Friction Pressure Method for Simulating Solute Drag and Particle Pinning in a Multi Phase Field Model

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Abstract. In many polycrystalline materials, second phase particles and solute atoms impose a drag pressure on the motion of grain boundaries. The drag effect occurs on a scale comparable to the particle diameter and interface thickness. However, to simulate grain growth with numerical efficiency one requires a model that captures the drag pressure on the interfaces but does not resolve the fine particles or solute segregation spike. In this article, a multi-phase field model is proposed to simulate the evolution of microstructure under constant and velocity dependent drag pressures. The accuracy of the model is confirmed in comparison with analytical expressions for a shrinking circular grain. Application of the model is presented for grain growth in two dimensions under particle pinning. Measuring curvature of grain boundary segments reveals that in the completely pinned structure, the average driving pressure is not equal but lower than the pinning pressure. Considering this effect, the predicted limiting grain size is about three times larger than that assumed in conventional mean field theories. Based on this observation, a correction factor is introduced for these mean field models. The proposed phase field formulation is also applied to simulate grain growth in the presence of solute drag. The grain growth kinetics follows a phenomenological relationship that can be described with a power law with a time exponent in the range of 0.35 to 0.50. The deviation of the time exponent from 0.5, associated with ideal grain growth, and its correlation to the solute drag parameters is discussed.

PACS numbers: 61.50.-f, 61.50.Ah, 61.72.Mm, 81.30.-t

1. Introduction

Second phase particles and solute atoms have been used as an important constituent to design materials and processes due to their ability to control motion of grain boundaries [1, 2]. As an example, in high strength low alloy (HSLA) steels, addition of small amount of microalloying elements creates fine carbide precipitates. These particles limit motion of grain boundaries by the particle pinning mechanism. In addition, solute atoms in the matrix segregate to grain boundaries and impede their movement by solute drag [3].

In many polycrystalline materials, second phase particles have diameters on the order of tens of nanometres and segregation of solutes atoms occurs on a scale comparable to the grain boundary thickness. However, other microstructural features like grains are usually on the micron scale. Theoretical approaches have addressed the multi-scale nature of interface drag. The Zener-Smith theory [4] and its later modifications [5] represent a mean field approach to model particle pinning. Similarly, mean field models for solute drag explain segregation of solutes at the grain boundaries and provide the drag pressure [6, 7, 8, 9]. Mean field models are, however, unable to capture the role of pinning and solute drag pressure on topology of grain structure. Therefore, more detailed investigation of the phenomenon requires microstructural evolution models, e.g. phase field method, that are formulated on the length scale of the microstructure, the so called meso-scale, while including the effect of interface drag.

Phase field models have been established as a powerful and versatile simulation method to describe microstructural evolution in materials [10, 11]. In particular, several multi-phase field models are capable of simulating grain growth [12, 13, 14]. Grain boundary drag phenomenon has also been studied with the phase field approach. Moelans et al. [15] proposed a model in which particles and grains are resolved in the same scale. Using this model, grain growth in two dimensions [16], three-dimensional thin-films [17] and three-dimensional bulk systems [18] has been simulated. Modelling solute drag is more challenging since the difference between the length scale of solute segregation and the grain microstructure is several orders of magnitude. As a result, solute drag has been only studied in one dimension for a flat grain boundary [19, 20, 21] or two-dimensional nano grain structures [22].

Apel et al. [23] developed a dual-scale approach to simulate particle pinning. In the smaller scale, they modelled motion of a grain boundary moving through an array of particles and obtained a relationship between the grain boundary velocity and the applied driving pressure. This relationship was interpreted as an “effective mobility” that is a function of pinning pressure. The resulting grain boundary mobility was included in a phase field model to simulate grain growth on a larger scale.

The present work, however, builds a model on an energetic approach to include the drag pressure directly into the phase field equation. It avoids the phenomenological modification of the grain boundary kinetics through mobility. This approach can be readily extended to other more complex forms of interface drag including any velocity...
dependent drag pressure. Regardless of the source of drag, we propose a multi-phase field formulation to impose a generalized drag pressure on interfaces. Drag pressure can be constant (pinning with fine inert particles) or depend on the interface velocity (solute drag). We present an application of the proposed formulation to simulate grain growth in a system containing fine second phase particles and a system with the presence of solute drag.

2. Phase Field Formulation

2.1. Multi-Phase Field Formulation and Driving Pressure

Based on the diffuse interface model of Allen and Cahn [24], Chen et al [13, 14] proposed a multi-phase field formulation that can be applied to model grain growth. Moelans [25, 26] proposed a methodology to relate phase field model parameters to the physical characteristics of the system. Using this approach, one can quantitatively model a particular system with known grain boundary properties.

In the multi-phase field formulation, at any point in space, \( r \), each grain is identified as a separate field or order parameter \( \eta_i \). Order parameters have the equilibrium value of one inside a grain and zero outside and gradually change at the grain boundary from one to zero. According to the Allen-Cahn theory [24, 13] one can expand the energy of the system into two contributing sources; (i) the local energy density at each point, \( f_{\text{loc}} \), and (ii) a term related to the gradient of the order parameter, \( \nabla \eta_i \). Therefore, the following relationship for the energy of the system is obtained:

\[
F = \int \left[ f_{\text{loc}} + \kappa \sum_{i=1}^{p} |\nabla \eta_i|^2 \right] d^3 r \tag{1}
\]

where \( F \) is the free energy of the system with respect to a standard state, \( p \) is the number of order parameters (grains) and \( \kappa \) is a phenomenological constant adjusting the energy contribution from the gradient of \( \eta \) at the interface. \( f_{\text{loc}} \) consists of two parts: (i) variation of local energy density across the interface \( (f_0) \) and (ii) a contribution due to the internal bulk energy of the grains \( (f_d) \), i.e:

\[
f_{\text{loc}} = f_0 + f_d. \tag{2}
\]

