Numerical Simulation of the Critical Velocity for Particle Pushing/Engulfment Transition in Fe–C alloys Using a Phase-field Model

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The particle/interface problem is numerically analyzed using a phase-field model in thin interface limit. A new double-well potential function in free energy density of the alloy is defined using a dilute solution approximation. With the function, a negative value of double-well potential height is usable and the mesh size restriction is largely relaxed. A pushing force for alloy systems is also introduced so as to relate to the interface energy change caused by the deformation of interface shape. With the pushing and drag forces calculated from the interface shape, the acceleration and velocity of the particle are estimated and the particle movement relative to the interface is analyzed. Using the model, the particle pushing and engulfment behaviors are successfully reproduced for the system of Fe–C alloys and an alumina particle. The critical velocities for the pushing/engulfment transition are determined for the particles with different diameters. The effect of initial carbon content on critical velocity is also examined and discussed in terms of the pushing force.

KEY WORDS: phase-field model; thin interface limit; particle–interface problem; critical velocity for pushing/engulfment transition; Fe–C alloy.

1. Introduction

Several investigations have been reported on the interaction between an insoluble particle and solid/liquid interface during solidification.1–8) In the experiments the velocity and shape of the interface around a particle are in-situ measured using an optical microscope for transparent materials1,2) and an X-ray transmission microscope for metals3) and the critical interface velocity for the pushing/engulfment transition is determined. Theoretical investigations have also been done to estimate the critical velocity.4,5) In the theory, it is defined as the velocity at which the dragging force becomes equal to the pushing force. In case of pure materials the dragging force is due to the viscosity flow of liquid and the pushing force is due to the difference of interface energies when the particle contacts with solid. In case of alloys the interface never touches the particle because of solute-enriched liquid between interface and particle. Hence different types of pushing force for alloy systems have been proposed.6,7) Han and Hunt, however, have pointed out all of them have some contradictory aspects.8) In addition, it is difficult to know the exact shape of the interface necessary for the estimation of the force. At present time, only the phase field model enables us to solve the problem.

A phase field model is a powerful tool that can describe the complex interface pattern evolution.9–16) It treats the interface as a finite area where the phase gradually changes from solid to liquid. Hence all the governing equations can be written in unified forms without distinguishing the interface and it has a great advantage in numerical calculations. In its practical application, however, there is the restriction on the mesh size. The conventional way to derive phase-field parameters, so-called sharp interface limit, assumes thermodynamic valuables to be constant in the interface region though it is not the case in reality. This difference causes the misestimation of the driving force for interface movement and the interface thickness in simulations must be smaller than capillary length to reduce the calculation error.12,14) Hence the calculation under a low undercooling condition or a small interface velocity condition requires a huge calculation time. Karma and Rappel derived the parameter relationships in pure materials under a thin interface limit condition to improve computational efficiency.14,15) Kim et al. extended the thin interface limit to alloy systems.16) In the previous work, we have simulated the change of interface shape and the solute redistribution around an insoluble particle in Fe–C alloys using their model.17) The calculation time, however, was still too large.
to estimate the critical velocity for the pushing/engulfment transition.

In the present work a new definition is applied to the double-well potential in free energy density for alloys. It relaxes the restriction on the mesh size and improves the computational efficiency by about hundred times comparing to the previous calculations. A new pushing force, which is derived from the increase rate of total interface energy according to the interface shape change, is also defined. The numerical calculations have been carried out for the system of Fe–C alloys and an alumina particle using the new model. The change of the interface shape during particle pushing and engulfment is shown and the critical velocity for the pushing/engulfment transition is estimated. The effects of particle diameter and initial carbon content of the alloys on the critical velocity are also discussed.

