Phase-field simulations of grain growth in materials containing second-phase particles

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28 February 2008
Part I

Scientific context
Outline part I: Scientific context

- Grain growth in polycrystalline materials
- Pinning effect of second-phase particles
- Phase-field method for simulating microstructural evolution
- Incentives of the research
Role of microstructures in materials science

Chemical composition
+ Temperature, pressure, cooling rate,…

Microstructure
Shape, size and orientation of the grains, mutual distribution of the phases

Material properties
Strength, deformability, hardness, toughness, fatigue…
Polycrystalline microstructure with second-phase particles

- Mechanism for controlling the grain size of a material
  - E.g. microalloyed steels
    - Small grain size required for high strength
    - Addition of small amounts of Nb, Ti, Al, V,...
    - Formation of NbC, AlN, TiN,...
    - Pinning of grain boundaries during heat treatments or welding
Normal grain growth

- Surface tension

\[ P_g = \text{driving pressure for grain boundary movement} \]

\[ P_g = \frac{\alpha \sigma_{gb}}{R} \]

\[ \Rightarrow P_g \text{ decreases in time} \]
Normal grain growth

- Surface tension + topological considerations

Isotropic:
\[ \alpha_1 = \alpha_2 = \alpha_3 = 120^\circ \]

Smaller grains shrink and larger grains grow
Zener pinning

- Grain boundary area is reduced when a particle is located on a grain boundary
  - Particles exert a back force on moving grain boundaries

\[ F_{Z}^{\text{max}} = \pi r \sigma_{gb} \]

- Dimple-shape

**MnS precipitate in low-C steel**
Final grain size – Zener relation

• Calculation of the total pinning pressure of the particles $P_z$ requires
  • Number of particles in contact with a grain boundary
  • Angle at which grain boundary meets the particle

• Grain growth stops when $P_g = P_z$

$$\frac{R_{lim}}{r} = K \frac{1}{f_V^b}$$

From P.A. Manohar (1998)
Pinning effect: experimental observation

- Fe-0.09 to 0.53 w% C-0.02 w% P containing Ce$_2$O$_3$ inclusions
  - PhD – work M. Guo
  - Pinned austenite grain boundaries
Phase-field simulations of microstructural evolution

- **Experiments, atomistic simulations and thermodynamic models**
  - Crystal structure, phase stabilities, interfacial properties (energy, mobility, structure, anisotropy), diffusion properties

- **Phase-field simulations**
  - Morphological evolution of the grains at the mesoscale during solidification, precipitation, solid-state phase transformations, grain growth, …

- **Models that predict macroscopic material properties**
  - Strength, deformability, hardness, toughness, fatigue, …
Phase field method

- van der Waals, Cahn-Hilliard (1958), Ginzburg-Landau (1950), Hohenberg and Halperin (1977)

- Microstructure evolution (started ± 20 years ago)
  - Solidification
  - Ordering reactions
  - Martensitic transformation

- Nowadays
  - Wide range of applicabilities
  - Quantitative aspects
    - Parameter determination
    - Numerical implementation
Representation of microstructures in the phase-field method

- Phase-field variables: continuous functions in space and time
  - Local composition \( x_B(r,t) \)
  - Local structure and orientation \( \eta(r,t) \)

Binary alloy A-B
- Phase \( \alpha \): \( \eta = 0 \)
- Phase \( \beta \): \( \eta = 1 \)
Diffuse-interface description

- **Sharp interface**
  - Discontinuous variation in properties
  - Requires tracking of the interfaces
  - Simplified grain morphologies

- **Diffuse interface**
  - Continuous variation in properties
  - Interfaces implicitly given by local variations in phase-field variables
  - Complex grain morphologies
Phase-field simulation technique

- **Microstructural representation:**
  \[ x_B(r,t) \]
  \[ \eta(r,t) \]

- **Thermodynamic and kinetic equations**
  - Phase stabilities
  - Interfaces
  - Elastic energy due to volume effects
  - Orientation dependence
  - Solute diffusion

- **Parameter determination**

- **Numerical solution**
Intermediate conclusions
Incentives of the research

- Pinning effect of second-phase particles on grain boundaries and final grain size still not understood

- Mesoscale grain growth simulations can give important insights

- Phase-field method for simulating microstructure evolution
  - General technique based on nonequilibrium thermodynamic principles
  - Complex phenomena and morphologies

⇒ Phase-field simulations of grain growth in materials containing second-phase particles
Part II

Research work

Model description and simulation results
Outline part II: Results

- Phase-field model for grain growth and Zener pinning
- Study of the pinning mechanism
- Simulation results for polycrystalline materials
- Ongoing/future research
Representation of a polycrystalline structure

- Extension grain growth model
  D. Fan and L.-Q. Chen (1997)