Here \( f_0 \) is chosen to have a minimum of 0 inside each grain and a maximum at the interface resembling a double-well shape. Based on the formulation of Fan and Chen [14], the following form for \( f_0 \) is proposed:

\[
f_0 = m \left( \sum_{i=1}^{p} \frac{\eta_i^4}{4} - \sum_{i=1}^{p} \frac{\eta_i^2}{2} + \frac{3}{2} \sum_{i=1}^{p} \sum_{j \neq i}^{p} \eta_i^2 \eta_j^2 + \frac{1}{4} \right), \tag{3}
\]

where \( m \) is a constant that determines the height of the energy peaks between each minima.
The bulk energy of grain $i$, $G_i$, is considered in comparison to a standard state. In a case where bulk energy of all grains is equal ($G_i = G_j$), $f_d = \text{const.}$ and the driving pressure for grain boundary movement is only due to its curvature [27]. However, in a case of different bulk energies, $G_i \neq G_j$, there is an additional driving pressure of $\Delta G_{ij} = G_j - G_i$ on the grain boundaries. In this situation, $f_d$ interpolates the bulk energy of each grain across the grain boundary. For two order parameters the contribution to the local energy density due to the bulk energy is written as:

$$f_d = 3 \sum_{i=1}^{2} \sum_{j \neq i}^{2} \left( \frac{\phi_{ij}^2}{2} - \frac{\phi_{ij}^3}{3} \right) \Delta G_{ij} + \frac{1}{2} \sum_{i=1}^{2} G_i \tag{4}$$

The driving pressure function is formulated using a variable, $\phi_{ij}$ which is 0 inside grain $i$ and 1 inside grain $j$, i.e:

$$\phi_{ij} = \frac{\eta_j - \eta_i + 1}{2} \tag{5}$$

Fig. 1 shows the local energy density, $f_{\text{loc}}$ of a system with $G_1 = 0.3$ and $G_2 = 0$. The value of $f_{\text{loc}}$ at $\eta_i = 1$ and $\eta_j = 0$ is equal to the $G_i$ value. Across the interface, the local energy density follows a curve of the double well shape.

The functional $F$ determines the thermodynamic properties of the system. Evolution of the system follows the Allen-Cahn relaxation dynamics [24], resulting in a set of partial differential equations (PDEs):

$$\frac{\partial \eta_i}{\partial t} = -L \frac{\delta F}{\delta \eta_i} = -L \left( \frac{\partial f_0}{\partial \eta_i} + \frac{\partial f_d}{\partial \eta_i} - \kappa \nabla^2 \eta_i \right). \tag{6}$$

Here $\delta$ denotes functional derivation and $L$ is a kinetic constant. The derivation of $f_d$ can be simplified using $\eta_j = 1 - \eta_i$ [25], as:

$$\frac{\partial f_d}{\partial \eta_i} = 3\eta_i\eta_j \Delta G_{ij}. \tag{7}$$

It is shown in Appendix A that Eq. 7 satisfy the sharp interface limit assumptions. Each pair of order parameters between $\eta_i$ and $\eta_j$ has a contribution to the $\partial f_d/\partial \eta_i$ term. Therefore, a set of evolution PDEs are obtained by generalizing Eq. 7 over all order parameter pairs:

$$\frac{\partial \eta_i}{\partial t} = -L \left[ m \left( \eta_i^3 - \eta_i + 3\eta_i \sum_{j \neq i}^{p} \eta_j^2 \right) - 3\eta_i \sum_{j \neq i}^{p} \eta_j \Delta G_{ij} - \kappa \nabla^2 \eta_i \right]. \tag{8}$$

In this formulation, model parameters are related to the energy ($\sigma$), mobility ($M$) and thickness ($l$) of the grain boundary as follows [25, 26]:
\[ \sigma = \frac{1}{3} \sqrt{2m\kappa} \]  
\[ M = \frac{3}{2} L \sqrt{\frac{2\kappa}{m}} \]  
\[ l = \sqrt{\frac{8\kappa}{m}} \]  
(9)  
(10)  
(11)

2.2. Applying Drag Pressure

In the previous section, we described how to apply a driving pressure on an interface by shifting the local energy density. In this section, we use the same method to apply a drag pressure on the interface. For the sake of simplicity, we assume that the driving pressure \( P_d \) on an interface is only due to its curvature. A drag (or friction) pressure, \( P_f \), can be considered as a negative driving pressure or a pressure that opposes movement of the interface. If the driving pressure is smaller than the drag pressure, the interface stops moving. To create the effect of drag, the local energy density can be shifted depending on the direction of the interface velocity as it is shown schematically in Fig. 2. In general, the drag pressure can be a function of the normal interface velocity \( v_{ij} \) of grain \( i \) moving into grain \( j \). In such a formalism the energy shift between grain \( i \) and \( j \) can be written as:

\[
\Delta G_{ij}(r) = \begin{cases} 
-P_f(v_{ij}) & P_d > P_f \\
-P_d & P_d < P_f 
\end{cases}
\]

(12)

Applying this approach to the phase field method, we define the normal velocity in the curvilinear coordinate system as a scalar velocity field for each order parameter \( \eta \) using the chain rule, i.e.

