

Co-Based Superalloy Morphology Evolution: A Phase Field Study Based on Experimental Thermodynamic and Kinetic Data

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Abstract

Cobalt-based superalloys with two phase γ/γ' microstructures offer great promise as candidates for next-generation high-temperature alloys for applications, such as turbine blades. It is essential to understand the thermodynamic and kinetic factors that influence the microstructural evolution of these alloys in order to optimize the alloy compositions and processing steps with a goal to improve their coarsening, creep and rafting behavior. We are using a continuum phase field approach to study the diffusion process and to predict the equilibrium shapes of Co-Al-W γ' precipitates. In order to obtain quantitatively predictive capabilities, we extract chemical free energies for the γ/γ' phases based on CALculation of PHase Diagrams (CALPHAD) thermodynamic data and diffusion mobilities for Co alloys based on CALPHAD kinetic data. We also use experimental or first-principles data for other quantities, such as misfit strain and interface information, for the parameterization of our model. A particular focus of our study is to understand how different energy balances, misfit strain and kinetics affect the coarsening and rafting behavior of γ' precipitates, and the sensitivity of the final precipitate shape to materials parameters. We find that the equilibrium shape of the precipitate results from a delicate competition between chemical, interfacial, and elastic energies, and it is very sensitive to changes in model parameters. We examine how modeling input parameters affect the equilibrium shape of precipitates and relate these parameters to experimentally available values.

Keywords: Cobalt-based superalloys, Morphology evolution, Phase field model

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1. Introduction

Co-based superalloys with two phase γ/γ' microstructures offer great promise as candidates for next-generation alloys for high-temperature applications, such as turbine blades. Compared with the traditional Ni-based superalloys, Co-based superalloys have higher melting temperatures and superior hot corrosion resistance, which allow for a potentially higher operating temperature, and thus improve the efficiency in high-temperature applications, such as turbines [1, 2]. A γ/γ' microstructure in Co-Al-W superalloys was first reported by Sato et al. in 2006 [3]. Just as in the Ni-based superalloys, the γ/γ' microstructure plays an important role in enhancing the strength and creep resistance of the Co-based superalloys [4]. Both phases have face centered cubic (FCC) based crystal structures; the γ (A1) phase is a disordered solid solution and makes up the matrix of the superalloy, while the γ' phase, which has the form $\text{Co}_3(\text{Al}, \text{W})$, is an ordered L1_2 structure and constitutes the precipitate used to strengthen the alloy.

The Center for Hierarchical Materials Design (CHiMaD) [5] has spent substantial efforts on Co-based superalloy development over the past years, employing an integrated computational materials engineering (ICME) approach to accelerate the development cycle. In ICME, materials models and experimental data at multiple length scales are linked together. The CHiMaD database group focuses on developing the necessary CALPHAD-based thermodynamic and kinetic databases for the Co-based superalloys [6], other efforts employ techniques such as 3D atom-probe tomography (3D-APT) to measure the composition and study the interface of the Co-based superalloys [7], and phase field models based on the databases and other experimental results are used to investigate both the equilibrium shapes and coarsening behavior of γ' precipitates.

There are many studies on the equilibrium shapes of precipitates [8, 9, 10, 11, 12, 13, 14, 15] using general models investigating the balance between elastic and interfacial energies, and also including imperfections such as dislocations and structural ledges, e.g., Shi et al.[16]. Johnson and Cahn[8] first investigated shape transitions of inclusions due to elastic misfit and interfacial energy for isotropic systems. Using symmetry arguments and simple calculations in two and three dimensions they showed that when the inclusion is softer than the matrix, the inclusion shape has a transition to a lower-symmetry shape. Thomson, Su, and Voorhees[9] examined equilibrium morphologies of inclusions under tetragonal or dilatational misfit in an elastically anisotropic cubic medium. Using numerical simulations in two dimensions, they showed that the shape can bifurcate from a circle to a four-fold (purely dilatational) or two-fold (tetragonal misfit) symmetry shape. Mueller and Gross[11] performed three-dimensional (3D) simulations of equilibrium shapes of precipitates in a matrix using generalized thermodynamic forces, including interfacial and elastic energies, acting on the interface. They discussed their results in the context of Ni-based superalloys, showing that there is a size-induced transition from spherical to cubic precipitates as the size of the precipitate grows, and also that there is a morphology transition, rafting, to elongated precipitates for high ratios between the precipitate and matrix shear moduli. More recently, Li et al.[14] investigated the equilibrium shape of precipitates under misfit strain in an elastically anisotropic system in two and three dimensions, and derived a corresponding

Gibbs-Thomson equation in terms of a characteristic ratio L' between elastic and interfacial energies. Generally, a large L' the shape bifurcates into a lower-symmetry stable shape and a higher-symmetry metastable shape.

These works studied general isotropic or cubic materials, and sometimes discussed the results in the context of Ni-based superalloys, and did not discuss effects of the size of the precipitate (except for Mueller and Gross[11]). In a recent ~~an earlier~~ work, Jokisaari et al. used a phase field model to predict the equilibrium precipitate shape in the Co-Al-W 3D system [17]. That model incorporated elastic energy and interfacial energy without directly including phase compositions. Even though the model was simple, it provided valuable insight into how variations in elastic moduli, interfacial energy and γ/γ' misfit strain affect the precipitate morphology.