2. Calculations

2.1. Governing Equations using a Dilute Solution Approximation

A phase-field model for solidification is based on the Ginzburg–Landau free energy functional. We employ the recent model derived from the second law of thermodynamics. The governing equations for alloy solidification are given by

\[
\frac{\partial \phi}{\partial t} = M (e^{2\nabla^2 \phi} - f_\phi) \quad \text{(1)}
\]

\[
\frac{\partial c}{\partial t} = \nabla \left( \frac{D(\phi)}{f_{cc}} \nabla f_c \right) \quad \text{(2)}
\]

\[
f(c, \phi) = h(\phi) f_\phi^2(c) + (1 - h(\phi)) f_c^2(c) + W g(\phi)
\]

\[
h(\phi) = \phi (6\phi^2 - 15\phi + 10)
\]

where \(\phi\) is the phase-field, \(c\) is solute concentration, \(t\) is time, \(f\) is free energy density, \(D(\phi)\) is solute diffusion coefficient, the superscripts of \(S\) and \(L\) mean solid and liquid, the subscripts of \(\phi\) and \(c\) mean the first partial derivatives by \(\phi\) and \(c\), the subscript \(cc\) means the second derivative by \(c\), and \(M\) and \(e\) are the phase field parameters. The function \(g(\phi)\) is a double-well function which has local minima at \(\phi = 0\) and \(\phi = 1\), and a local maximum at \(0 < \phi < 1\), and \(W\) is the height of the double well potential.

In a dilute solution, chemical potentials of solvent A and solute B are given by,

\[
\mu^*_B(c) = \mu^*_B + \frac{RT}{V_m} \ln(\gamma_B c^*_B)
\]

\[
\mu^*_A(c) = \mu^*_A + \frac{RT}{V_m} \ln(\gamma_A c^*_A)
\]

\[
\mu^*_B(c) = \mu^*_B + \frac{RT}{V_m} \ln(1-c)
\]

\[
\mu^*_A(c) = \mu^*_A + \frac{RT}{V_m} \ln(1-c)
\]

where \(R\) is gas constant, \(T\) is temperature, \(V_m\) is molar volume, \(\gamma\) is activity coefficient and the superscript 0 means the quantity in the standard state. The equilibrium conditions are,

\[
\mu^*_B + \frac{RT}{V_m} \ln(\gamma_B c^*_B) = \mu^*_B + \frac{RT}{V_m} \ln(\gamma_B c^*_B)
\]

\[
\mu^*_A + \frac{RT}{V_m} \ln(1-c^*_B) = \mu^*_A + \frac{RT}{V_m} \ln(1-c^*_B)
\]

\[
\mu^*_B + \frac{RT}{V_m} \ln(1-c) = \mu^*_B + \frac{RT}{V_m} \ln(1-c)
\]

\[
\mu^*_A + \frac{RT}{V_m} \ln(1-c) = \mu^*_A + \frac{RT}{V_m} \ln(1-c)
\]

where \(c^*_B\) and \(c^*_S\) are the equilibrium concentrations of liquid and solid. As the free energy density is the sum of the free energy for liquid and solid phases and that for interface it is given by,

\[
\phi(c, \phi) = h(\phi) (c\mu^*_B + (1-c)\mu^*_B) + (1-h(\phi))(c\mu^*_L + (1-c)\mu^*_L) + W g(\phi)
\]

\[
= -\frac{RT}{V_m} h(\phi) \left[ c \ln \frac{c^*_S}{c^*_L} + (1-c) \ln \frac{1-c^*_S}{1-c^*_L} \right] + (c\mu^*_B + (1-c)\mu^*_L) + W g(\phi) \quad \text{(5)}
\]

By differentiating Eq. (5) the functions of \(f_\phi\), \(f_c\), and \(f_{cc}\) are obtained as,

\[
f_\phi = \frac{RT}{V_m} h'(\phi) \left[ c \ln \frac{c^*_S}{c^*_L} + (1-c) \ln \frac{1-c^*_S}{1-c^*_L} \right] + W g'(\phi)
\]

\[
f_c = -h(\phi) \frac{RT}{V_m} \ln \frac{c^*_S}{c^*_L} (1-c^*_S) + (c\mu^*_B - \mu^*_L) + \frac{RT}{V_m} \ln \frac{\gamma_B c}{1-c}
\]

\[
f_{cc} = \frac{RT}{V_m} \ln \frac{c}{1-c}
\]

By substituting them into Eqs. (1) and (2), we get the governing equations for a dilute solution.