\[
v_{\eta i}(r) = \frac{\partial n_i}{\partial t} = \frac{\partial \eta_i}{\partial t} \frac{\partial n_i}{\partial \eta_i}.
\]

(13)

Here \( n_i \) is the distance along the curvilinear coordinate \( n_i \) normal to the interface pointing from within grain \( i \), into grain \( j \). The order parameter profile across the interface is known analytically, i.e. [26]:

\[
\frac{\partial \eta_i}{\partial n_i} = \frac{1}{2} \sqrt{\frac{m}{2\kappa}} \left[ 1 - \tanh^2 \left( \sqrt{\frac{m}{2\kappa}} n_i \right) \right] = \frac{1}{2} \sqrt{\frac{m}{2\kappa}} \left[ 1 - (2\eta_i - 1)^2 \right].
\]

(14)

At each point on the \( n_i \) axis through the boundary between grains \( i \) and \( j \), \( v_{\eta i} \) has a same value and is equal to the local grain boundary velocity \( v_{\eta i} = |v_{ij}| \) and \( v_{\eta j} = -v_{\eta j} \). According to Eq. 12, the value of the local energy shift \( \Delta G_{ij} \) in Eq. 8 can be obtained with the condition that \( P_f(-v_{\eta i}) = -P_f(v_{\eta i}) \), since \( \Delta G_{ij} = -\Delta G_{ji} \). The curvature driven pressure in the phase field is distributed across the interface. Similar to Eq. 7, \( f' = 3\eta_i \eta_j P_d \) where \( f' \) is:
\[
f_i' = \frac{\partial f_0}{\partial \eta_i} - \kappa \nabla^2 \eta_i. \tag{15}
\]

Consequently, by substituting appropriate values for the energy shift in Eq. 8, the phase field equation can be obtained:

\[
\frac{\partial \eta_i}{\partial t} = -L \left[ \frac{\partial f_0}{\partial \eta_i} - \kappa \nabla^2 \eta_i - P_f' \right]. \tag{16}
\]

Here \( P_f' \) is given by:

\[
P_f' = \begin{cases} 
3\eta_i(1 - \eta_i)P_f(v_{\eta_i}) & |f_i'| > |3\eta_i(1 - \eta_i)P_f| \\
\frac{3\eta_i(1 - \eta_i)P_f(v_{\eta_i})}{f_i'} & |f_i'| \leq |3\eta_i(1 - \eta_i)P_f| 
\end{cases}
\tag{17}
\]

such that the boundary becomes immobile when the drag pressure is larger than the driving pressure due to its curvature.

Eq. 16 is applicable for any general form of grain boundary drag. In the special case of pinning by a stationary particle distribution, the pinning pressure is a constant opposing the motion of the interface. Consequently, considering a constant and positive value for the pinning pressure, \( P_z \), the friction pressure in Eq. 17 (for \( P_d > P_f \)) is written as:

\[
P_f = -\text{sgn}(v_{\eta_i})P_z. \tag{18}
\]

According to Eq. 6, \( f_i' \) and \( d\eta_i/dt \) have opposite signs. Therefore, the drag pressure term (Eq. 17) in the phase field equation simplifies to:

\[
P_f' = \begin{cases} 
3\eta_i(1 - \eta_i)\text{sgn}(f_i')P_z & |f_i'| > |3\eta_i(1 - \eta_i)P_f| \\
\frac{3\eta_i(1 - \eta_i)\text{sgn}(f_i')P_z}{f_i'} & |f_i'| \leq |3\eta_i(1 - \eta_i)P_f| 
\end{cases}
\tag{19}
\]

In another spacial case, i.e. sole solute drag, the drag pressure, \( P_f = -P_s \) is never larger than the driving pressure. In other words, the grain boundary never stops unless the driving pressure goes to zero. Since the case of Fig. 2b never occurs, the resulting drag pressure term is given by:

\[
P_f' = -3\eta_i(1 - \eta_i)P_s(v_{\eta_i}). \tag{20}
\]

Accurate solution of Eq. 16 with the drag term given by Eq. 20 requires an iterative process since velocity is also a function of \( \partial \eta_i/\partial t \). A typical algorithm is, at a time step \( t_n \), to first calculate \( v_{\eta_i} \) from the previous value of \( \partial \eta_i/\partial t \) at a grid point. Then solve the PDE for the new value of \( \partial \eta_i/\partial t \). Repeat the process until convergence of \( v_{\eta_i} \) is obtained. Using a sufficient number of grid points through the interface (\( l/\Delta x > 8 \)), convergence is attained within 4 to 5 iterations. Numerically, the velocity field can be obtained only at the interface where \( d\eta_i/dn_i \) is not zero.
3. Numerical Testing and Benchmarking

A simple geometry of a circular grain shrinking inside a larger grain is considered to test the accuracy of the proposed phase field model. The phase field equation (Eq. 16) is solved with the drag pressure term given by Eq. 19 and Eq. 20, respectively. Grid spacing of $\Delta x = 1$, time step of $\Delta t = 0.5$, $m = 2$ and $\kappa = 4$ are used with the forward Euler finite difference method. Only two order parameters are considered. The initial condition is created by setting $\eta_1$ to the value of 1 inside a circle with a prescribed radius and the value of 0 outside. In the entire domain $\eta_2$ is set to $1 - \eta_1$. The phase field equation is solved with no drag pressure for 300 time steps in order to reach the steady state profile shape for the order parameter across the grain boundary.