Recently, Wang et al. [18] studied the rafting behavior and creep properties of the Co-Al-W ternary system taking the formulation of chemical free energy into account, with different W composition and under different applied stress. In this work, we expand on the model by Jokisaari et al. [17] to incorporate composition-dependent chemical free energies for the γ/γ' phase as well as mobilities based on CALPHAD thermodynamic and kinetic data developed at National Institute of Standards and Technology (NIST) [6]. We also use experimental and first-principles data for quantities such as lattice misfit and interfacial width that are needed for parameterization of other energy terms in the phase field model. We first test our model on two-dimensional (2D) systems before going to much more computationally expensive three-dimensional (3D) simulations. We observe similar behavior of the system evolution in 2D and 3D simulations and will here only present and discuss results of the 3D simulations. In addition to the shape bifurcation behavior reported by Jokisaari et al., [17], we observe other characteristic microstructures as a result of the competition between the chemical free energies for the γ/γ' phase, interfacial energy, and elastic energy. A particular focus of our study is to understand how different energy balances, misfit strain and kinetics affect the coarsening and rafting behavior of γ' precipitates. Furthermore, we examine how modeling input parameters, e.g., the initial alloy composition, the barrier height of the double-well potential, the gradient energy coefficient, and the misfit strain affect the size and shape of the equilibrium precipitates, and perform a sensitivity study of the results to input parameters. It is our hope that such sensitivity analysis can provide useful guidance both for future computational as well as experimental efforts.

2. Model formulations

In the phase field model, the γ/γ' two-phase microstructure can be described by a non-conserved order parameter η that takes the value of zero in the γ phase, unity in the γ' phase, and varies smoothly through the interfaces, together with a conserved composition field variable c (in mole fraction) that describes the local Co concentration. We can use only one composition field to describe the ternary phase of Co-Al-W superalloy because the composition of Al during the microstructural evolution remains approximately the same in the two phases. This is confirmed from both 3D-APT experiments and phase diagrams generated by CALPHAD calculations. Bocchini et al. [7] used 3D-APT to measure the composition of Co, W, Al vs. distance across the γ/γ' interface in the Co-9.7Al-10.8W alloy at 900 °C. While the mole fraction of Co and W can be 6 % to 7 % different in the

two phases, the difference of the Al mole fraction is smaller than 1 %. Furthermore, the phase diagram of Co-Al-W at 900 °C (Fig. 1) shows that the tie line between the disordered FCC γ phase and the ordered L1₂ γ' phase is parallel to the base line of the triangle, which indicates that the composition of Al is the same in the two phases. Furthermore, W is a heavy element and its mobility is much less than the other two elements and diffusion of W can be ignored. Therefore, we can reduce the phase field model only to include diffusion of Co in our simulations.

We use a Kim-Kim-Suzuki (KKS) formulation [19] of the phase field model, which assumes an equal chemical potential in the two phases so that the bulk chemical contribution from the interfacial region can be removed. The free energy of the system is then given by the function

$$\mathcal{F}[c, \eta] = \int_V [f_{\text{chem}}(c, \eta) + f_{\text{grad}}(\eta) + f_{\text{elas}}(\eta)] dV, \quad (1)$$

where $f_{\text{chem}}(c, \eta)$ is the local chemical free energy density, $f_{\text{grad}}(\eta)$ is the gradient energy density, and $f_{\text{elas}}(\eta)$ is the elastic energy density. The gradient energy density is independent of composition and is expressed as $\frac{\kappa}{2}(\nabla\eta)^2$, where κ is the gradient energy density coefficient in units of energy per unit length. The elastic free energy density also depends only on the order parameter η (and not on the composition field c) to represent the difference in elasticity between the two phases. The elastic energy of the precipitate-matrix system is driven by a misfit strain between the crystal structure of the matrix phase γ and precipitate phase γ' .

2.1. Chemical free energy density

The local chemical free energy density includes the free energy density of each phase, and a double-well potential which describes the structural difference between the disordered γ matrix and the ordered γ' precipitates. It is given by

$$f_{\text{chem}}(c, \eta) = f_{\gamma}(c_{\gamma})(1 - h(\eta)) + f_{\gamma'}(c_{\gamma'})h(\eta) + \omega g(\eta), \quad (2)$$

where $f_{\gamma}(c_{\gamma})$ and $f_{\gamma'}(c_{\gamma'})$ are the free energy densities of the γ and γ' phase, respectively, c_{γ} and $c_{\gamma'}$ are the phase compositions which are defined in the individual phase regions and overlapping in the interface, $h(\eta) = \eta^3(6\eta^2 - 15\eta + 10)$ is the interpolation function, such that $h(0) = h'(0) = h'(1) = 0$ and $h(1) = 1$, ω is the barrier height for the double-well potential, and the double-well potential $g(\eta)$ has the same 10th-order polynomial form as what is used in Jokisaari et al. [17], where $g(0) = g(1) = g'(0) = g'(1) = 0$ and the energy density remains downward concave between the two energy wells. [This particular double-well potential is chosen to prevent the actual value of \$\eta\$ in each phase from shifting significantly from its equilibrium value due to the presence of a curved interface or elastic strain.](#) An illustration of function g is shown in Fig. 2.