- Phase field variables:
  \[ \eta_1, \eta_2, ..., \eta_i(r,t), ..., \eta_p \]

- Particles: \( \Phi = 1 \)
  \[ (\eta_1, \eta_2, ..., \eta_i, ..., \eta_p) = (0, 0, ..., 0, ..., 0) \]

- Grain i of matrix-phase: \( \Phi = 0 \)
  \[ (\eta_1, \eta_2, ..., \eta_i, ..., \eta_p) = (0, 0, ..., \pm 1, ..., 0) \]
Representation of the grain boundaries

Grain $i$: $\eta_i = 0$

Grain $j$: $\eta_j = 0$

Distance (g.p.)
Representation of a particle

- Evolution of $\eta_i$ across a particle in grain $i$
Free energy and kinetic equations

- **Thermodynamic free energy**

  \[ F = \int_V \left[ m \left( \sum_{i=1}^{p} \left( \frac{\eta_i^4}{4} - \frac{\eta_i^2}{2} \right) + \sum_{i=1}^{p} \sum_{j \neq i}^{p} \eta_i^2 \eta_j^2 + \varepsilon \Phi \sum_{i=1}^{p} \eta_i^2 \right) + \sum_{i=1}^{p} \frac{\kappa}{2} (\nabla \eta_i)^2 \right] dV \]

- **Equilibrium**
  - \( \Phi = 0 \):
    \[ (\eta_1, \eta_2, ..., \eta_p) = (1, 0, ..., 0), (0, 1, ..., 0), ..., (0, 0, ..., 1), (-1, 0, ..., 0), ... \]
  - \( \Phi = 1 \):
    \[ (\eta_1, \eta_2, ..., \eta_p) = (0, 0, ..., 0) \]

- **Kinetic equations (Ginzburg-Landau)**

  \[ \frac{\partial \eta_i(\vec{r}, t)}{\partial t} = -L \frac{\partial F}{\partial \eta_i(\vec{r}, t)} = -L \left( \frac{\partial f_0(\eta_1, \eta_2, ...)}{\partial \eta_i(\vec{r}, t)} - \kappa \nabla^2 \eta_i(\vec{r}, t) \right) \]
\[
\frac{\partial \eta_i(r,t)}{\partial t} = -L \left( \frac{\partial f(\eta_1, \eta_2, \ldots)}{\partial \eta_i(r,t)} - \kappa \nabla^2 \eta_i(r,t) \right)
\]

\[
f = m \left( \sum_{i=1}^{p} \left( \frac{\eta_i^4}{4} - \frac{\eta_i^2}{2} \right) + \sum_{i=1}^{p} \sum_{j \neq i} \eta_i^2 \eta_j^2 + \varepsilon \Phi \sum_{i=1}^{p} \eta_i^2 \right)
\]

- **Grain boundary energy:** \(0.58 \sqrt{\kappa m}\)
- **Grain boundary velocity:** \(V = \kappa L (\lambda_1 + \lambda_2)\)
- **Interfacial energy particles:** \(f(\varepsilon) \sqrt{\kappa m}\)
- **Parameter choice:** \(\kappa = 0.5, m = 1, L = 1, \varepsilon = 1\)
Local interaction: spherical grain

- Grain boundary passing a particle
  - 3-D simulation
  - Geometry

- Dimple-shape
- Break-free at $\beta > \pi/4$
Temporal evolution spherical grain

\[ r = 8 \]
\[ R_0 = 90 \]
\[ d_{c-p} = 76 \]
Large-scale 2-D simulations

- Isotropic grain boundary properties
- Random dispersion of round particles
  \[ r = 2.5 \qquad r = 3 \]
- Area fraction: \( f_a = 0.004 - 0.16 \)
- Initial microstructure:
  - Grain nucleation in presence of particles \( (R_0=0) \)
  - Grain nucleation and initial grain growth without particles \( (R_0>0) \)
  \[ \frac{R_{\text{lim}}}{r} = K \frac{1}{f_a^{b}} \]
Large-scale 2D-simulations: initial microstructure

- Grain nucleation in the presence of particles
  - $\overline{R_0} = 0$
- Most particles on grain boundaries

$r = 3, f_a = 0.02$
Large-scale 2D-simulations: initial microstructure

- Grain nucleation and initial growth without particles
Large-scale 2D-simulations: initial microstructure

- Grain nucleation and initial growth without particles
- Addition of particle when $\bar{R}_0 > 0$
- Many particles within grains

$r = 3, f_a = 0.02, \bar{R}_0 = 13.6$
Large-scale 2-D simulations

- $R_0 = 0$
  - $r = 3, f_a = 0.04, \bar{R}_0 = 0, \bar{R}_{\text{lim}} = 22.2$

- $R_0 > 0$
  - $r = 3, f_a = 0.04, \bar{R}_0 = 13.6, \bar{R}_{\text{lim}} = 26.2$
Role initial grain size