The resulting circular grain is set to shrink with the drag pressure. In this case, driving pressure on the interface ($P_d$) is only due to the grain boundary curvature. For a circular grain, $P_d = \sigma/r$, where $r$ is the radius of the circle. Since the grain always remains a circle, $r$ is calculated from the area of the domain under $\eta_1$. Interface energy and mobility are given by Eqs. 9 and 10. The driving pressure increases monotonically as the circle shrinks. Thus, the grain boundary velocity can be obtained for low to high driving pressures in one simulation. The velocity of the grain boundary is calculated from the radius of the circle as a function of time. The velocity is then compared to the corresponding analytical expressions for shrinking of a circular grain. Based on the sharp interface assumption, the following relationship is expected to be observed in the model.

\[ v = \begin{cases} 
M(P_d - P_f) & P_d > P_f \\
0 & P_d < P_f 
\end{cases} \]

(21)

For the first case of “Zener pinning”, a constant pinning pressure is considered. Eq. 16 is solved for the constant drag pressure $P_z$ in the range of 0 to 0.0354. In Fig. 3 the velocity of the interface is plotted against the driving pressure. Each line is for a simulation with different $P_z$. The solid line with slope of 1 is for a shrinking circle with no drag pressure. As the pinning pressure increases, the curves shift towards higher driving pressures but its slope remains unchanged. The velocity of an interface with driving pressure below $P_z$ is zero as implemented by Eq. 17. These phase field simulations reproduce the results according to Eq. 21.

For the second case we consider a velocity dependent drag pressure caused by solute drag. Cahn [6] proposed the following relationship for the drag pressure:

\[ P_s = \frac{av}{1 + bv^2}. \]

(22)

Here $v$ is the velocity of the grain boundary, $a$ and $b$ are constants. Eq. 16 is solved for the shrinking circular grain using the drag pressure of Eq. 22 in Eq. 20. The calculations were performed with $a = 1$ and $b = 200$ to 50,000. The calculated velocity of the grain boundary is plotted in Fig. 4 as a function of the driving pressure and
compared with the analytical solutions. The analytical solution for a grain boundary velocity as a function of driving pressure can be obtained by inserting Eq. 22 into Eq. 21 (with \( P_f = P_s \)) and solving for \( v \) with respect to \( P_d \).

The parameter \( a \) determines the slope of the low velocity regime and \( b \) determines the driving pressure where the transition from the low to the high velocity regime occurs. For all calculations there is good agreement between the solution of the phase field equation and the corresponding analytical solution. These two examples demonstrate that the phase field equations satisfy the presumed sharp interface limit assumptions.

At grain junctions more than two order parameters have a nonzero value. Application of Eq. 16 at junctions distributes the drag pressure to the existing order parameters however, since \( \sum \eta_i \neq 1 \) [26], the combined drag force is slightly higher than the expected value. The effect of this error on the overall grain boundary movement is small if the diameter of grains are much larger in comparison to the interface thickness and area under the junctions. To test this effect, the shrinkage of a four sided grain was simulated with symmetric boundary condition (Fig. 5) using Eq. 19. The velocity of the shrinking grain’s interface was compared to a corresponding sharp interface assumption in Eq. 21. There is less than 5% error in the observed value of \( P_f \) and less than 1% for \( M \) for grains with diameter as small as 5 times the interface thickness.

4. Grain Growth Simulations

4.1. Simulation Methodology

Grain growth under the influence of particle pinning and solute drag is simulated in two dimensions by solving Eq. 16 with the drag pressure terms given by Eq. 19 and 20, respectively. To facilitate the simulation of a system which contains thousands of grains we implement a numerical procedure known as a sparse data structure [28] which reduces the number of equations locally. The sparse data structure stores only 5 order parameters and their corresponding index at each grid point in the two dimensional domain. The PDEs are solved by a finite difference method with the 9 point stencil [29] and forward Euler scheme. At each grid point, a search function surveys all the 8 neighbouring grids to find the index \( i \) corresponding to the \( \eta_i \) and calculates the Laplacian term in the PDEs. Then the value of \( \eta_i \) for the next time step is calculated. At the end of each time step only a list of active order parameters (\( \eta_i > 10^{-8} \)) at each grid point is stored for the next time step. Numerical parameters are \( \Delta x = 1 \) and \( \Delta t = 0.1 \). The system size of either 2000 \( \times \) 2000 or 2400 \( \times \) 2400 grid points are used depending on the grain size to ensure the presence of a statistically significant number of grains.

The initial grain structure is created by setting 0.5% of the grid points in the sparse data structure with the value of 1, each associated with an order parameter \( \eta_i \). These seeds are positioned randomly in a spatially uniform distribution. The remainder of unselected grid points are initialized to 0. The seeds grow due to the higher energy
of the matrix and subsequently impinge on each other creating a fine grain structure. Subsequently, a self-similar grain size distribution is attained by evolving the grain structure in the absence of a drag pressure. This grain structure, which contains approximately 6400 grains (for the $2000 \times 2000$ domain) is used as initial condition for simulations with the presence of pinning or solute drag pressure.