Equality of chemical potentials through the interface is required to smoothly connect the phase compositions coexisting in the interface. The following constraint equations are added

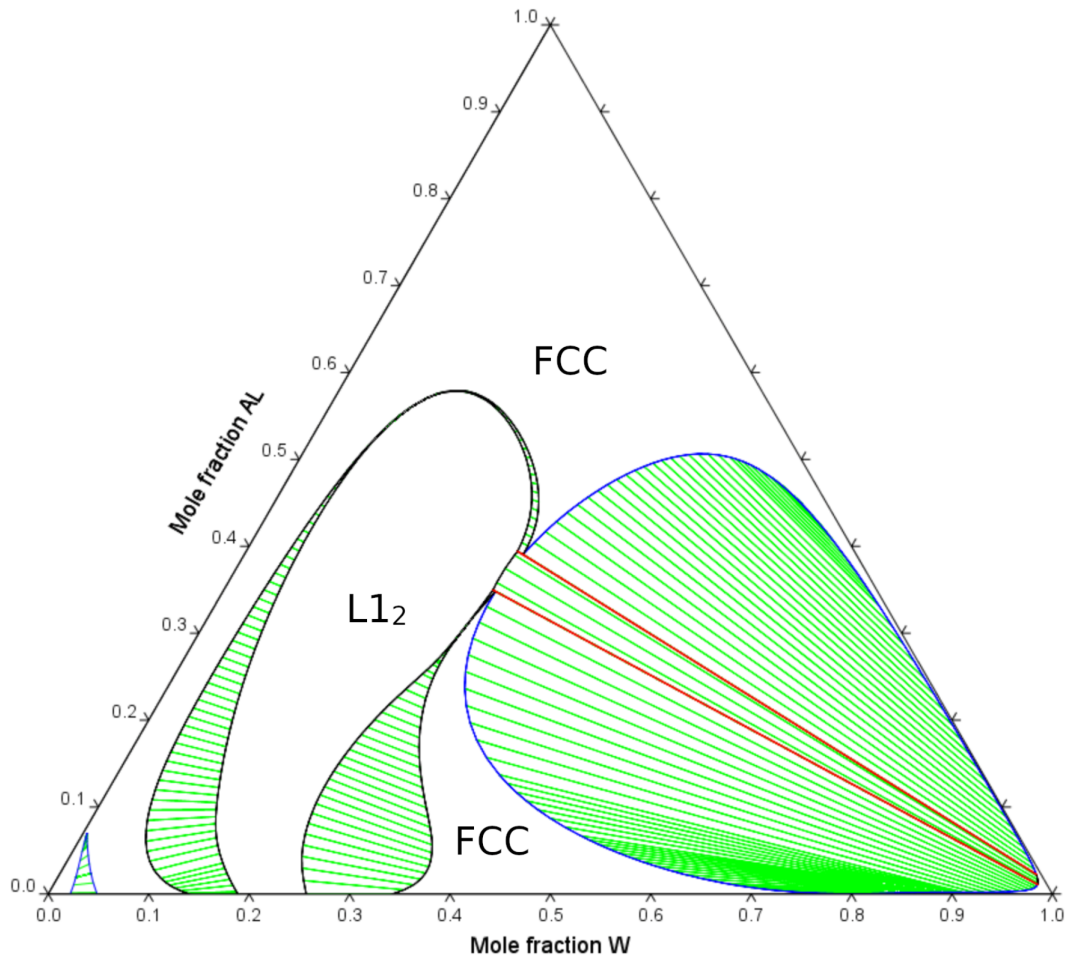


Figure 1: The γ/γ' phase diagram of Co-Al-W at 900 °C. The red lines represent degenerated three phase equilibria $\gamma'(L1_2)+\gamma$ (FCC) + γ (FCC) resulting from the miscibility gap in the γ (FCC) phase.

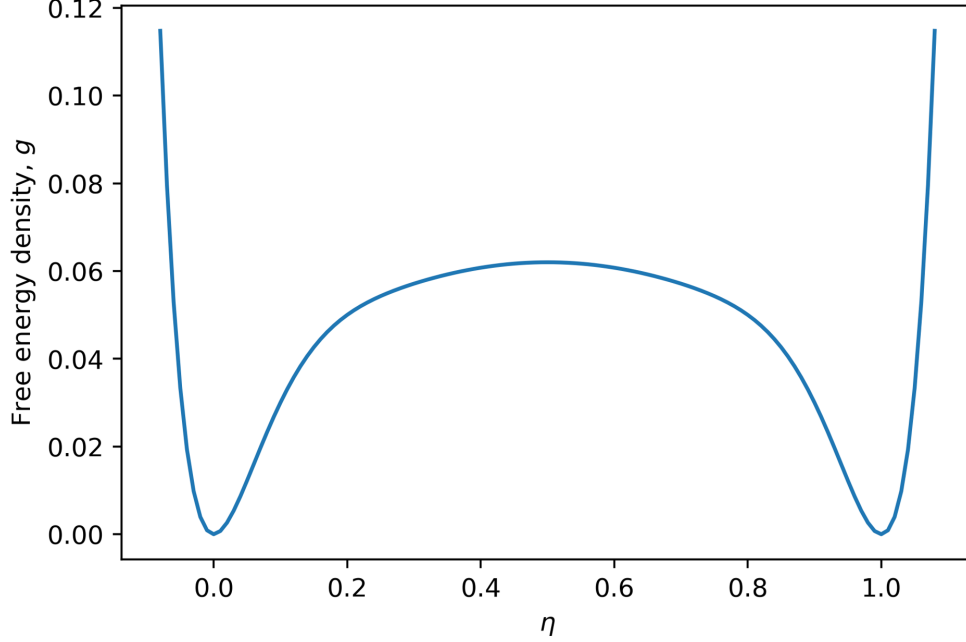


Figure 2: The double-well energy density g . The energy wells are narrow and deep compared to the common double-well potential $\eta^2(1 - \eta)^2$, to prevent the actual value of η in each phase from shifting significantly from its equilibrium value since a large shift would introduce non-negligible error into the elastic energy calculation.

