Phase-field model for solid state transformation in a two-phase binary system with elastic inhomogeneity

A. Durga[*], Nele Moelans[†], Patrick Wollants[‡]

Department of Metallurgy and Materials Engineering, Katholieke Universiteit Leuven,

Kasteelpark Arenberg 44, B-3001 Leuven, Belgium

Abstract

A phase-field model is presented to study binary systems having two phases with different elastic properties. The evolution of two phase-field variables, namely, composition and non-conserved phase-field, are considered. This model uses the approach of Steinbach and Apel [1] to model the dependence of total strain on the phase-field, which assumes equal elastic stresses in the different phases at the diffuse interface. The elasticity equations for mechanical equilibrium are solved by a Fourier transform-based iterative approach similar to Hu and Chen [2]. The phase-field equations are solved by the finite difference method. Growth of a second-phase particle in an elastic matrix and the dependence of the precipitate shape on the elastic properties of the two phases are studied.

Keywords: Phase-field model, Elasticity, Microstructure

1 Introduction

Phase-field modelling as a tool to study microstructure evolution is becoming increasingly important with its capability to combine various phenomena like elasticity, magnetism, etc. with the conventional model. In the field of phase-field microelasticity, Khachaturyan's model [3] is widely used and there have also been attempts to combine it with plasticity models to study various phenomena like dislocation dynamics, crack propagation and so on. The focus of the present study is to examine the effect of using two different interpolations for elastic properties when elasticity effects are treated in a phase-field model. Recently, Steinbach and Apel [1] have proposed a multiphase-field model which includes elasticity and is consistent with the Kim et al. [4] model for the chemical contribution. It uses Reuss-Sachs model [5] for interpolating the stiffness tensor between the different phases, instead of the Khachaturyan's model. Hu and Chen [2] established a Fourier transform-based iterative approach for solving the elasticity equations at mechanical equilibrium to obtain the elastic strain for Khachaturyan's model. In the present study, the model formulation of Steinbach and Apel [1] is used to treat a binary two-phase system and the elasticity equation is solved by an iterative approach similar to that of Hu and Chen. This method is compared with the Khachaturyan's scheme of interpolation [5].

In Section 2, a description of the proposed model is presented, comparing it with Khachaturyan's model and a brief description of Hu-Chen's method to solve the elasticity equations is given. In Section 3, phase-field equations are introduced. Finally, in Sections 4 and 5, results are presented for a precipitate growing in an elastic matrix for two different interpolations of the stiffness tensor and for different elastic properties.

2 Model formulation

Two components A and B forming two possible phases, α and β with different elastic properties are considered in this study. Two phase-field variables, which are functions of space and time are used: the conserved molar fraction field, c (molar fraction of A), and the non-conserved phase-field, ϕ. A value of ϕ = 1 indicates the presence of only α phase and ϕ = 0, only β phase. Any intermediate value refers to the interface region between the two phases, which is considered as a mixture of the two phases.

The total free energy F of the system is defined as:

$$F = \int f^{gb} + f^{ch} + f^{el} dV \tag{1}$$

where f^{gb} refers to grain boundary energy density, f^{ch} is the chemical free energy density and f^{el}, elastic energy density.

2.1 Grain boundary energy density

The grain boundary energy density is given by:

$$f^{gb} = \frac{\kappa}{2}(\vec{\nabla}\phi)^2 + Wg(\phi) \tag{2}$$

$$g(\phi) = \phi^2(1-\phi)^2 \tag{3}$$

where κ is the gradient energy coefficient, $g(\phi)$ is the double-well function and W, depth of the double-well. The two terms contribute only in the interface region. κ and W are adjustable parameters, related to the interfacial energy and the interfacial thickness [6].

2.2 Chemical free energy density

The chemical free energy density is interpolated between the two phases using a smooth function $p(\phi)$ and is given by:

---

[*] Corresponding author: durga.ananthanarayanan@mtm.kuleuven.be, Tel: +32 16 321781, Fax: +32 16 321991