4.2. Grain Growth with Pinning Pressure

Using the above initial structure, we consider the effect of a constant pinning pressure on the final grain size. Grain growth is simulated using Eq. 19 with the pinning pressures in the range of $P_z = 0.01$ to $0.05$ and $m = 2$, $\kappa = 3$ and $L = 1$. In all simulations the applied pinning pressure is selected to be lower than the initial driving pressure allowing grain growth to occur. Fig. 6 shows two-dimensional grain structures for different times in a simulation with pinning pressure of $P_z = 0.02$. At the beginning of the simulation grain growth occurs in all regions of the domain. As the simulation progresses, some grain boundary segments with lower curvature and stable topological neighbourhood (grains with 6 sides) freeze. An example for such regions is shown with the box in Figs. 6b and 6c. The fraction of the frozen regions increases with time and eventually results in a total halt in the grain boundary motion. The final structure consists of grains with different sizes and topological classes. The frozen grain boundary segments can be curved or flat as shown in Fig. 6c with arrows I and II respectively.

To characterize the grain size, the area of each grain is measured and the radius of a circle with the same area is considered as the equivalent grain radius. The average grain radius increases with time but due to the pinning pressure reaches a limiting size associated with the final frozen structure. The average grain radius vs time is plotted in Fig. 7a for different pinning pressures. The limiting grain size increases as the pinning pressure decreases. The limiting grain size, $R_{lim}$, obtained in the phase field simulations, is inversely proportional to the pinning pressure $P_z$, as shown in Fig. 7b.

The balance between the pinning and driving pressure determines the limiting grain size. The local driving pressure on a curved grain boundary segment is $P_d = \sigma K$. Where $K$ is defined as the sum of the two principal curvatures of the surface ($K = K1 + K2$). In two dimensions, one principal curvature is zero (i.e. $K = K1$) and $K$ can be determined for each boundary segment as described in Appendix B. For a grain structure, the overall driving pressure of grain growth is an average quantity, $\bar{P}_d$, that is defined over all grain boundary segments. The average has to be obtained with respect to the segments area (or length in two dimensions), $ds$ such that:

$$\bar{P}_d = \frac{\int_S \sigma K ds}{\int_S ds} = \sigma \bar{K}. \quad (23)$$

The integral is over the entire grain boundary network ($S$). Phenomenologically, the driving pressure for grain growth is expressed in terms of the grain size, i.e.
\[
\bar{P}_d = \frac{\alpha \sigma}{\bar{R}},
\]
(24)
where \(\bar{R}\) is the average equivalent grain radius. For the limiting grain structure, the geometrical constant \(\alpha\) is obtained from Eqs. 23 and 24 to be:

\[
\alpha = \bar{K}_{\text{lim}} \bar{R}_{\text{lim}}.
\]
(25)

Mean field theories of Zener pinning suggest that the limiting grain size occurs when the pinning pressure is equal to the average driving pressure, i.e. \(\bar{P}_d = P_z = \alpha \sigma / \bar{R}_{\text{lim}}\) [30]. This relationship however, seems not to be accurate since the motion of the entire grain boundary network stops when the most curved segment in the system, with the maximum value of \(P_d\), reaches the \(P_z\) value. To test this hypothesis, the distribution of grain boundary segment curvature is determined. Fig. 8 shows a histogram of the grain boundary curvature at the limiting structure for the simulation with \(P_z = 0.02\). Curvature positions for values of \(\bar{K} = \bar{P}_d / \sigma\) and \(P_z / \sigma\) are shown by solid lines in the histogram.

From Fig. 8 it is evident that for the limiting grain structure, \(\max(P_{d,\text{lim}}) = P_z\) and \(P_d < P_z\). Therefore, to quantify the relationship between driving and pinning pressures, a scaling factor, \(\beta\), can be introduced as:

\[
\bar{P}_{d,\text{lim}} = \beta P_z.
\]
(26)

For the frozen grain structure, Eqs. 24 and 26 are combined to obtain the limiting grain size as:

\[
\bar{R}_{\text{lim}} = \frac{\alpha \sigma}{\beta P_z}.
\]
(27)

Here, \(\alpha\) is numerically calculated for the frozen structure at the limiting grain size \(\bar{R}_{\text{lim}}\). To obtain \(\alpha\), first the grain boundary curvature at each point is calculated. Then, the average curvature of the grain boundary network \(\bar{K}_{\text{lim}}\) is obtained by a discretized form of the integrals in Eq. 23. The value of \(\alpha\) remains constant for simulations with different \(P_z\) and is obtained to be \(\alpha = 0.26 \pm 0.05\). The constant \(\beta\) is calculated using Eq. 26. For different simulations, the value of \(\beta = 0.34 \pm 0.02\) is obtained by averaging the driving pressure from the grain boundary curvature distributions (Fig. 8).

The ratio of \(\alpha / \beta\) is obtained from the slope of the line in Fig. 7b to be 0.73. On the other hand, based on Eqs. 25 and 26 \(\alpha / \beta = 0.76 \pm 0.16\) which confirms Eq. 27 and demonstrates good agreement between the theoretical analysis and the simulation results.

As it is demonstrated above, the geometrical constants \(\alpha\) and \(\beta\) can be calculated from the grain boundary curvature distributions. Statistically, \(\alpha\) and \(\beta\) are independent. \(\alpha\) is a statistical measure that correlates curvature of the grain boundary segments to the size of the grains. \(\beta\) on the other hand is a measure of the width of the grain boundary curvatures distribution for the limiting structure. Neighbouring large grains with six sides have very flat segments (Fig. 6) thus reducing the overall average driving
pressure while configuration of small four sided grains near a large grain can create the highest curvature equal to \( P_z/\sigma \). These particular topological arrangements control the width of the curvature distribution represented by the constant \( \beta \).