in the KKS model in order to maintain the mass conservation and equality of the chemical potentials:

$$c = (1 - h(\eta))c_\gamma + h(\eta)c_{\gamma'} \quad (3)$$

$$\mu_c = \frac{\partial f_{\text{chem}}}{\partial c} = \frac{\partial f_\gamma}{\partial c_\gamma} = \frac{\partial f_{\gamma'}}{\partial c_{\gamma'}}. \quad (4)$$

The free energies of the γ and γ' phases have been determined from measurements and thermodynamic analyses as a function of composition and temperature [6]. In this work, the temperature is fixed at 900 °C, thus f_γ and $f_{\gamma'}$ are functions of composition only. In the γ phase, all lattice sites are equivalent and the disordered solution is formed from a pure substance by including substitutions of atoms on the lattice sites. Following the general guideline by Kattner [20], we describe the free energy density of the disordered γ phase in terms of mole fractions using a substitutional solution model:

$$\begin{aligned} f_\gamma = & c_{\text{Al}}G_{\text{Al}} + c_{\text{Co}}G_{\text{Co}} + c_{\text{W}}G_{\text{W}} + RT(c_{\text{Al}}\ln c_{\text{Al}} + c_{\text{Co}}\ln c_{\text{Co}} + c_{\text{W}}\ln c_{\text{W}}) \\ & + c_{\text{Al}}c_{\text{Co}}[L_{\text{Al,Co}}^0 + L_{\text{Al,Co}}^2(c_{\text{Al}} - c_{\text{Co}})^2] + c_{\text{Al}}c_{\text{W}}[L_{\text{Al,W}}^0 + L_{\text{Al,W}}^1(c_{\text{Al}} - c_{\text{W}})] \\ & + c_{\text{Co}}c_{\text{W}}[L_{\text{Co,W}}^0 + L_{\text{Co,W}}^1(c_{\text{Co}} - c_{\text{W}})] + c_{\text{Al}}c_{\text{Co}}c_{\text{W}}[c_{\text{Al}}L_{\text{Al,Co,W}}^0 + c_{\text{Co}}L_{\text{Al,Co,W}}^1 \\ & + c_{\text{W}}L_{\text{Al,Co,W}}^2] + G_{\text{mag}}, \end{aligned} \quad (5)$$

where $c_{\text{Al}} = 0.1$ is a constant, c_{Co} corresponds to the composition field variable c used in the phase field formulation, and $c_{\text{W}} = 0.9 - c$, G_{Al} , G_{Co} and G_{W} are the Gibbs free energy densities of the pure Al, Co and W phase as given by Dinsdale [21], R is the gas constant,

$T = 1173$ K is the temperature, Ls are the Redlich-Kister coefficients [22], and G_{mag} is the magnetism contribution to the free energy density. The Redlich-Kister coefficient Ls and G_{mag} can be obtained from the CALPHAD thermodynamic database [6].

The γ' phase is an ordered form of the γ phase, and, therefore, can be described as one phase by using an order-disorder model [23] with two sublattices $(\text{Al}, \text{Co}, \text{W})_{\text{a}}(\text{Al}, \text{Co}, \text{W})_{\text{b}}$ where $a = 0.75$ and $b = 0.25$. The free energy density of the ordered γ' phase is composed of two parts – the Gibbs free energy density for the disordered γ phase f_{γ} described by Eq. (5) and the long range ordering contribution to the Gibbs free energy density ΔG^{ord} :

$$f_{\gamma'} = f_{\gamma} + \Delta G^{\text{ord}}. \quad (6)$$

In order to describe the constitution of a phase with two sublattices the mole fraction is not enough, we need another type of composition variable, the constituent fraction, which describes the composition of atoms within the individual sublattices. The free energy density ΔG^{ord} is then given by

$$\begin{aligned} \Delta G^{\text{ord}} = & G^{\text{ord}}(y'_{\text{Al}}, y''_{\text{Al}}, y'_{\text{Co}}, y''_{\text{Co}}, y'_{\text{W}}, y''_{\text{W}}) \\ & - G^{\text{ord}}(y'_{\text{Al}} = c_{\text{Al}}, y''_{\text{Al}} = c_{\text{Al}}, y'_{\text{Co}} = c_{\text{Co}}, y''_{\text{Co}} = c_{\text{Co}}, y'_{\text{W}} = c_{\text{W}}, y''_{\text{W}} = c_{\text{W}}), \end{aligned} \quad (7)$$

where y'_A is the constituent fraction of A in the first sublattice and y''_A is the constituent fraction of A in the second sublattice. ($A \in \{\text{Al}, \text{Co}, \text{W}\}$) y' and y'' are equal to 0 when the ordering disappears. The constituent fractions can be related to the mole fraction via:

$$ay'_A + by''_A = c_A, \quad (8)$$

and G^{ord} can be described by the Compound Energy Formalism (CEF) [24]:

$$\begin{aligned}
G^{\text{ord}} = & y'_{\text{Al}} y''_{\text{Al}} G_{\text{Al:Al}} + y'_{\text{Al}} y''_{\text{Co}} G_{\text{Al:Co}} + y'_{\text{Al}} y''_{\text{W}} G_{\text{Al:W}} + y'_{\text{Co}} y''_{\text{Al}} G_{\text{Co:Al}} + y'_{\text{Co}} y''_{\text{Co}} G_{\text{Co:Co}} \\
& + y'_{\text{Co}} y''_{\text{W}} G_{\text{Co:W}} + y'_{\text{W}} y''_{\text{Al}} G_{\text{W:Al}} + y'_{\text{W}} y''_{\text{Co}} G_{\text{W:Co}} + y'_{\text{W}} y''_{\text{W}} G_{\text{W:W}} \\
& + RT (a(y'_{\text{Al}} \ln y'_{\text{Al}} + y'_{\text{Co}} \ln y'_{\text{Co}} + y'_{\text{W}} \ln y'_{\text{W}}) + b(y''_{\text{Al}} \ln y''_{\text{Al}} + y''_{\text{Co}} \ln y''_{\text{Co}} + y''_{\text{W}} \ln y''_{\text{W}})) \\
& + y'_{\text{Al}} y'_{\text{Co}} \left(\sum_{j \in \{\text{Al, Co, W}\}} y''_j \sum_{i=0}^1 L_{\text{Al, Co: } j}^i (y'_{\text{Al}} - y'_{\text{Co}})^i \right) \\
& + y'_{\text{Al}} y'_{\text{W}} \left(\sum_{j \in \{\text{Al, Co, W}\}} y''_j \sum_{i=0}^1 L_{\text{Al, W: } j}^i (y'_{\text{Al}} - y'_{\text{W}})^i \right) \\
& + y'_{\text{Co}} y'_{\text{W}} \left(\sum_{j \in \{\text{Al, Co, W}\}} y''_j \sum_{i=0}^1 L_{\text{Co, W: } j}^i (y'_{\text{Co}} - y'_{\text{W}})^i \right) \\
& + y'_{\text{Al}} y'_{\text{Co}} y'_{\text{W}} (y''_{\text{Al}} L_{\text{Al, Co, W: Al}}^0 + y''_{\text{Co}} L_{\text{Al, Co, W: Co}}^0 + y''_{\text{W}} L_{\text{Al, Co, W: W}}^0) \\
& + y''_{\text{Al}} y''_{\text{Co}} \left(\sum_{j \in \{\text{Al, Co, W}\}} y'_j \sum_{i=0}^1 L_{j: \text{Al, Co}}^i (y''_{\text{Al}} - y''_{\text{Co}})^i \right) \\
& + y''_{\text{Al}} y''_{\text{W}} \left(\sum_{j \in \{\text{Al, Co, W}\}} y'_j \sum_{i=0}^1 L_{j: \text{Al, W}}^i (y''_{\text{Al}} - y''_{\text{W}})^i \right) \\
& + y''_{\text{Co}} y''_{\text{W}} \left(\sum_{j \in \{\text{Al, Co, W}\}} y'_j \sum_{i=0}^1 L_{j: \text{Co, W}}^i (y''_{\text{Co}} - y''_{\text{W}})^i \right).
\end{aligned} \tag{9}$$

where $G_{A:B}$ is the Gibbs energy of ‘compound’ $A_a B_b$, usually called an ‘endmember’ of the phase, and $L_{A,B:A}$ is the interaction parameter between A and B on the first sublattice when the second sublattice is filled with A . Notice that here $A \in \{\text{Al, Co, W}\}$, $B \in \{\text{Al, Co, W}\}$ and $A \neq B$.

The free energy densities of the γ and γ' phases are shown in Fig. 3. The free energy density of the γ' phase appears as a yellow band because we converted the constituent fractions into the mole fraction using Eq. (8). Different combinations of constituent fractions can result in the same mole fraction. However, site disorder equilibrates on a time scale much faster than the microstructural evolution. Therefore, we can restrict the free energy density of the γ' phase to the minimum of the site occupancies at a given mole fraction, shown as the red line in Fig. 3 (b). Notice that the red curve overlaps with the black curve (free energy density of the γ phase) when the mole fraction of Co is low, which means that the ordering of L1₂ γ' phase does not exist. Common tangent line construction based on Fig. 3 (b) yields equilibrium compositions $c_{\gamma}^{\text{eq}} = 0.8638$ and $c_{\gamma'}^{\text{eq}} = 0.7955$, in agreement with the phase diagram in Fig. 1. Since we are only interested in alloys with composition around Co-10Al-10W, and γ' phase does not exist when the Co composition is low, we use a quadratic fit for the free energy densities of the two phases at $c > 0.6$. The Gibbs free energy densities

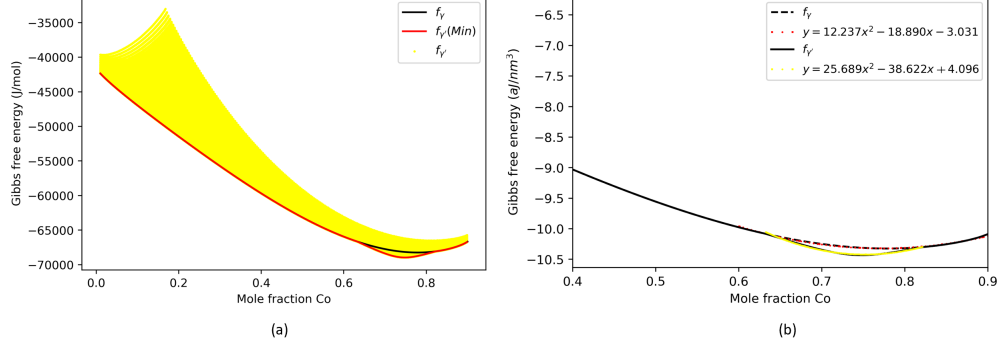


Figure 3: (a) The molar Gibbs free energy densities of γ and γ' phase, and (b) the volumetric Gibbs free energy densities of γ and γ' phase and the corresponding quadratic fits.