[†] nele.moelans@mtm.kuleuven.be

[‡] patrick.wollants@mtm.kuleuven.be

$$f^{ch} = p(\phi) f^{ch}_{\alpha} + [1-p(\phi)] f^{ch}_{\beta} \tag{4}$$

$$\text{where } p(\phi) = \phi^3(6\phi^2 - 15\phi + 10) \tag{5}$$

$p(\phi)$ equals 0 at ϕ=0 and equals 1 at ϕ=1. The chemical energy densities of the α and β phases are assumed to have a parabolic shape: $f^{ch}_{\alpha} = A^{\alpha}/2(c_{\alpha} - c_{\alpha,0})^2$, $f^{ch}_{\beta} = A^{\beta}/2(c_{\beta} - c_{\beta,0})^2$, where A^{α}, A^{β}, $c_{\alpha,0}$ and $c_{\beta,0}$ are constants. According to Kim et al.[4], every point in the interface is assumed to be a mixture of the two phases with compositions c_{α} and c_{β} (which are not necessarily equal to the compositions of the bulk α and β phases) at equal chemical potential. This is a thermodynamically consistent description of the interface region.

$$\text{Therefore, } \mu_{\alpha} = \mu_{\beta} \tag{6}$$

$$\text{i.e., } \frac{\partial f^{ch}_{\alpha}}{\partial c_{\alpha}} = \frac{\partial f^{ch}_{\beta}}{\partial c_{\beta}} \tag{7}$$

$$\Rightarrow A^{\alpha}(c_{\alpha} - c_{\alpha,0}) = A^{\beta}(c_{\beta} - c_{\beta,0}) \tag{8}$$

The composition c is related to the phase concentrations c_{α} and c_{β} as:

$$c = p(\phi) c_{\alpha} + [1-p(\phi)] c_{\beta} \tag{9}$$

This correctly reduces to the phase concentrations inside the bulk of the two phases. c_{α} and c_{β} are calculated using (8) and (9). The derivatives of the chemical free energy density with respect to c and ϕ are derived using equations (4)-(9).

2.3 Elastic energy density

The elastic stress is defined according to Hooke's law as: $\sigma_{ij} = C_{ijkl}\epsilon^{el}_{kl}$, where C_{ijkl} is the stiffness tensor and ϵ^{el}_{kl}, the elastic strain. The elastic strain is defined as the difference between the total strain, ϵ^{tot}_{kl}, and the eigenstrain (stress-free strain), ϵ^{*}_{kl}, which arises in the material due to phase transformation:

$$\epsilon^{el}_{kl} = \epsilon^{tot}_{kl} - \epsilon^{*}_{kl} \tag{10}$$

The total strain is the sum of homogeneous strain, which is a measure of the macroscopic deformation of the system, and local heterogeneous strain [7].

$$\epsilon^{tot}_{kl} = \overline{\epsilon_{kl}} + \delta\epsilon_{kl} \tag{11}$$

The heterogeneous strain is related to the displacement as:

$$\delta\epsilon_{kl} = \frac{1}{2}\left[\frac{\partial u_k(\overrightarrow{r})}{\partial r_l} + \frac{\partial u_l(\overrightarrow{r})}{\partial r_k}\right] \tag{12}$$

where $u_i(\overrightarrow{r})$ is the ith component of displacement. Two different formulations for calculating the elastic energy density are presented. In the proposed model, Steinbach and Apel's [1] approach is used. This is later compared with Khachaturyan's approach [5].

2.3.1 Steinbach and Apel's approach

According to Steinbach and Apel, the elastic energy density is interpolated between the phases, similar to the chemical energy density:

$$f^{el} = p(\phi) f^{el}_{\alpha} + [1-p(\phi)] f^{el}_{\beta} \tag{13}$$

The elastic energy densities of the two phases are given by:

$$f^{el}_{\alpha} = \frac{1}{2}\epsilon^{el,\alpha}_{ij} : C^{\alpha}_{ijkl} : \epsilon^{el,\alpha}_{kl} \tag{14}$$

$$f^{el}_{\beta} = \frac{1}{2}\epsilon^{el,\beta}_{ij} : C^{\beta}_{ijkl} : \epsilon^{el,\beta}_{kl} \tag{15}$$

where C^{α}_{ijkl} and C^{β}_{ijkl} are constants. Similar to phase compositions, phase strains $\epsilon^{el,\alpha}_{kl}$ and $\epsilon^{el,\beta}_{kl}$ are introduced.