The curvature distribution in Fig. 8 has two peaks at high and low curvatures. This is consistent with the final frozen structure in Fig. 6 where two distinct regions related to the highly curved segments and almost flat segments can be observed, as marked by the arrows I and II. However, the grain size distribution, shown in Fig. 9, does not have a bimodal distribution. It is expected that the topological restrictions in two dimensions causes some grain boundary segments to become flat as it was also observed in simulations with resolved particles [16].

4.3. Grain Growth with Solute Drag

The effect of solute drag on grain growth kinetics is studied in two-dimensional simulations. For a systematic study, Cahn’s expression for solute drag pressure, Eq. 22 is used in Eq. 20 with \( a = 1 \) and \( b \) varying from 200 to \( 4 \times 10^5 \). The parameter \( b \) determines the driving pressure where the transition from low to high velocity drag occurs (Fig. 4). With lower \( b \) values, higher driving pressures are required for the transition.

The grain growth simulations are performed with \( m = 2, \kappa = 4, L = 1, \Delta x = 1 \) and \( \Delta t = 0.05 \). The square of the average grain radius is plotted against time in Fig. 10. All curves start at an initial grain size, \( R_0 \), coinciding with the ideal grain growth line. The curves deviate from the ideal grain growth line and do not show any sign of saturation. This behaviour can be described phenomenologically by introducing a time exponent \( \lambda \), lower than 0.5 for the grain growth kinetics [2], i. e.

\[
\bar{R}^{1/\lambda} - \bar{R}_0^{1/\lambda} = kt, \quad \lambda \leq 0.5
\]

where \( k \) is a kinetic parameter related to the grain boundary energy, mobility and the grain geometry. For each simulation, the value of the time exponent \( \lambda \) is obtained by fitting Eq. 28 to the simulated grain growth kinetics. The straight solid lines in Fig. 10 represent parabolic grain growth with \( \lambda = 0.5 \) for the high and low velocity regimes, respectively.

Lower \( \lambda \) values indicate deviation from parabolic grain growth. For simulations with high \( b \) (e.g. \( b = 2 \times 10^4 \)) the time exponent is close to that of ideal grain growth. However as \( b \) decreases, the deviation from parabolic grain growth increases. For simulation with \( b = 4000 \) the maximum deviation is observed with \( \lambda = 0.35 \). Lowering \( b \) values further (e.g. \( b = 200 \)), reverses the trend and the grain growth becomes parabolic again with a very low kinetic coefficient \( k \). Fig. 11 illustrates the relationship between grain growth exponent, \( \lambda \) and the solute drag parameter \( b \). The minimum value of the exponent is positioned at intermediate values of \( b \).

Comparison with Fig. 4 reveals that curves with the intermediate values of \( b \) (e.g. \( b = 4000 \)) undergo a transition from the high to the low velocity regime. This transition occurs at grain boundary curvatures in the range of \( K = 0.01 \) to 0.02. In Fig. 10,
the average grain boundary curvature at the initial grain size is $\bar{K} = 0.02$. Therefore, each grain boundary segment follows the curves in Fig. 4 from $K = P_d/\sigma = 0.02$ to the lower values. Here, curves with $b = 200$ or $b = 5 \times 10^4$ entirely remain in the low and high velocity regime respectively. We conclude that the transition from high to low solute drag regime translates into grain growth kinetics with non-parabolic behaviour. However, for very weak or strong solute drag, the majority of the boundary segments remain in the high or low velocity regime respectively, such that grain growth kinetics is parabolic. A time exponent of 0.5, associated with ideal grain growth, has been rarely observed in materials [2]. The effect of solute drag pressure on the motion of grain boundaries can justify the reduction in the time exponent as can be seen in Fig. 11.

5. Summary and Conclusions

A multi-scale approach is required to model grain boundary drag and its effect on grain growth. In this work we presented a phase field formulation to model drag pressure on grain boundaries by introducing a friction pressure. The model is specifically evaluated for particle pinning with constant Zener pressure and solute drag with a velocity dependent drag pressure as described by Cahn [6]. The intrinsic nature of the interface drag phenomenon is not taken into account, but the resulting drag pressure is an input to the model. One can choose a constant drag pressure, any velocity dependent drag pressure or even a combination of both to simulate the evolution of the grain structure.

Grain growth simulations in two dimensions under particle pinning reveals an inverse relationship between $R_{lim}$ and pinning pressure in agreement with experimental observations [5]. The geometrical relationship between the limiting grain structure and the pinning pressure is investigated by analyzing the distribution of grain boundary segment curvatures and comparing the average driving pressure in the system with the pinning pressure. It is concluded that, the limiting grain structure is attained when the maximum value in the driving pressure distribution reaches the pinning pressure rather than the average driving pressure as assumed in conventional mean field theories. The scaling factor $\beta = 0.34$ is introduced to relate the average driving pressure to the pinning pressure. As a result the limiting grain size is observed to be about three times larger than the prediction by conventional theories [30]. These grain growth parameters, obtained for two-dimensional simulations have yet to be analyzed for the case of three-dimensional grain structures. Further, it is important to study the evolution of curvature distribution during grain growth and to determine whether $\alpha$, that relates the average driving pressure to the grain size, remains constant during grain growth.

Application of the friction pressure model is presented to simulate kinetics of grain growth with solute drag. It is observed that the time exponent of the grain growth kinetics has a non monotonic relationship with the solute drag parameter $b$. At very low or high $b$, the time exponent is close to 0.5. For intermediate values of $b$ the system falls into region of transition from the high to the low velocity regimes and a non-parabolic
grain growth results in time exponents as low as 0.35. Experimental observations of the
grain growth kinetics in materials with small amount of impurities shows time exponents
of $\lambda = 0.2 - 0.4$ [2] which is consistent with the trends of the simulations. Again, these
results need to be further confirmed with three-dimensional grain growth simulations.