after the unit conversion and the quadratic fits are shown in Figure 3(b). The final simplified forms of the free energy densities are then

$$f_{\gamma}(c_{\gamma}) = 12.24c_{\gamma}^2 - 18.89c_{\gamma} - 3.031 \quad (10)$$

and

$$f_{\gamma'}(c_{\gamma'}) = 25.69c_{\gamma'}^2 - 38.62c_{\gamma'} + 4.096. \quad (11)$$

2.2. Elastic energy

We apply the same approach to include f_{elas} as in Jokisaari et al. [17]. The elastic energy density is given as

$$f_{\text{elas}} = \frac{1}{2} \sigma_{ij} \epsilon_{ij}^{\text{elas}}, \quad (12)$$

where we use the Einstein summation convention over Cartesian indices i, j, k, l , $\sigma_{ij} = C_{ijkl}(\eta) \epsilon_{kl}^{\text{elas}}$ is the elastic stress, $\epsilon_{ij}^{\text{elas}}$ is the elastic strain, and $C_{ijkl}(\eta)$ is the elastic stiffness tensor which is interpolated smoothly from one phase to the other across the diffuse interface:

$$C_{ijkl}(\eta) = C_{ijkl}^{\gamma}(1 - h(\eta)) + C_{ijkl}^{\gamma'}h(\eta). \quad (13)$$

The lattice misfit strain needs to be considered when the lattice parameters of the two phases are different [25],

$$\epsilon_{ij}^{\text{elas}} = \epsilon_{ij}^{\text{total}} - \epsilon_{ij}^{\text{misfit}}h(\eta), \quad (14)$$

and the total strain is calculated from the displacement fields u_j as in [25]

$$\epsilon_{ij}^{\text{total}} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right). \quad (15)$$

Here, $\epsilon_{ij}^{\text{misfit}}$ is the crystallographic misfit strain tensor between the γ and γ' phases defined with respect to the γ phase. We do not include an explicit composition dependence in the strain as the elastic energy is mainly induced by the misfit strain between the two phases, which is a structural difference that is described by the phase field order parameter.

2.3. Kinetics

The equations of motion for this model are given by the Allen-Cahn equation for the non-conserved phase field order parameter η and the Fick's law of diffusion equation for the conserved composition field variable c , respectively:

$$\frac{\partial \eta}{\partial t} = -L \frac{\delta \mathcal{F}}{\delta \eta} \quad (16)$$

$$\frac{\partial c}{\partial t} = \nabla \cdot \left(M \nabla \frac{\delta \mathcal{F}}{\delta c} \right) \quad (17)$$

where L is the kinetic coefficient that governs the time evolution of the phase field η , M is the mobility that controls the kinetics of the composition field. The chemical potentials μ_η and μ_c are given by

$$\mu_\eta = \frac{\delta \mathcal{F}}{\delta \eta} = \frac{\partial f_{\text{chemical}}}{\partial \eta} + \omega \frac{dg}{d\eta} + \frac{\partial f_{\text{elas}}}{\partial \eta} - \nabla \cdot (\kappa \nabla \eta) \quad (18)$$

$$\mu_c = \frac{\delta \mathcal{F}}{\delta c} = \frac{\partial f_{\text{chemical}}}{\partial c} \quad (19)$$

The mobility of Co in the γ phase can be derived from the CALPHAD kinetic database, which is a function of Co composition, ranging from 0 to 0.9, and temperature (fixed at 900 °C in this work). We assume that it stays the same in the γ' phase since mobility data for γ' phase are, to the best of our knowledge, not presently available. Following Campbell [26], we define the atomic mobility of element $A \in \{\text{Al}, \text{Co}, \text{W}\}$ in terms of absolute reaction rate theory [27] as

$$M_A = \exp \left(\frac{-\Delta Q'_A}{RT} \right) \frac{1}{RT}, \quad (20)$$

where R is the gas constant, T is temperature, and $\Delta Q'_A$ is an activation energy, which is composition-dependent. The activation energy $\Delta Q'_A$ can be expanded as:

$$\Delta Q'_A = c_{\text{Al}} Q_A^{\text{Al}} + c_{\text{Co}} Q_A^{\text{Co}} + c_{\text{W}} Q_A^{\text{W}}, \quad (21)$$

where Q_A^B is the self-activation energy for $B \in \{\text{Al}, \text{Co}, \text{W}\}$ in pure A , which can be obtained from the CALPHAD kinetic database [28].

We note that the atomic mobility data provided by CALPHAD databases are based on a lattice frame of reference, while phase field methods adopt a volume-fixed frame of reference, therefore requiring transformation of mobility expressions before they can be used in our phase field model. The transformed Co mobility M for the phase field model can be computed using [29]

$$M = V_m ((1 - c)^2 M_{\text{Co}} + c^2 M_{\text{Al}} + c^2 M_{\text{W}}), \quad (22)$$

Here the Co mobility is calculated based on the ternary system database. V_m is the molar volume which can be obtained from Pyczak et al. [30]. M_{Al} and M_{W} are different as calculated from Eq. 20. It is interesting that the composition of Al is the same in the two phases as shown in Fig. 1, despite the fact that $M_{\text{Al}} > M_{\text{W}}$. Figure 4 illustrates how the mobility of Co changes with the Co composition after the unit conversion. A fourth-order polynomial