Using the Reuss-Sachs model, the elastic stress is equal in all the phases assumed to be present as a mixture at every point in the interface. This is analogous to the assumption of equal chemical potentials.

$$\text{i.e., } \sigma_{ij} = \sigma^{\alpha}_{ij} = \sigma^{\beta}_{ij} \tag{16}$$

$$\Rightarrow \frac{\partial f^{el}}{\partial \epsilon^{el}_{kl}} = \frac{\partial f^{el}_{\alpha}}{\partial \epsilon^{el,\alpha}_{kl}} = \frac{\partial f^{el}_{\beta}}{\partial \epsilon^{el,\beta}_{kl}} \tag{17}$$

$$\epsilon^{el}_{kl} C_{ijkl} = \epsilon^{el,\alpha}_{kl} C^{\alpha}_{ijkl} = \epsilon^{el,\beta}_{kl} C^{\beta}_{ijkl} \tag{18}$$

The overall elastic strain is interpolated between the phase strains as:

$$\epsilon^{el}_{kl} = p(\phi)\epsilon^{el,\alpha}_{kl} + [1-p(\phi)]\epsilon^{el,\beta}_{kl} \tag{19}$$

Substituting from (16) and (18), we get

$$\begin{aligned} \sigma_{ij} C^{-1}_{ijkl} &= p(\phi)\sigma^{\alpha}_{ij}[C^{\alpha}_{ijkl}]^{-1} \\ &\quad + [1-p(\phi)]\sigma^{\beta}_{ij}[C^{\beta}_{ijkl}]^{-1} \end{aligned} \tag{20}$$

$$\Rightarrow C_{ijkl} = [p(\phi) S^{\alpha}_{ijkl} + \{1-p(\phi)\} S^{\beta}_{ijkl}]^{-1} \tag{21}$$

This is rewritten as

$$C_{ijkl} = C^{\alpha}_{ijkl} - C'_{ijkl}(\overrightarrow{r}) \tag{22}$$

The solution to the elasticity equations to obtain the elastic strain field is elaborated in Section 2.4. From equation (18), we obtain

$$\epsilon^{el,\beta}_{kl} = [C^{\beta}_{ijkl}]^{-1} C^{\alpha}_{ijmn} \epsilon^{el,\alpha}_{mn} \tag{23}$$

$$= M_{klmn}\epsilon^{el,\alpha}_{mn} \tag{24}$$

$$\epsilon^{el}_{kl} = p(\phi)\epsilon^{el,\alpha}_{kl} + [1-p(\phi)] M_{klmn}\epsilon^{el,\alpha}_{mn} \tag{25}$$

[Using (19) and (24)]

$\epsilon^{el,\alpha}_{kl}$ is obtained by solving equation (25) and from that, $\epsilon^{el,\beta}_{kl}$ can also be obtained using equation (24). The elastic energy density and its derivative with respect to ϕ are calculated using equations (13)-(15).

2.3.2 Khachaturyan's approach

The elastic energy density is given by:

$$f^{el} = \frac{1}{2}\epsilon^{el}_{ij} : C_{ijkl} : \epsilon^{el}_{kl} \tag{26}$$

In the Khachaturyan's approach [5], a linear mixture model is used for the stiffness tensor:

$$C_{ijkl} = p(\phi) C^{\alpha}_{ijkl} + [1-p(\phi)] C^{\beta}_{ijkl} \tag{27}$$

$$= C^{\alpha}_{ijkl} - [1-p(\phi)](C^{\alpha}_{ijkl} - C^{\beta}_{ijkl}) \tag{28}$$

$$= C^{\alpha}_{ijkl} - C'_{ijkl}(\overrightarrow{r}) \tag{29}$$

where C^{α}_{ijkl} and C^{β}_{ijkl} are constants. For the eigenstrain also, a linear mixture model is used:

$$\epsilon^{*}_{kl} = p(\phi)\epsilon^{*,\alpha}_{kl} + [1-p(\phi)]\epsilon^{*,\beta}_{kl} \tag{30}$$

The elastic energy density and its derivative with respect to ϕ are calculated using the above equations and the elastic strain defined earlier. The solution to the elasticity equations to obtain the elastic strain field is elaborated in Section 2.4.

2.4 Solution of mechanical equilibrium equations for both models

For solving the elasticity equations for the two approaches described in Sections 2.3.1 and 2.3.2 in order to obtain the elastic strain field at every time step, the approach of Hu and Chen [2] is implemented.

$$\begin{aligned}\sigma_{ij} &= [C^{\alpha}_{ijkl} - C'_{ijkl}(\overrightarrow{r})] \\ &\quad [\overline{\epsilon_{kl}} + \frac{1}{2}\left(\frac{\partial u_k(\overrightarrow{r})}{\partial r_l} + \frac{\partial u_l(\overrightarrow{r})}{\partial r_k}\right) \\ &\quad -p(\phi)\epsilon^{*,\alpha}_{kl} - (1-p(\phi))\epsilon^{*,\beta}_{kl}] \qquad (31) \\ &\quad \text{[From Section 2.3]}\end{aligned}$$