The proposed friction pressure approach has here been discussed for grain growth
simulations. This approach however, can readily be applied when an additional driving
pressure $\Delta G_{ij} \neq 0$ is present, e.g. for recrystallization and phase transformations. To
apply the proposed method to a specific material it will be required to find the relevant
friction pressure ($P_z$, $a$ and $b$, etc.) from independent experimental or simulation studies.
The drag pressure in this model is applied uniformly in the domain. In principle, this
is true for a limiting case where particles are very fine compared to grains and each
segment interacts with large number of particles. In addition, placing particles at grain
boundary junctions may insert different drag pressures than what is calculated using
this method. However, if the size of grains are much larger than the area under the
 triple junctions, the error is negligible on the overall grain growth kinetics.

Acknowledgement

The authors would like to acknowledge the Natural Sciences and Engineering Research
Council of Canada for providing financial support. Helpful discussions with Janin Eiken
at Access, Aachen are greatly appreciated.

Appendix A: Interpolation Function for Driving pressure

Here we demonstrate that the driving pressure function, Eq. 4 and its resulting term
(Eq. 7) for the phase field PDE satisfies the fundamental sharp interface relationship
between interface velocity and the applied driving pressure, i.e. $v = M \Delta G$. We consider
an interface between two order parameters with energy difference $\Delta G_{ij}$. An unknown
term can be assumed for the function $g'(\eta)$ that interpolates $\Delta G_{ij}$ across the interface.
Eq. 8 for two order parameters can be written as,

$$\frac{\partial \eta_i}{\partial t} = -L \left[ \frac{\partial f_0}{\partial \eta_i} - \kappa \nabla^2 \eta_i + g'(\eta_i) \Delta G_{ij} \right]. \tag{A.1}$$

For the order parameter $\eta_i$, the curvilinear coordinate system is defined with a
vector $\mathbf{n}_i$ that is normal to the iso-$\eta_i$ profile. In the curvilinear system, the Laplacian
term can be rearranged using $\nabla \eta_i = (\partial \eta_i / \partial n_i) \mathbf{n}_i$ and $\nabla^2 \eta_i = \nabla \cdot \nabla \eta_i = \nabla \cdot \left( \frac{\partial^2 \eta_i}{\partial n_i^2} + \nabla \cdot \mathbf{n}_i \frac{\partial \eta_i}{\partial n_i} \right)$. Therefore, Eq. A.1 is written as:

$$\frac{\partial \eta_i}{\partial t} = -L \left[ \frac{\partial f_0}{\partial \eta_i} - \kappa \left( \frac{\partial^2 \eta_i}{\partial n_i^2} + \nabla \cdot \mathbf{n}_i \frac{\partial \eta_i}{\partial n_i} \right) + g'(\eta_i) \Delta G_{ij} \right] \tag{A.2}$$

The weighting function $g'(\eta)$ is chosen to satisfy two conditions: (i) application of
the driving pressure ($\Delta G_{ij}$) does not change the shape of the interface profile, (ii)
relationship between the interface velocity and the driving pressure is linear \((v = M \Delta G_{ij})\). If the curvature radius of the interface is much larger than the interface thickness, the curved interface has the same order parameter profile in the curvilinear coordinate system as a flat interface. For a flat interface \(\nabla \cdot \mathbf{n}_i = 0\) and without a driving pressure \((\Delta G_{ij} = 0)\), it remains stationary i.e. \(\partial \eta_i / \partial t = 0\). Thus, the following equality can be obtained from Eq. A.2:

\[
\frac{\partial \eta_i}{\partial t} = 0 \Rightarrow \frac{\partial f_0}{\partial \eta_i} = \kappa \frac{\partial^2 \eta_i}{\partial n_i^2} \tag{A.3}
\]

Using Eq. A.3 and \(\partial \eta_i / \partial t = \partial \eta_i / \partial n_i \cdot \partial n_i / \partial t\) simplifies Eq. A.2 to the following equation:

\[
v_{\eta_i} = \frac{\partial n_i}{\partial t} = -L \left[ -\kappa \nabla \cdot \mathbf{n}_i + g'(\eta_i) \Delta G_{ij} \frac{\partial n_i}{\partial \eta_i} \right] \tag{A.4}
\]

where \(v_{\eta_i}\) is the velocity field (Eq. 13) associated with \(\eta_i\) and is equal to the interface velocity, \(v\), between grain \(i\) and \(j\). The gradient of a normal vector, \(\nabla \cdot \mathbf{n}_i\), equals to the curvature of the interface \([31]\). Moelans et al. \([26]\) showed \(L \kappa = M \sigma\). In the sharp interface limit we have:

\[
v = M (\sigma K + \Delta G_{ij}) \tag{A.5}
\]

By comparing Eqs. A.4 and A.5, \(g'\) can be obtained:

\[
g'(\eta_i) = -\frac{M}{L} \frac{\partial \eta_i}{\partial n_i} \tag{A.6}
\]

If the interface profile does not change when an additional driving pressure is applied then we can use the profile obtained for a stationary flat interface. The interface profile can be obtained by solving Eq. A.3 for two order parameters \([26]\):

\[
\eta_i = \frac{1}{2} \left[ 1 - \tanh \left( \sqrt{\frac{m}{2\kappa}} n_i \right) \right] \tag{A.7}
\]

