KKS Phase Field Model of Precipitate Evolution

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The model employed in this application is described in detail in the article:

DeWitt et al., Misfit-driven β''' precipitate composition and morphology in Mg-Nd alloys,
Acta Materialia **137**, 378-389 (2017).

1 Variational formulation

The total free energy of the system (neglecting boundary terms) is of the form,

$$\Pi(c, \eta_1, \eta_2, \eta_3, \boldsymbol{\epsilon}) = \int_\Omega f(c, \eta_1, \eta_2, \eta_3, \boldsymbol{\epsilon}) \, dV \tag{1}$$

where c is the concentration of the β phase, η_p are the structural order parameters and $\boldsymbol{\epsilon}$ is the small strain tensor. f, the free energy density is given by

$$f(c, \eta_1, \eta_2, \eta_3, \boldsymbol{\epsilon}) = f_{chem}(c, \eta_1, \eta_2, \eta_3) + f_{grad}(\eta_1, \eta_2, \eta_3) + f_{elastic}(c, \eta_1, \eta_2, \eta_3, \boldsymbol{\epsilon}) \tag{2}$$

where

$$f_{chem}(c, \eta_1, \eta_2, \eta_3) = f_\alpha(c, \eta_1, \eta_2, \eta_3) \left(1 - \sum_{p=1}^{3} H(\eta_p) \right) + f_\beta(c, \eta_1, \eta_2, \eta_3) \sum_{p=1}^{3} H(\eta_p) + W f_{Landau}(\eta_1, \eta_2, \eta_3) \tag{3}$$

$$f_{grad}(\eta_1, \eta_2, \eta_3) = \frac{1}{2} \sum_{p=1}^{3} \kappa_{ij}^{\eta_p} \eta_{p,i} \eta_{p,j} \tag{4}$$

$$f_{elastic}(c, \eta_1, \eta_2, \eta_3, \boldsymbol{\epsilon}) = \frac{1}{2} C_{ijkl}(\eta_1, \eta_2, \eta_3) \left(\varepsilon_{ij} - \varepsilon_{ij}^0(c, \eta_1, \eta_2, \eta_3) \right) \left(\varepsilon_{kl} - \varepsilon_{kl}^0(c, \eta_1, \eta_2, \eta_3) \right) \tag{5}$$

$$\varepsilon^0(c, \eta_1, \eta_2, \eta_3) = H(\eta_1) \varepsilon_{\eta_1}^0(c_\beta) + H(\eta_2) \varepsilon_{\eta_2}^0(c_\beta) + H(\eta_3) \varepsilon_{\eta_3}^0(c_\beta) \tag{6}$$

$$\boldsymbol{C}(\eta_1, \eta_2, \eta_3) = H(\eta_1) \boldsymbol{C}_{\eta_1} + H(\eta_2) \boldsymbol{C}_{\eta_2} + H(\eta_3) \boldsymbol{C}_{\eta_3} + (1 - H(\eta_1) - H(\eta_2) - H(\eta_3)) \boldsymbol{C}_\alpha \tag{7}$$

Here $\varepsilon_{\eta_p}^0$ are the composition dependent stress free strain transformation tensor corresponding to each structural order parameter, which is a function of the β phase concentration, c_β, defined below.

In the KKS model (Kim 1999), the interfacial region is modeled as a mixture of the α and β phases with concentrations c_{alpha} and c_{beta}, respectively. The homogenous free energies for each phase, f_α and f_β in this case, are typically given as functions of c_α and c_β, rather than directly as functions of c and η_p. Thus, $f_{chem}(c, \eta_1, \eta_2, \eta_3)$ can be rewritten as

$$f_{chem}(c, \eta_1, \eta_2, \eta_3) = f_\alpha(c_\alpha) \left(1 - \sum_{p=1}^{3} H(\eta_p) \right) + f_\beta(c_\beta) \sum_{p=1}^{3} H(\eta_p) + W f_{Landau}(\eta_1, \eta_2, \eta_3) \tag{8}$$

The concentration in each phase is determined by the following system of equations:

$$c = c_\alpha \left(1 - \sum_{p=1}^{3} H(\eta_p) \right) + c_\beta \sum_{p=1}^{3} H(\eta_p) \tag{9}$$

$$\frac{\partial f_\alpha(c_\alpha)}{\partial c_\alpha} = \frac{\partial f_\beta(c_\beta)}{\partial c_\beta} \tag{10}$$