Phase-field equation:

\[
\frac{1}{M} \frac{\partial \phi}{\partial t} = e^{2\nabla^2 \phi} - W g'(\phi)
\]

\[
+ \frac{RT}{V_m} h'(\phi) \left[ c \ln \frac{c^*_S}{c^*_L} + (1-c) \ln \frac{1-c^*_S}{1-c^*_L} \right] \quad \text{(6)}
\]

Diffusion equation:

\[
\frac{\partial c}{\partial t} = \nabla \cdot (D(\phi) c (1-c)) \left[ \frac{c}{1-c} - h(\phi) \ln \frac{c^*_S (1-c^*_S)}{c^*_L (1-c^*_L)} \right] \quad \text{(7)}
\]

2.2. Phase-field Parameters

In the phase field equation, there are three unknown parameters of \(e\), \(W\) and \(M\). The parameters \(e\) and \(W\) are related to the interface energy \(\sigma\) and the interface width \(2\lambda\) using one-dimensional stationary profiles of phase-field \(\phi(x)\) and solute concentration \(c_s(x)\). When the interface region is defined as \(0.1 < \phi < 0.9\), \(2\lambda\) and \(\sigma\) are written as follows,\(^{16}\)
In order to obtain the relationship between necessary conditions stated above. With the new function has a double-well shape and satisfies necessary conditions stated above. With the new function we get (see Appendix I),

\[ \frac{d\phi_0}{dx} = -\frac{1}{\varepsilon} \sqrt{2 \left( \frac{W + RT}{V_m} \right) g(\phi_0)} \]

As \( g(\phi) \) is always positive only \( W + RT/V_m \) should be positive and the negative value of \( W \) is allowed. In the previous work the positive value of \( W \) is necessary to obtain the relationship between \( \sigma \) and the parameters and it gives a strong limitation for the interface width.\(^{17} \) By employing the new \( g(\phi) \), the limitation is relaxed and we can use the larger value of \( 2\lambda \) than the previous one. As a result, we can make the calculation mesh size at least five times larger and thus the time step about 25 times larger than before. Therefore, the small interface velocity can be attained only with the new \( g(\phi) \), which is necessary for determining the critical velocity of the particle pushing.

The parameter \( M \) is related to the kinetic coefficient \( \mu^k \). In order to obtain the relationship between \( M \) and \( \mu^k \) we need to rewrite the phase field equation in a 1-D steady state into the compatible equation of interface temperature. According to the reference,\(^{16} \) the interface temperature is given by,

\[ T_i = T_m - m^2 c_1 \left[ 1 + \left( \frac{k^2 - k}{1 - k^2} \right) \alpha m^2 \right] \phi \] ...((9))

where \( m^2 \) is slope of liquidus line, \( k \) is distribution coefficient, \( k^2 \) is equilibrium distribution coefficient and \( \nu_n \) is normal interface velocity. As the third term in the right hand side of Eq. (9) corresponds to the kinetic undercooling, is given by,

\[ \mu^k = \frac{1 - k^2}{\alpha m^2} \]

where \( \alpha \) is (see Appendix II),

\[ \alpha = \frac{V_m}{MRT} \frac{\sigma}{\varepsilon^2} \left( \frac{\varepsilon}{\sqrt{2(W + RT/V_m)}} \right)^2 \]

The resultant phase field parameters are as followings

\[ \varepsilon = \frac{2\lambda \sigma}{I_1 I_2} \]

\[ W = \frac{\sigma I_2}{4\lambda I_1} - \frac{RT}{V_m} \]

\[ 1 \leq \frac{2RT\lambda}{M V_m} \left( \frac{1}{\mu^k} \frac{1 - k^2}{m^2} - \frac{2\lambda}{I_2} \right) \] \quad ...((10))

\[ I_1 = \int_0^\infty \sqrt{g(\phi_0)} \text{d}\phi_0 \]

\[ I_2 = \int_0^\infty \frac{1}{\sqrt{g(\phi_0)}} \text{d}\phi_0 \]