Since the mechanical equilibrium is attained much faster than the chemical equilibrium because of the slow diffusion processes, it is assumed that the system is always at mechanical equilibrium, i.e., $\frac{\partial \sigma_{ij}}{\partial r_j} = 0$. The mechanical equilibrium equation cannot be solved analytically because of the non-linearity. If the inhomogeneity in elasticity that arises from the term with C'_{ijkl} is ignored, i.e. $C'_{ijkl} = 0$, then the equation becomes

$$\begin{aligned}&\frac{1}{2}C^{\alpha}_{ijkl}\left(\frac{\partial^2 u^0_k(\overrightarrow{r})}{\partial r_j \partial r_l} + \frac{\partial^2 u^0_l(\overrightarrow{r})}{\partial r_j \partial r_k}\right) \\ &= C^{\alpha}_{ijkl}\frac{\partial p(\phi)}{\partial r_j}(\epsilon^{*,\alpha}_{kl} - \epsilon^{*,\beta}_{kl}) \qquad (32)\end{aligned}$$

This can be solved in the Fourier space. The zeroth-order approximation of the displacement field in the real space in 2-D can be obtained by taking the back Fourier transform of the displacement field in the Fourier space. Including the terms with C'_{ijkl} and retaining the same left hand side as equation (32), the mechanical equilibrium equation becomes

$$\begin{aligned}&\frac{1}{2}C^{\alpha}_{ijkl}\left(\frac{\partial^2 u^n_k(\overrightarrow{r})}{\partial r_j \partial r_l} + \frac{\partial^2 u^n_l(\overrightarrow{r})}{\partial r_j \partial r_k}\right) \\ &= (\epsilon^{*,\alpha}_{kl} - \epsilon^{*,\beta}_{kl})C^{\alpha}_{ijkl}\frac{\partial p(\phi)}{\partial r_j} \\ &+(\overline{\epsilon_{kl}} - \epsilon^{*,\beta}_{kl})\frac{\partial C'_{ijkl}(\overrightarrow{r})}{\partial r_j} + \frac{1}{2}\frac{\partial}{\partial r_j}[C'_{ijkl}(\overrightarrow{r})\delta\epsilon^{n-1}_{kl}] \\ &-(\epsilon^{*,\alpha}_{kl} - \epsilon^{*,\beta}_{kl})\frac{\partial}{\partial r_j}(C'_{ijkl}(\overrightarrow{r})p(\phi)) \qquad (33)\end{aligned}$$

The n^{th} order solution for displacement field can be obtained by substituting the $(n-1)^{\text{th}}$ order solution on the right hand side and following a similar procedure of using Fourier transforms. In this study, the system is constrained such that there is no macroscopic deformation. Therefore, the homogeneous strain, $\overline{\epsilon_{kl}} = 0$. The iterative approach described above is followed till the displacement field converges. Since the system in consideration is 2-D, we obtain displacement fields in two directions. The heterogeneous strain is calculated from the two final displacement fields using equation (12) and elastic strain field is calculated using equation (10).

3 Phase-field equations

The temporal evolution of the non-conserved phase-field variable, $\phi(\overrightarrow{r}$,t) is given by Ginzburg-Landau equation:

$$\begin{aligned}\frac{\partial \phi}{\partial t} &= -L\frac{\delta F}{\delta \phi} \qquad (34) \\ &= -L\left(\frac{\partial f^{gb}}{\partial \phi} + \frac{\partial f^{ch}}{\partial \phi} + \frac{\partial f^{el}}{\partial \phi}\right) \qquad (35) \\ \text{where } \frac{\partial f^{gb}}{\partial \phi} &= -\kappa \overrightarrow{\nabla}^2\phi + Wg'(\phi) \qquad (36)\end{aligned}$$

The temporal evolution of the conserved molar fraction field, c($\overrightarrow{r}$,t) is given by Cahn-Hilliard equation [8]:

$$\begin{aligned}\frac{\partial c}{\partial t} &= \overrightarrow{\nabla}.\left(M\overrightarrow{\nabla}\frac{\delta F}{\delta c}\right) \qquad (37) \\ &= \overrightarrow{\nabla}.\left(M\overrightarrow{\nabla}\frac{\partial f^{ch}}{\partial c}\right) \qquad (38)\end{aligned}$$

4 Details of the simulation

Firstly, the proposed model is compared with Khachaturyan's model. The strain fields for both approaches are solved by the iterative approach explained in Section 2.4. The phase-field equations given in Section 3 are solved by finite difference method. Periodic boundary conditions are assumed for the phase field, composition field and displacement fields.