The derivative \(\partial \eta_i / \partial n_i\) as a function of \(\eta_i\) is

\[
\frac{\partial \eta_i}{\partial n_i} = -2 \sqrt{\frac{m}{2\kappa}} (\eta_i - \eta_i^2). \tag{A.8}
\]

And according to Eq. 10:

\[
L = \frac{2}{3} \sqrt{\frac{m}{2\kappa}} M. \tag{A.9}
\]

Consequently, by substituting Eqs. A.9 and A.8 into Eq. A.6 the weighting function can be obtained, i.e.

\[
g'(\eta_i) = 3(\eta_i - \eta_i^2) = 3\eta_i(1 - \eta_i). \tag{A.10}
\]
During the derivation of $g'$, it is assumed that the applied driving pressure does not change the order parameter profile. Therefore, the profile of a moving interface, must be a solution of phase field PDE (Eq. 8). Transition of the coordinate system to a co-moving frame of reference allows to write the following equation for the moving $\eta_i$.

$$\eta_i = \frac{1}{2} \left[ 1 - \tanh \left( \sqrt{\frac{m}{2\kappa}} n_i - vt \right) \right] \tag{A.11}$$

Rewriting the velocity of the diffuse interface in the curvilinear coordinate system (Eq. A.4) results in

$$v_{\eta_i} = -L \left[ -\kappa K + 3\eta_i(1 - \eta_i) \Delta G_{ij} \frac{\partial n_i}{\partial \eta_i} \right]. \tag{A.12}$$

The sharp interface velocity (Eq. A.5) is together with Eq. 9 and Eq. A.9 substituted into Eq. A.12 to obtain the equality:

$$-\sqrt{\frac{2m}{\kappa}} \eta_i(1 - \eta_i) \frac{\partial n_i}{\partial \eta_i} = 1. \tag{A.13}$$

The profile of a moving interface given in Eq. A.11 is indeed a solution of Eq. A.13. Therefore, it satisfies the phase field equation and demonstrate that applying driving pressure with the given $g'$ does not change the shape of the order parameter profile. $g'$ can be simplified further using $\eta_i = 1 - \eta_j$ [26]. Consequently, the driving pressure term in the phase field equation (Eq. 7) is written as:

$$\frac{\partial f_d}{\partial \eta_i} = g'(\eta_i) \Delta G_{ij} = 3\eta_i \eta_j \Delta G_{ij}. \tag{A.14}$$

The contribution of the driving pressure to the energy functional, $f_d$, (Eq. 4), is obtained by integration of Eq. A.14 with the condition that $f_d(\eta_i = 1) = G_i$.

### Appendix B: Measuring Interface Curvature

Curvature of a grain boundary segment can be measured from order parameter fields in the phase field simulations. The following relationship is used to measure curvature at each point on the grain boundary [24]:

$$\nabla^2 \eta_i = \frac{\partial^2 \eta_i}{\partial n_i^2} + \frac{\partial \eta_i}{\partial n_i} K \tag{B.1}$$

Applying Eq. B.1 on a curved interface between $\eta_i$ and $\eta_j$ results in a $K$ which is positive for the grain with a convex segment and negative for the grain with a concave segment. The local value of curvature is obtained for each order parameter only for $0.2 < \eta_i < 0.8$ resulting in a uniform sampling of curvature on all grain boundary segments. Statistical sampling is done by applying Eq. B.1 to all order parameters. The redundant negative curvature values are eliminated. The representative curvature of a segment is determined by averaging grid points in a box at the centre of the segment.
The size of the box is 0.2 times the segment length in order to avoid sampling points close to the triple junctions.

References


Figure 1. Local energy density, $f_{\text{loc}}$ as a function of order parameters in a system with two grains. The solid line is energy density associated with the equilibrium value of order parameters across the interface for $G_1 = 0.3$ and $G_2 = 0$.

Figure 2. Schematic representation of the friction pressure on an interface. Local energy density of two neighbouring grains in (a) and (c) is shifted in order to create an opposing pressure to the interface movement. (b) When the driving pressure is smaller than the pinning pressure, the interface stops.
Figure 3. Grain boundary velocity of a shrinking circular grain as a function of driving pressure as obtained in phase field simulations with pinning pressure.

Figure 4. Grain boundary velocity of a shrinking circular grain as a function of driving pressure in the presence of solute drag pressure. Simulated curves (dot markers) are compared with the analytical solution (solid lines) for different values of $b$. 
Figure 5. A four sided grain shrinks in a symmetric boundary condition from equivalent radius of (a) 37 grid points to (b) 13 grid points. The interface thickness is 5 grid points approximately.

Figure 6. Snapshots of grain growth simulation with $\sigma = 1.1547$ and $M = 2.598$ at times of (a) 200, initial structure, (b) 2000 and (c) 20000, final frozen structure.
Figure 7. (a) Equivalent grain radius as a function of time for grain growth simulations with different pinning pressures. (b) Limiting grain size as a function of inverse pinning pressure.
**Figure 8.** Distribution of local grain boundary curvature ($K = P_d/\sigma$) for the limiting grain structure with $P_z = 0.02$. Vertical lines represent the curvature values corresponding to the value of the mean driving pressure and pinning pressure.

**Figure 9.** Grain size distribution for the limiting grain structure with $P_z = 0.02$. 
Figure 10. Kinetics of grain growth with the presence of solute drag pressure for simulations with different $b$ values in Eq. 22. Solid lines represent parabolic grain growth.

Figure 11. Time exponent coefficient of grain growth ($\lambda$ in Eq. 28) as a function of solute drag parameter $b$ in Eq. 22.