Given the following parabolic functions for the single-phase homogenous free energies:

$$f_\alpha(c_\alpha) = A_2 c_\alpha^2 + A_1 c_\alpha + A_0 \tag{11}$$

$$f_\beta(c_\beta) = B_2 c_\beta^2 + B_1 c_\beta + B_0 \tag{12}$$

the single-phase concentrations are:

$$c_\alpha = \frac{B_2 c + \frac{1}{2}(B_1 - A_1)\sum_{p=1}^{3} H(\eta_p)}{A_2 \sum_{p=1}^{3} H(\eta_p) + B_2\left(1 - \sum_{p=1}^{3} H(\eta_p)\right)} \tag{13}$$

$$c_\beta = \frac{A_2 c + \frac{1}{2}(A_1 - B_1)\left[1 - \sum_{p=1}^{3} H(\eta_p)\right]}{A_2 \sum_{p=1}^{3} H(\eta_p) + B_2\left[1 - \sum_{p=1}^{3} H(\eta_p)\right]} \tag{14}$$

2 Required inputs

- $f_\alpha(c_\alpha), f_\beta(c_\beta)$ - Homogeneous chemical free energy of the components of the binary system, example form given above

- $f_{Landau}(\eta_1, \eta_2, \eta_3)$ - Landau free energy term that controls the interfacial energy and prevents precipitates with different orientation varients from overlapping, example form given in Appendix I

- W - Barrier height for the Landau free energy term, used to control the thickness of the interface

- $H(\eta_p)$ - Interpolation function for connecting the α phase and the pth orientation variant of the β phase, example form given in Appendix I

- $\boldsymbol{\kappa}^{\eta_p}$ - gradient penalty tensor for the pth orientation variant of the β phase

- $\boldsymbol{C}_{\eta_p}$ - fourth order elasticity tensor (or its equivalent second order Voigt representation) for the pth orientation variant of the β phase

- $\boldsymbol{C}_\alpha$ - fourth order elasticity tensor (or its equivalent second order Voigt representation) for the α phase

- $\boldsymbol{\varepsilon}_{\eta_p}^0$ - stress free strain transformation tensor for the pth orientation variant of the β phase

In addition, to drive the kinetics, we need:

- M - mobility value for the concentration field

- L - mobility value for the structural order parameter field

3 Variational treatment

We obtain chemical potentials for the chemical potentials for the concentration and the structural order parameters by taking variational derivatives of Π:

$$\mu_c = f_{\alpha,c}\left(1 - H(\eta_1) - H(\eta_2) - H(\eta_3)\right) + f_{\beta,c}\left(H(\eta_1) + H(\eta_2) + H(\eta_3)\right) + \boldsymbol{C}_{ijkl}(-\varepsilon_{ij,c}^0)\left(\varepsilon_{kl} - \varepsilon_{kl}^0\right) \tag{15}$$

$$\mu_{\eta_p} = [f_\beta - f_\alpha - (c_\beta - c_\alpha)f_{\beta,c_\beta}]H(\eta_p)_{,\eta_p} + W f_{Landau,\eta_p} - \kappa_{ij}^{\eta_p}\eta_{p,ij} + \boldsymbol{C}_{ijkl}(-\varepsilon_{ij,\eta_p}^0)\left(\varepsilon_{kl} - \varepsilon_{kl}^0\right) + \frac{1}{2}\boldsymbol{C}_{ijkl,\eta_p}\left(\varepsilon_{ij} - \varepsilon_{ij}^0\right) \tag{16}$$

4   Kinetics

Now the PDE for Cahn-Hilliard dynamics is given by:

$$\frac{\partial c}{\partial t} = \boldsymbol{\nabla} \cdot \left(\frac{1}{f_{,cc}} M \boldsymbol{\nabla} \mu_c \right) \tag{17}$$

where M is a constant mobility and the factor of $\frac{1}{f_{,cc}}$ is added to guarentee constant diffusivity in the two phases. The PDE for Allen-Cahn dynamics is given by:

$$\frac{\partial \eta_p}{\partial t} = -L \mu_{\eta_p} \tag{18}$$

where L is a constant mobility.

5   Mechanics

Considering variations on the displacement u of the from $u + \epsilon w$, we have

$$\delta_u \Pi = \int_{\Omega} \boldsymbol{\nabla} w : \boldsymbol{C}(\eta_1, \eta_2, \eta_3) : \left(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^0(c, \eta_1, \eta_2, \eta_3) \right) \ dV = 0 \tag{19}$$

$$\tag{20}$$

where $\boldsymbol{\sigma} = \boldsymbol{C}(\eta_1, \eta_2, \eta_3) : \left(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^0(c, \eta_1, \eta_2, \eta_3) \right)$ is the stress tensor.