2.3. Forces Acting on a Particle

From the obtained interface shapes we can calculate the drag and pushing forces to the particle. The drag force, \( F_D \), is due to convective flow and is given by\(^{5} \)

\[ F_D = 6\pi \eta \frac{R^2}{d} \left( \frac{R_l}{R_l - R} \right) \]

where \( R \) is particle radius, \( \eta \) is viscosity, \( V \) is interface velocity, \( d \) is the distance between interface and particle, and \( R_l \) is curvature radius of the interface. In case of pure materials the pushing force, \( F_p \), is due to the difference of interface energies between particle/solid, particle/liquid and solid/liquid \( \Delta \sigma = \sigma_{ps} - (\sigma_{pl} + \sigma_{sl}) \), and is usually expressed by\(^{5} \)

\[ F_p = 2\pi R \cdot \Delta \sigma \left( \frac{a_0}{a_0 + d} \right)^n \left( \frac{R_l}{R_l - R} \right) \]

where \( a_0 \) is sum of the radii of atoms, and \( n \) has the constant value from 2 to 7. The force above is derived from the assumption that the interface is in contact with the particle. It is not applicable to the alloy system as pointed out by Hunt, because there always exists the solute-enriched liquid between the interface and the particle. Therefore, we need a different definition of the pushing force for the alloy system.

When the interface approaches to the particle the interface becomes concave and the total interface area increases. As the interface energy is regarded as the interface tension for isotropic materials, the pushing force could be related to the increase of the total interface energy. Then, the force can be proportional to the derivative of the interface energy by the representative length of the interface shape. For simplicity, we assume the interface shape is a sinusoidal one, \( z = l \sin(\pi r/r_0) \), as shown in Fig.1, where \( l \) is the amplitude and \( 2r_0 \) is the wavelength. Hence the total area of the interface is given by, \( S = 2\pi r_0 l \), then the pushing force is defined by,

\[ F_l = \frac{\partial (S \sigma)}{\partial l} = \pi r_0 \sigma \]
The pushing force should quickly decrease as the interface–
particle distance increases. Therefore, we assume the same
shape of a dumping factor in Eq. (12), which is a function
of the interface–particle distance.

\[ F_1 = \pi \sigma \left( \frac{a_0}{a_0 + d} \right)^n \] ..........................(13)

By substituting the relationship, \( r_0 = \pi \sqrt{R_1 l} \), into Eq. (13),
the pushing force is finally obtained as:

\[ F_1 = \pi \sigma \cdot \pi \sqrt{R_1} \left( \frac{a_0}{a_0 + d} \right)^n \] ..........................(14)

In the present work we assume the value of \( n \) to be two\(^{5}\)
and \( R_1 = R + d \) for simplicity. Using Eqs. (11) and (14) we
can estimate the net force acting on the particle, the accel-
eration and velocity of the particle.

2.4. Numerical Calculations

As we are considering a spherical particle, we assume the
system to be axial-symmetric. Hence we get the governing
equations in the cylindrical coordinates.

Phase-field equation:

\[ \frac{\partial \phi}{\partial t} = M \left[ c^2 \left( \frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} + \frac{\partial^2 \phi}{\partial z^2} \right) \right] - W g(\phi) \]

\[ + \frac{RT}{V_m} h(\phi) \left( c \ln \frac{c_s}{c_L} + (1-c) \ln \frac{1-c_s}{1-c_L} \right) \] ..........................(15)

Diffusion equation:

\[ \frac{\partial c}{\partial t} = \frac{D(\phi)}{r} \left[ \ln \frac{c}{1-c} \right]^{-1} \frac{\partial}{\partial r} \left\{ -h(\phi) \ln \frac{c_s(1-c_s)}{c_L(1-c_L)} \right\} \]

\[ + \frac{RT}{V_m} (\mu_{bl} - \mu_{bl}) + \ln \frac{\gamma_{bc}}{1-c} \] ..........................(16)

where \( D(\phi) = D^L (0 < \phi < 0.9) \), \( D(\phi) = D^S (0.9 < \phi \leq 1) \), \( D^L \)
and \( D^S \) are the solute diffusivity in liquid and in solid, respec-
tively. The equations are numerically solved using an ex-
licit finite difference scheme. Note that the system is
assumed to be isothermal and the effect of latent heat gen-
eration is neglected because of the computing time limitation.
It would be acceptable for the case the growth is mainly
dominated by solute diffusion.

