the evolution pattern of continuous nucleation along one-dimensional line in two-dimensional system. When $t \simeq 260$ the grains start to touch each other, and the site saturation growth rather than the continuous growth starts to occur.

4 Conclusion and Discussion

In this study, we used a cell dynamics method to test the validity of the Kolmogorov-Johnson-Mehl-Avrami (KJMA) kinetic theory of phase transformation [17]. We simulated the growth of an ensemble of nuclei under the conditions of both the site saturation and continuous nucleation. We found a nearly linear behavior of the KJMA plot with the Avrami exponent close to the KJMA predicted value. We also suggested several extensions of the cell dy-
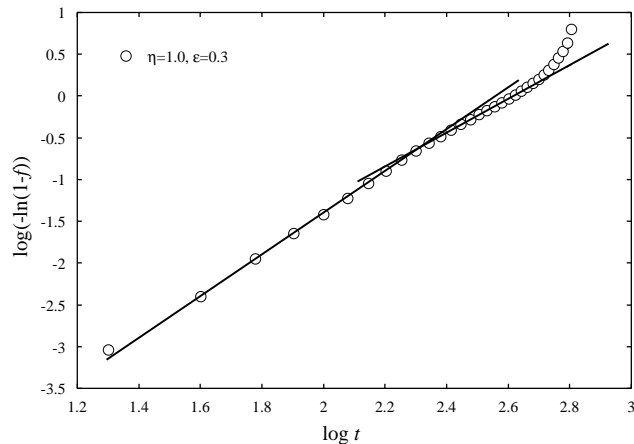
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Fig. 4. Evolution of volume fraction f calculated by cell dynamics simulation for continuous nucleation along a one-dimensional line in two-dimensional system as a function of time. The double logarithmic KJMA plot is used. There is a transition from the continuous nucleation with Avrami exponent $n = 2.48$ to the site-saturation nucleation with $n = 2.01$ when $\log t \simeq 2.4$, therefore $t \simeq 260$.
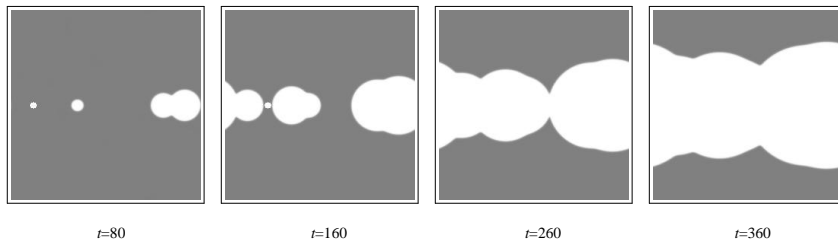

Fig. 5. Typical evolution pattern of continuous nucleation along one-dimensional line when $\eta = 1.0$ and $\epsilon = 0.3$. The transition from the continuous nucleation to the site-saturation nucleation occurs when $t \simeq 260$

namics method to study various contributions that may lead to the nonlinear KJMA behavior or nonideal Avrami exponent.

The results obtained in this study are summarized as follows:

(1) The cell dynamics method with a realistic free energy can simulate the growth of multiple nuclei and confirm the time evolution of the volume fraction of the transformed material predicted from the KJMA kinetic theory [1] and numerical simulation using TDGL [9].
(2) Therefore, the cell dynamics method can be used to simulate more complex scenarios of nucleation and growth such as the nucleation along a one-dimensional grain boundary.

The cell dynamics method is similar to the time-dependent Ginzburg-Landau or Cahn-Hilliard model based on the free energy functional. In contrast to the conventional cellular automaton approach to the phase transformation [12,4,15,16], no phenomenological energy that induces phase transformation is necessary.

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Therefore, the cell dynamics method is numerically efficient as a cellular automaton, yet it keeps the direct connection to the equilibrium phase diagram. This cell dynamics method can be used to test various scenarios of nucleation and growth in a unified manner [17].

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