6   Time discretization

Using forward Euler explicit time stepping, equations 17 and 18 become:

$$c^{n+1} = c^n + \Delta t \left[\nabla \cdot \left(\frac{1}{f_{,cc}} M \nabla \mu_c \right) \right] \tag{21}$$

$$\eta_p^{n+1} = \eta_p^n - \Delta t L \mu_{\eta_p} \tag{22}$$

7   Weak formulation

Writing equations 17 and 18 in the weak form, with the arbirary variation given by w yields:

$$\int_{\Omega} w c^{n+1} dV = \int_{\Omega} w c^n + w \Delta t \left[\nabla \cdot \left(\frac{1}{f_{,cc}} M \nabla \mu_c \right) \right] dV \tag{23}$$

$$\int_{\Omega} w \eta_p^{n+1} dV = \int_{\Omega} w \eta_p^n - w \Delta t L \mu_{\eta_p} dV \tag{24}$$

The expression of $\frac{1}{f_{,cc}}\mu_c$ can be written as:

$$\frac{1}{f_{,cc}}\nabla\mu_c = \nabla c + (c_\alpha - c_\beta)\sum_{p=1}^{3}H(\eta_p)_{,\eta_p}\nabla\eta_p$$

$$+ \frac{1}{f_{,cc}}\left[\sum_{p=1}^{3}(C_{ijkl}^{\eta_p} - C_{ijkl}^{\alpha})\nabla\eta_p H(\eta_p)_{,\eta_p}\right](-\epsilon_{ij,c}^0)(\epsilon_{ij} - \epsilon_{ij}^0)$$

$$- \frac{1}{f_{,cc}}C_{ijkl}\left[\sum_{p=1}^{3}\left(H(\eta_p)_{,\eta_p}\epsilon_{ij,c}^{0\eta_p} + \sum_{q=1}^{3}\left(H(\eta_p)\epsilon_{ij,c\eta_q}^{0\eta_p}\right)\right)\nabla\eta_p + H(\eta_p)\epsilon_{ij,cc}^{0\eta_p}\nabla c\right](\epsilon_{kl} - \epsilon_{kl}^0)$$

$$+ \frac{1}{f_{,cc}}C_{ijkl}(-\epsilon_{ij,c}^0)\left[\nabla\epsilon_{kl} - \left(\sum_{p=1}^{3}\left(H(\eta_p)_{,\eta_p}\epsilon_{kl}^{0\eta_p} - \sum_{q=1}^{3}\epsilon_{kl,\eta_q}^{\eta_q}H(\eta_q)\right)\nabla\eta_p + H(\eta_p)\epsilon_{kl,c}^{0\eta_p}\nabla c\right)\right]$$

$$\tag{25}$$

Applying the divergence theorem to equation 23, one can derive the residual terms r_c and r_{cx}:

$$\int_\Omega wc^{n+1}dV = \int_\Omega w\underbrace{c^n}_{r_c} + \nabla w\cdot\underbrace{(-\Delta tM\frac{1}{f_{,cc}}\nabla\mu_c)}_{r_{cx}}dV \tag{26}$$

Expanding μ_{η_p} in equation 24 and applying the divergence theorem yields the residual terms r_{η_p} and $r_{\eta_p x}$:

$$\int_\Omega w\eta_p^{n+1}dV =$$

$$\int_\Omega w\left\{\underbrace{\eta_p^n - \Delta tL\left[(f_\beta - f_\alpha)H(\eta_p^n)_{,\eta_p} - (c_\beta - c_\alpha)f_{\beta,c_\beta}H(\eta_p^n)_{,\eta_p} + Wf_{Landau,\eta_p}\right.}_{r_{\eta_p}}\right.$$

$$\underbrace{\left.-C_{ijkl}\left(H(\eta_p)_{,\eta_p}\epsilon_{ij}^{0\eta_p}\right)(\epsilon_{kl} - \epsilon_{kl}^0) + \frac{1}{2}\left[(C_{ijkl}^{\eta_p} - C_{ijkl}^\alpha)H(\eta_p)_{,\eta_p}\right](\epsilon_{ij} - \epsilon_{ij}^0)(\epsilon_{kl} - \epsilon_{kl}^0)\right]}_{r_{\eta_p}\ cont.}\right\}$$

$$+ \nabla w\cdot\underbrace{(-\Delta tL\boldsymbol{\kappa}_{ij}^{\eta_p}\eta_{p,i}^n)}_{r_{\eta_p x}}dV \tag{27}$$

8 Appendix I: Example functions for f_α, f_β, f_{Landau}, $H(\eta_p)$

$$f_\alpha(c_\alpha) = A_2 c_\alpha^2 + A_1 c_\alpha + A_0 \tag{28}$$

$$f_\beta(c_\beta) = B_2 c_\beta^2 + B_1 c_\beta + B_0 \tag{29}$$

$$f_{Landau}(\eta_1, \eta_2, \eta_3) = (\eta_1^2 + \eta_2^2 + \eta_3^2) - 2(\eta_1^3 + \eta_2^3 + \eta_3^3) + (\eta_1^4 + \eta_2^4 + \eta_3^4) + 5(\eta_1^2\eta_2^2 + \eta_2^2\eta_3^2 + \eta_1^2\eta_3^2) + 5(\eta_1^2\eta_2^2\eta_3^2) \tag{30}$$

$$H(\eta_p) = 3\eta_p^2 - 2\eta_p^3 \tag{31}$$