In the particle region, the phase-field mobility and the
solute flux are put to be zero. In case of the moving parti-
cle, we need another condition for the mass conservation.
Strictly speaking, it is necessary to consider the liquid flow
according to the movement of the particle and to solve the
Navier–Stokes equation. We, however, adopt a simple pro-
cedure for satisfying the mass conservation. By advancing
the particle, just as solute of the initial concentration is
subtracted from the front as shown in

Fig. 2. Procedure for mass conservation around an advancing
particle.

\[ \frac{\partial}{\partial r} \left[ (\mu_{bl} - \mu_{bl}) + \ln \frac{\gamma_{bc}}{1-c} \right] \] ..........................(16)

The number of
meshes in radius is put in the area. Boundary conditions
and sizes are schematically shown in

Fig. 3. The number of
meshes in the interface region is four. Mesh size and time
step are \( 5.0 \times 10^{-3} \text{ m} \) and \( 2.5 \times 10^{-3} \text{ s} \), respectively. The val-
ues of \( 8 \times 10^{-10} \text{ m} \) are used for \( a_0 \) in Eq. (13), which are
taken from the literatures.\(^{5}\)

We assume that the system temperature is uniform and
decreases with a constant rate as followings, \( T = T_0 - \delta T \times N \), where \( T_0 \) is initial temperature which is slightly lower than the liquidus temperature of the alloy (=1800 K), \( N \) is number of time steps and \( \delta T \) is cooling rate per time step. Interface velocity depends on \( \delta T \) and the interface location. Therefore, the average velocity at the height of the particle center is used as a representative value. The interface velocities for different cooling rates obtained by the preliminary calculations are tabulated in Table 1. In the calculation, the system of Fe–0.5mol% C alloy and a silica or an alumina particle is examined and their material properties are shown in Table 2.

### Table 1. Interface velocity at each cooling rate obtained in preliminary calculations.

<table>
<thead>
<tr>
<th>( \delta T ) (K/step)</th>
<th>Cooling rate (K/s)</th>
<th>Interface velocity (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0 \times 10^6</td>
<td>40</td>
<td>4.02 \times 10^4</td>
</tr>
<tr>
<td>1.5 \times 10^6</td>
<td>60</td>
<td>6.23 \times 10^4</td>
</tr>
<tr>
<td>2.5 \times 10^6</td>
<td>100</td>
<td>1.01 \times 10^2</td>
</tr>
<tr>
<td>5.0 \times 10^6</td>
<td>200</td>
<td>1.78 \times 10^3</td>
</tr>
<tr>
<td>7.5 \times 10^6</td>
<td>300</td>
<td>2.29 \times 10^3</td>
</tr>
<tr>
<td>1.0 \times 10^7</td>
<td>400</td>
<td>2.38 \times 10^3</td>
</tr>
</tbody>
</table>

### Table 2. Physical properties of Fe–C alloy and alumina.

<table>
<thead>
<tr>
<th>Properties of Fe–C</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>interface energy ( \alpha ) (J/m(^2))</td>
<td>0.204</td>
</tr>
<tr>
<td>melting point ( T_m (K) )</td>
<td>1810</td>
</tr>
<tr>
<td>solute diffusivity (liquid) ( D_s (m^2/s) )</td>
<td>2.0 \times 10^{-8}</td>
</tr>
<tr>
<td>solute diffusivity (solid) ( D_s (m^2/s) )</td>
<td>6.0 \times 10^{-9}</td>
</tr>
<tr>
<td>molar volume ( V_m (m^2/mol) )</td>
<td>7.7 \times 10^{-4}</td>
</tr>
<tr>
<td>Equilibrium distribution coefficient ( \alpha )</td>
<td>0.204</td>
</tr>
<tr>
<td>slope of liquidus ( m_l (K/mol%C) )</td>
<td>-18/2</td>
</tr>
<tr>
<td>viscosity ( \eta (Pa\cdot s) )</td>
<td>4.75 \times 10^{-3}</td>
</tr>
<tr>
<td>kinetic coefficient ( \mu (m/sK) )</td>
<td>0.003</td>
</tr>
<tr>
<td>Property of the particle</td>
<td></td>
</tr>
<tr>
<td>Density of ( Al_2O_3 ) ( \rho_{Al_2O_3} (kg/m^3) )</td>
<td>3.99 \times 10^3</td>
</tr>
</tbody>
</table>