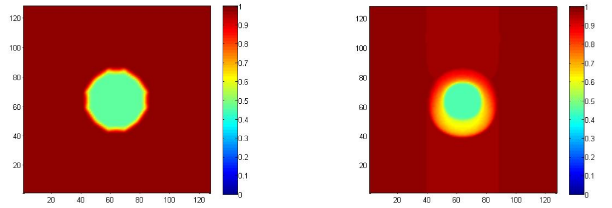
A matrix of 128 μm×128 μm is considered with a circular precipitate of radius 20 μm located at the centre. The Cu-Sn system is considered with α as the Sn (bct) phase and β as the Cu_6Sn_5 phase. Parabolic free energies with the following constants are used: A^{α} = 10^9 J/m^3, A^{β} = 10^{10} J/m^3, $c_{\alpha,0}$ = 0.979 and $c_{\beta,0}$ = 0.455 [9]. These values correspond to an approximation of the Gibbs free energies of the two phases calculated at 180°C. The stiffness tensors of the two phases are: C^{α}_{11} = 82.74 GPa, C^{α}_{12} = 57.85 GPa and C^{α}_{44} = 28.18 GPa, C^{β}_{11} = 168.57 GPa, C^{β}_{12} = 69.11 GPa, C^{β}_{22} = 165.39 GPa and C^{β}_{44} = 45.981 GPa [10, 11]. The eigenstrain is taken as 1% in the precipitate phase and zero in the matrix phase. The simulations are started from the equilibrium composition values. Time step of 0.001 sec is used.

The proposed model derived from Steinbach and Apel's approach is used for further simulations. According to Hu and Chen[2], the shape of a second-phase particle in a matrix depends on the ratio of shear moduli of the two phases. Hence, in this set of simulations, the values of the elastic modulus of the β phase are changed such that the bulk modulus B (= $C_{11} + C_{12}$) and the ratio of anisotropy δ (= $2C_{44}/(C_{11} - C_{12})$) are the same as that of the α phase. Simulations are performed for three different values of the ratio of the shear modulus ($C^{\beta}_{44}/C^{\alpha}_{44}$). A matrix of 128×128 cells is considered with a square 20×20 precipitate located at the centre. Parabolic free energies with the following constants are used: A^{α} = 0.173*10^9 J/m^3, A^{β} = 0.86*10^9 J/m^3, $c_{\alpha,0}$ = 0.053 and $c_{\beta,0}$ = 0.947. The eigenstrains are taken as 1% and 2% in the two phases respectively. Initial composition of 0.053 in the α phase and 0.947 in the β phase are used. Other parameters are assumed to be dimensionless.

5 Results and Discussion

The results from the proposed model and Khachaturyan's approach are compared for the growth of a Cu_6Sn_5 precipitate in

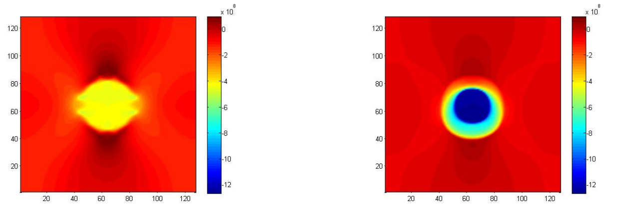
Sn matrix after 20000 time steps. The composition fields for the two approaches are shown in Figures 1(a) and 1(b). The evolution of the precipitate in Khachaturyan's approach is much faster than in the proposed model. This indicates that the elastic energy density contribution to the evolution of the system is higher in Khachaturyan's approach when realistic parameters are used. The proposed model is still able to predict the evolution with a stable shape of the precipitate. Due to higher eigenstrain in the precipitate phase, the precipitate tends to shrink in both cases.



(a) Proposed model (b) Khachaturyan's approach

Figure 1: Comparison of the composition fields obtained using the two approaches

Figures 2(a) and 2(b) compare the stress profile of σ_{11} component for the two approaches. The magnitude of stress inside the precipitate is higher in Khachaturyan's approach. The magnitude of the stress is highest inside the precipitate for both approaches, due to the presence of eigenstrain in the precipitate phase only.