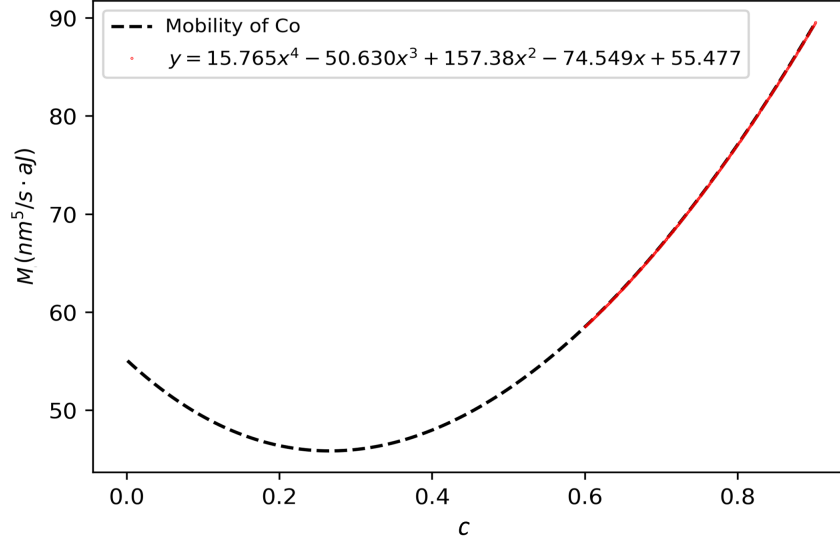


Figure 4: The mobility of Co and the corresponding fourth-order polynomial fit.

fit is made for the mobility data at $c > 0.6$ since this is the relevant composition range for Co-rich Co-Al-W alloys. If we make an estimate of reasonable values for scales from the simulation system, e.g., $\lambda = 5$ nm for the interface between the precipitate and the matrix, the composition change from γ to γ' phase will be approximately 0.068 and the energy density change will be approximately 0.15 aJ nm^{-3} . The mobility in this case is about $80 \text{ nm}^5 \text{ s}^{-1} \text{ aJ}^{-1}$, and the time scale for the mobility driving this composition change according to the discrete form of Eq. (17) is of the order of 0.01 s.

2.4. Interfacial and elastic energy densities

In addition to the chemical free energy density and mobilities that we extracted from the CALPHAD database, we need realistic models for the interfacial and elastic free energies, as well as for the phase field double-well potential. We use a combination of experimental and modeling data [17] to construct this, as illustrated in Fig. 5.

Information about the interface, such as the interfacial energy σ and interfacial width λ , is needed to get a reasonable value for the barrier height ω of the double-well potential energy density and for the gradient energy density coefficient κ used in the phase field model. The interfacial energy obtained from the density functional theory (DFT) calculation at 0 K is 98 mJ m^{-2} [17]. We assume that the interfacial energy at 900 °C stays the same, but note that thermal effects may be significant. The interfacial width is about 5 nm as measured by 3D-APT [7]. We can then obtain ω and κ from the following relations [19]:

$$\sigma = \frac{\kappa\sqrt{\omega}}{3\sqrt{2}} \quad (23)$$

$$2\lambda = 2.94\sqrt{2}\frac{\kappa}{\sqrt{\omega}}. \quad (24)$$

Table 1: The Latin hypercube used for parameter sensitivity analysis in this work. Sample No. 0 uses the initial parameterization. Parameters in other samples are chosen around the initial parameters in sample No. 0. c_0 is the initial mole fraction, ω is the barrier height, κ is the gradient energy coefficient, ϵ_{misfit} is the misfit strain, R_0 is the initial radius of the precipitate and L is the phase field kinetic coefficient.

Sample No.	c_0	ω (aJ nm ⁻³)	κ (aJ nm ⁻¹)	ϵ_{misfit} (%)	R_0 (nm)	L (nm ³ s ⁻¹ aJ ⁻¹)
0	0.830	0.173	1.000	0.500	75.00	100.0
1	0.849	0.128	1.366	0.545	95.93	55.37
2	0.803	0.161	1.180	0.324	25.38	87.21
3	0.834	0.297	0.699	0.852	184.6	119.0
4	0.843	0.191	0.762	0.265	53.12	184.4
5	0.860	0.314	0.607	0.990	120.0	66.92
6	0.841	0.209	1.007	0.757	175.6	76.32
7	0.823	0.111	0.535	0.424	67.22	60.26
8	0.817	0.141	1.834	0.474	77.51	114.0
9	0.829	0.096	1.592	0.373	45.22	155.4
10	0.808	0.251	0.995	0.595	34.66	150.0

This yields $\omega = 0.173 \text{ aJ nm}^{-3}$, and $\kappa = 1.0 \text{ aJ nm}^{-1}$ for our initial parameterization. Although we are using a different form of the double-well function than the usual quartic function, the shape of the 10th-order double-well function, especially the the distance between the two wells and the height of the barrier, follows closely the shape of the common fourth-order double-well function, and the relations expressed in Eqs. (23) and (24) remain unchanged except for some insignificant correction factors.

The misfit strains of Co-based alloys have been reported in several works in the literature [3, 30, 31, 32]. Usually the misfit strain is dependent on the temperature and alloying elements. There is an appreciable variation in the reported data from 0.1 % to 0.9 %, and we choose $\epsilon_{11}^{\text{misfit}} = \epsilon_{22}^{\text{misfit}} = \epsilon_{33}^{\text{misfit}} = 0.5 \%$ for our initial parameterization, and we will use the notation ϵ_{misfit} for simplicity in the following. We use the same elastic stiffness tensor as in Jokisaari et al. [17].