3. Results and Discussions

#### 3.1. Particle Pushing and Engulfment by the Interface

Figure 4 shows the change in the interface shape with time when the particle of 2 \( \mu m \) in diameter is pushed by the interface, where the cooling rate and the interface velocity are 40 K/s and 4.02 \times 10^{-2} m/s, respectively. As the interface approaches to the particle, the solute concentration in the liquid increases and the interface becomes concave. Then, the particle gradually begins to move forward as the pushing force exceeds the drag force. At the early stage of pushing, the particle sinks into the interface because the particle velocity is smaller than that of the interface. As the interface becomes more concave and the pushing force increases, the particle velocity increases. When the both velocities become to be equal the particle moves steadily with the interface.

![Fig. 3. Calculation area and boundary conditions.](image1)

![Fig. 4. Time history of the interface shape during particle pushing. Particle diameter is 2 \( \mu m \) and interface velocity is 4.02 \times 10^{-4} m/s.](image2)

(a) 5.0 ms (b) 7.0 ms (c) 11.0 ms (d) 17.0 ms

![Fig. 5. Time history of the interface shape during particle engulfment. Particle diameter is 2 \( \mu m \) and interface velocity is 2.38 \times 10^{-3} m/s.](image3)

(a) 1.25 ms (b) 1.5 ms (c) 2.0 ms (d) 2.5 ms
On the contrary, the particle engulfment occurs when the interface velocity always exceeds the particle one. Figure 5 shows the change in the interface shape with time when the particle of 2 \( \mu m \) is engulfed by the interface. In the case of Fig. 5 the cooling rate is increased to 400 K/s and the interface velocity to 2.38 \( \times 10^{-3} \) m/s. At the early stage the particle is slightly pushed ahead but it is soon engulfed by the interface because the interface velocity is much larger than the particle one. No significant liquid trench ahead of the particle reported in the previous work\(^{17}\) is formed and the interface promptly covers the particle. It is due to the difference between the present 3-D and the previous 2-D calculations.

### 3.2. Critical Velocity for Pushing/Engulfment Transition

As shown in the previous section, we can numerically reproduce the transition from pushing to engulfment of a particle by changing the cooling rate or the interface velocity. For the particles of 1, 2 and 4 \( \mu m \) in diameter the calculations are carried out in the range of the interface velocity from 2\( \times 10^{-4} \) m/s to 4\( \times 10^{-3} \) m/s. Figure 6 shows the obtained results for an alumina particle, in which the pushing and engulfment of the particle are shown as open and filled circles, respectively. The critical velocities for the pushing/engulfment transition are about 3\( \times 10^{-3} \) m/s, 8\( \times 10^{-4} \) m/s and 2.5\( \times 10^{-4} \) m/s for the particles of 1, 2 and 4 \( \mu m \), respectively. These values are larger than the reported ones which are between 20 and 10\( \times 10^{-6} \) m/s.\(^{5}\) The difference, however, is not critical because most of experiments are performed for pure materials and the data for small particles are very limited. In addition, the calculated critical velocity depends on the magnitude of the defined pushing force, which is affected by the value of \( a_0 \) in Eq. (14). For example, the critical velocity for the particle of 2 \( \mu m \) becomes three times larger by increasing the value of \( a_0 \) by three times, though the further increase of \( a_0 \) does not affect much. Therefore, we should be careful in the quantitative comparison between experiments and calculations.

In Fig. 6 the well-known relationship between critical velocity and particle diameter, \( V_c \sim 1/R \), are also shown as a broken curve. The critical velocity for a small particle is only much larger than the curve. It is presumably due to the curvature effect. The interface near the particle has negative curvature and its equilibrium temperature is raised by Gibbs–Thomson effect. In the present calculations, the system is assumed to be isothermal and thus the degree of interface undercooling is locally different. When the diameter of the particle is small the curvature effect is not negligible, and the undercooling and velocity of the interface near the particle become large. As a result, the interface shape spreads widely so that the small particle becomes difficult to be engulfed by the interface and the critical velocity increases.