- $R_0$ important for high $f_a$

Fraction of particles on grain boundaries
Experimental study
- H.P. Longworth and C.V. Thompson
- Al films with CuAl₂ precipitates
- Annealing at 500°C:
  - Grain growth → pinning
  → abnormal grain growth
- Huge grain size required for micro-electronic applications
- Grain growth in thin films usually treated in 2-D
3-D simulations for thin films

• 2-D columnar grain structure

• 3-D interaction particle-grain boundary
  • $\Rightarrow$ curvature out of the plane

• Film thickness

• Particles in the middle of the film are more effective

$r = 3, f_a = 0.12, l_3 = 21$
Comparison with experimental data

\[
\frac{R_{\text{lim}}}{r} = 1.28 \frac{1}{f_a^{0.5}}
\]

- \( R_0 = 0, r = 2.5 \)
- \( R_0 = 0, r = 3 \)
- Fitted curve for \( R_0 = 0 \)
- \( R_0 \approx 14.4, r = 3 \)
- \( R_0 \approx 28.2, r = 3 \)
- \( R_0 > 0, r = 2.5 \)
- 3-D film, \( r = 3, l_3 = 21 \)
- Al film (exp data)
Comparison with experimental data

Al film: hot-stage TEM micrograph
(H.P. Longworth and C.V. Thompson, 1991)

\[ f_a = 0.086 \]

3-D simulation
\[ f_a = 0.08 \]

2-D simulation
\[ f_a = 0.08 \]

• Other effects
  - Surface grooving
  - Semi-coherent particle-matrix interface
  - Evolution particles
Ongoing /future research

- 3-D simulations for bulk materials
  - Bounding box algorithm

- Evolution, interfacial properties and shape of the particles

- Different grain boundary energies
  - Misorientation and inclination dependence

- Thermal grooving
Arenberg Castle, Leuven, Belgium
• Thank you for your attention!

• Acknowledgment:
  • Nele Moelans is Postdoctoral Fellow of the Research Foundation - Flanders (FWO-Vlaanderen)
  • Simulations were performed on the HP-computing infrastructure of the K.U.Leuven (operational since May 2005)

• More information on http://nele.studentenweb.org
Ongoing/future research

• 3D simulations for bulk materials
  • Bounding box algorithm
    – Equations for $\eta_i$ are only solved locally for grain $i$

Limiting grain size

$$\frac{\bar{R}_{\text{lim}}}{r} = K \frac{1}{f_Y^m}$$
Ongoing /future research

- Evolving particles
  - Coupling with Cahn-Hilliard equations for $\Phi$

Grain structure: $f_v=0.12, L=10M$  
Distribution $\Phi$
Ongoing/future research

• Anisotropic grain boundary properties

Misorientation dependence of grain boundary energy
Ongoing/future research

• Thermal grooving

<table>
<thead>
<tr>
<th>Environment</th>
<th>( \psi = 1 )</th>
<th>( \eta_1 = 0 )</th>
<th>( \eta_2 = 0 )</th>
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</thead>
<tbody>
<tr>
<td>Film</td>
<td>( \psi = 0 )</td>
<td>( \eta_1 = 1 )</td>
<td>( \eta_2 = 0 )</td>
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</tbody>
</table>
Interaction energy

- Energetic consideration
  - Geometry
  - Theoretical interaction energy
    \[ 2r\sigma_{gb}(2D) \]
    \[ \pi r^2 \sigma_{gb}(3D) \]

- Diffuse grain boundaries ⇒
  - Interaction energy slightly too negative
  - Lower limit on particle size
Interaction energy
Comparison with theory

- High scatter for low $f_a$
- Fitting:
  \[ \frac{R_{\text{lim}}}{r} = \frac{b}{f_V^\beta} \]
  - Theory: $\beta = 0.5$
  - Phase field ($R_0=0$):
    \[ \beta = 0.48, b = 1.32 \]
  - Monte Carlo:
    \[ \beta = 0.5, b = 1.7 \]
    \[ \beta = 0.54, b = 1.2 \]
  - Front-tracking: $\beta = 0.46$
    \[ \beta = 0.5 \]
Computational considerations

- **2D:** $R_{lim}/r$ could be reproduced
  - **High $f_a$:**
    - system size 256, 20000 time steps
    - $\Rightarrow$ 10 hours
  - **Low $f_a$:**
    - system size 512, >60000 time steps
    - $\Rightarrow$ 10 days

- **3D:**
  - $R_{lim}/r$: x10 $\Rightarrow$ system size: x10
  - Third power of system size
  - Computer requirements: $x10^5$
Pinning mechanism

- Zener

- Rios

- Coherent/incoherent particles-matrix interface