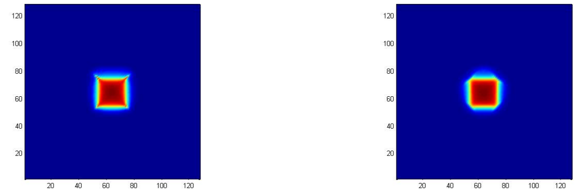
(a) Proposed model (b) Khachaturyan's approach

Figure 2: Comparison of the stress fields of σ_{11} component obtained using the two approaches

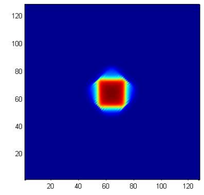
In the next set of simulations, the influence of the ratio of shear modulus on the precipitate shape is studied for 3 values of $C^{\beta}_{44}/C^{\alpha}_{44}$: 1.63, 0.81 and 0.54. Figures 3(a), 3(b) and 3(c) show the shape of the precipitate for the 3 cases after 50000 time steps by plotting the phase-field. For $C^{\beta}_{44}/C^{\alpha}_{44}$ = 1.63, the corners become pointed. For $C^{\beta}_{44}/C^{\alpha}_{44}$ = 0.81 and for $C^{\beta}_{44}/C^{\alpha}_{44}$ = 0.54, the corners become flat. Simulations performed for longer times will reveal more marked changes in the precipitate shape. The effect of ratio of the shear moduli is clearly seen.

6 Conclusions

A phase-field model for studying a binary system with elastic inhomogeneity was derived and implemented and simulation results for Cu_6Sn_5 particle in Sn matrix were compared with results obtained using Khachaturyan's model. The elastic contribution seemed to dominate the results in Khachaturyan's model, resulting in fast shrinkage of the precipitate, whereas the proposed model was able to demonstrate a stable evolution of the precipitate for realistic parameters. The proposed model also showed the effect of changing the ratio of shear modulus between the precipitate and the matrix phase on the precipitate shape.



(a) $C^{\beta}_{44}/C^{\alpha}_{44}$=1.63 (b) $C^{\beta}_{44}/C^{\alpha}_{44}$=0.81



(c) $C^{\beta}_{44}/C^{\alpha}_{44}$=0.54

Figure 3: Particle shapes for different ratios of shear moduli

Acknowledgements

References

[1] I. Steinbach and M. Apel, "Multi phase field model for solid state transformation with elastic strain," *Physica D*, vol. 217, pp. 153–160, 2006.

[2] S. Y. Hu and L. Q. Chen, "A phase-field model for evolving microstructures with strong elastic inhomogeneity," *Acta mater.*, vol. 49, pp. 1879–1890, 2001.

[3] L. Q. Chen, "Phase-field models for microstructure evolution," *Annu. Rev. Mater. Res.*, vol. 32, pp. 113–114, 2002.

[4] S. G. Kim, W. T. Kim, and T. Suzuki, "Phase-field model for binary alloys," *Phys. Rev. E*, vol. 60, pp. 7186–7196, 1999.

[5] K. Ammar, B. Appolaire, G. Cailletaud, and S. Forest, "Combining phase field approach and homogenization methods for modelling phase transformation in elastoplastic media," *European Journal of Computational Mechanics*, vol. 18, no. 5-6, pp. 485–523, 2009.

[6] N. Moelans, B. Blanpain, and P. Wollants, "An introduction to phase-field modeling of microstructure evolution," *Calphad*, vol. 32, pp. 268–294, 2008.

[7] A. G. Khachaturyan, *Theory of Structural Transformations in Solids*. New York: Wiley, 1983.

[8] J. W. Cahn and J. E. Hilliard, "Free energy of a nonuniform system. I. Interfacial energy," *J. Chem. Phys.*, vol. 28, no. 2, pp. 258–267, 1958.

[9] N. Moelans, "A phase-field model for multi-component and multi-phase systems," *Arch. Metall. Mater.*, vol. 53, no. 4, pp. 1147–1154, 2008.

[10] J. Chen and Y.-S. Lai, "Towards elastic anisotropy and strain-induced void formation in Cu-Sn crystalline phases," *Microelectron. Reliab.*, vol. 49, pp. 264–268, 2009.

[11] G. Ghosh, "First-Principles Calculation of Phase Stability and Cohesive Properties of Ni-Sn Intermetallics," *Metall. Mater. Trans. A*, vol. 40A, pp. 4–23, 2009.