Figure 7 shows the critical velocities for a particle with smaller density. For example, we input the density of the silica particle (2.65\( \times 10^3 \) kg/m\(^3 \)). The value of critical velocity is different from that of alumina particle only when the particle diameter is 1 \( \mu m \). The deviation of the critical velocities from the broken curve becomes larger than that of alumina particle. These results show the critical velocity for a heavier particle agrees better with the curve of 1/\( R \).

For discussing the relationship between critical velocity and particle diameter in detail, we need the results for larger particles. It is still difficult to attain such small interface velocity as to estimate the critical velocity for large particles because of computing time limitation.

### 3.3. Effect of Carbon Content on Critical Velocity

It is useful to investigate the effect of the initial concentration of the alloy on the critical velocity. The additional calculations were carried out for the alloys with 0.1, 0.25 and 1.0 mol% C and the critical velocities were examined for the particle of 2 \( \mu m \). Figure 8 shows the change in critical velocity for the silica/alumina particle with initial carbon content. The results of these two kinds of particle are the same at the calculated points. The critical velocity slightly decreases with decrease of carbon content from 1.0 to 0.25 mol% C. That of Fe–0.1mol%C alloy is, however, larger than those of others. It is explained from the functional form of the pushing force. As shown in Fig. 9 the distance between particle and interface, \( d \), becomes large as the initial carbon content increases because the solute is enriched more ahead of the interface. It makes the pushing force and then the critical velocity smaller because the dumping factor in Eq. (14) decreases. On the other hand, the curvature radius of the interface increases with increase of \( d \) and it makes the pushing force and the critical velocity
larger. Therefore, the critical velocity is determined by the two counter effects and has a minimum at 0.25 mol% C. Note that the interface shape shown in Fig. 9 changes with carbon content. The interface comes close to the side of the particle as carbon content decreases. It causes the error in estimating the pushing force because the distance between particle and interface has two counter effects on the force. The results in the present work are quite satisfactory and the wide potentiality of the phase-field model to interface-particle problems is demonstrated. For the further quantitative investigation, the experimental data for small particles in alloy systems are specially required.

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REFERENCES


Appendix 1

Under the boundary conditions of $\phi_0(-\infty)=1$, $\phi_0(\infty)=0$, $c_0(-\infty)=c_s^e$ and $c_0(\infty)=c_L^e$, we get one-dimensional stationary solutions for phase-field $\phi_0(x)$ and solute concentration $c_0(x)$ by solving the following equations.

Phase-field equation:

$$
\varepsilon^2 \frac{d^2 \phi_0}{dx^2} - W g(\phi_0) + \frac{RT}{\Gamma_m} h'(\phi_0) \left[ c_0(x) \ln \frac{c_0^e}{c_L^e} + (1-c_0(x)) \ln \frac{1-c_0^e}{1-c_L^e} \right] = 0
$$

...........................................(A-1)
Diffusion equation:
\[
\ln \frac{c_0 - h(\phi_0)}{1 - c_0} \ln \frac{e\gamma^5(1 - c^5_f)}{c^5_f(1 - c^5_f)} = \text{const.} \quad \ldots \ldots (A-2)
\]

By using the boundary conditions \( c_0(-\infty) = c^5_f \), \( c_0(\infty) = c^5_f \), \( h_0(-\infty) = 1 \) and \( h_0(\infty) = 0 \), we obtain,
\[
c_0(x) = \tilde{c}_0(\phi_0) = \frac{c^5_f}{c^5_f + (1 - c^5_f)p \gamma^5(\phi_0)}
\]

\[p = \frac{c^5_f(1 - c^5_f)}{c^5_f(1 - c^5_f)}\]

By integrating Eq. (A-1) by \( x \) from \( -\lambda \) to \( x \) (or by \( \phi_0 \) from 1 to \( \phi_0 \)) after multiplying \( d\phi_0/dx \),
\[
\mathcal{E}^2 \left( \frac{d\phi_0}{dx} \right)^2 \int_{-\lambda}^{x} -W(\phi_0) \, d\phi_0 = \int_{-\lambda}^{x} \left( \frac{c^5_f}{c^5_f + (1 - c^5_f)p \gamma^5(\phi_0)} \right) \, d\phi_0 = 0
\]

With the integral of \( c_0(x) \) and the condition \( d\phi_0/dx = 0 \) at \( x = -\lambda \),
\[
\int_{-\lambda}^{x} c_0(x) \, dh(\phi_0) = \int_{-\lambda}^{x} \frac{c^5_f}{c^5_f + (1 - c^5_f)p \gamma^5(\phi_0)} \, d\phi_0
\]

we get
\[
\mathcal{E}^2 \left( \frac{d\phi_0}{dx} \right)^2 -W(\phi_0) = \int_{-\lambda}^{x} \left[ \frac{c^5_f}{c^5_f + (1 - c^5_f)p \gamma^5(\phi_0)} \right] \, d\phi_0 = 0
\]

When we define \( g(\phi_0) \) as,
\[
g(\phi_0) = -\ln \left[ \frac{c^5_f}{c^5_f + (1 - c^5_f)p \gamma^5(\phi_0)} \right] = 0
\]

then
\[
\frac{d\phi_0}{dx} = -\frac{1}{\mathcal{E} \sqrt{2(W + RT/V_m)}} g(\phi_0) \quad \ldots \ldots (A-3)
\]

From Eqs. (A-3) and (A-4) we obtain the interface width \( 2\lambda \) and the interface energy \( \sigma \) as the functions of \( W \) and \( \mathcal{E} \).
\[
2\lambda = \int_{-\lambda}^{x} dx = \int_{0}^{1} \frac{dx}{d\phi_0} \, d\phi_0
\]
\[
\sigma = \mathcal{E}^2 \int_{0}^{\lambda} \left( \frac{d\phi_0}{dx} \right)^2 \, dx = \mathcal{E}^2 \int_{0}^{1} \frac{d\phi_0}{dx} \, d\phi_0
\]

Appendix II

From Eq. (36) in the reference (note that the definition of \( \alpha \) is different from that in the reference by the factor \( V_m/RT \) and \( f_{\alpha} \), in the main text, \( \alpha \) is given by,
\[
\alpha = \frac{V_m}{\mathcal{E}^2} \int_{0}^{1} \left( \frac{d\phi_0}{dx} \right)^2 \, dx
\]

\[
+ \frac{V_m}{RT} \int_{0}^{\lambda} \frac{f_{\alpha}(\phi_0)}{D(\phi_0)} (c_0(x) - c^5_f) \, dx \, d\phi_0
\]

\[
= \frac{V_m}{\mathcal{E}^2} \int_{0}^{\lambda} \frac{\sigma}{\sigma} \frac{\sigma}{\sigma} \, dx \, d\phi_0
\]

\[
+ \int_{0}^{1} \left( \int_{0}^{\lambda} \frac{c_0(x) - c^5_f}{D(\phi_0)c_0(x)(1 - c_0(x))} \, dx \, d\phi_0 \right) \, d\phi_0
\]

Using Eq. (A-5) and \( d\phi_0/d\phi_0 \) obtained by differentiating Eq. (A-2), we get,
\[
\alpha = \frac{V_m}{\mathcal{E}^2} \int_{0}^{\lambda} \frac{\sigma}{\sigma} \frac{\sigma}{\sigma} \, dx \, d\phi_0
\]

\[
\zeta = \int_{0}^{1} \left( \int_{0}^{\lambda} \frac{\tilde{c}_0(\phi_0) - c^5_f}{D(\phi_0)c_0(\phi_0)(1 - c_0(\phi_0))} \, dx \, d\phi_0 \right) \, d\phi_0
\]

\[
\times \left( \frac{\tilde{c}_0(\phi_0) - c^5_f}{D(\phi_0)c_0(\phi_0)(1 - c_0(\phi_0))} \frac{1}{\gamma(\phi_0)} \right)
\]

\[
\times h'(\phi) \tilde{c}_0(\phi_0)(1 - \tilde{c}_0(\phi_0)) \ln p d\phi_0
\]