To study how the parameterization could influence the size and shape of the γ' precipitate we build a Latin hypercube as shown in Table 1 and carry out a sensitivity analysis for parameters including the above mentioned barrier height of double-well potential (ω), the gradient energy coefficient (κ), the misfit strain (ϵ_{misfit}), as well as the initial composition of Co in the alloy (c_0), the initial radius of the precipitate (R_0) and the kinetic coefficient in Eq. (16) (L). Notice that we choose $c_0 = (c_{\gamma}^{\text{eq}} + c_{\gamma'}^{\text{eq}})/2 = 0.830$, $R_0 = 75 \text{ nm}$, and $L = 100.0 \text{ nm}^3 \text{ s}^{-1} \text{ aJ}^{-1}$ for our initial parameterization. All these compositions are within the two-phase region, i.e., $c_{\gamma'}^{\text{eq}} < c_0 < c_{\gamma}^{\text{eq}}$.

2.5. Initial conditions

As initial condition we use a spherical precipitate of radius R_0 for the structural order parameter η . Following Jokisaari et al. [17], we set the initial value of η in the matrix γ phase to be slightly greater than zero, e.g., 0.005, and the initial value of η in the precipitate γ' phase is set to be unity. The two phases are connected through a smooth interface of

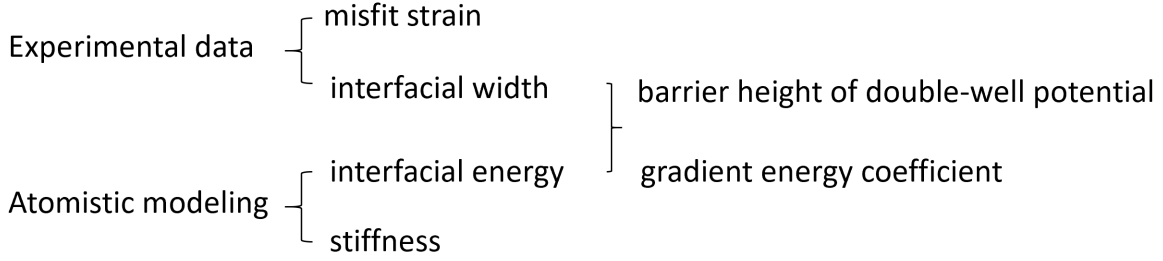


Figure 5: A diagram that illustrates the data that we are gathering from the experiments and atomistic modeling. The phase field modeling parameters such as barrier height of double-well potential and gradient energy coefficient can be derived from the interfacial width and interfacial energy.

width ℓ , which is set to be 5 nm when $R_0 \leq 100$ nm, and $\ell = R_0/20$ when $R_0 > 100$ nm to ensure a large ratio of the bulk to interfacial regions [17],

$$\eta_0(\mathbf{r}) = \begin{cases} 1 & |\mathbf{r}| \leq R_0 - \frac{\ell}{2} \\ 0.005 + \frac{1}{2}(1 - 0.005) \left(1 + \cos\left(\pi \frac{|\mathbf{r} - R_0 + \frac{\ell}{2}}{\ell}\right) \right) & R_0 - \frac{\ell}{2} \leq |\mathbf{r}| \leq R_0 + \frac{\ell}{2} \\ 0.005 & |\mathbf{r}| \geq R_0 + \frac{\ell}{2} \end{cases} \quad (25)$$

For the composition field variable c we use a nearly uniform initial condition c_0 , with an added small random perturbation of ± 0.001 drawn from a uniform distribution.

3. Numerical methods

Aagesen et al. first implemented the KKS model within the Multiphysics Object-Oriented Simulation Environment (MOOSE) framework [33, 34, 35] to study the U-Si system [36] and quantify elastic energy effects on interfacial energy [37]. We use a similar application within the MOOSE framework for our simulations on Co-based superalloys. The computational domains are meshed using cubic eight-node hexahedral elements. First-order Lagrange shape functions are employed for all nonlinear variables. The system of nonlinear equations is solved with the full Newton method that uses the single-matrix preconditioning with the additive Schwarz preconditioner and ILU¹ sub-preconditioning. The second backward differentiation formula (BDF2) time integration scheme is applied. The simulations are solved with a nonlinear relative tolerance of 10^{-6} and a nonlinear absolute tolerance of 10^{-9} . A maximum of 100 nonlinear iterations per solve and a maximum of 10 000 linear iterations are specified.

A computational domain of size 750 nm $\times$ 750 nm $\times$ 750 nm is used for all the simulations in the present work, unless the precipitate keeps growing until it engulfs the entire computational domain. The domain is large enough that the domain boundaries do not influence the precipitate size or morphology. We note that an alternative model using the volume-preserved algorithm [38] can be used when the size of the computational domain is an issue. To improve computational efficiency we employ adaptive meshing and adaptive

¹Incomplete factorizations based on the computation of triangular factors L and U (ILU).

time stepping, and also exploit the symmetry of the system. The computational domain of $750\text{ nm} \times 750\text{ nm} \times 750\text{ nm}$ is first meshed into $32 \times 32 \times 32$ elements. This translates to an element size of $\Delta x = 23.4\text{ nm}$. Gradient jump indicators [39] for c , η , and three displacement variables u_i are used to determine mesh adaptivity. A single element can be refined up to three times for adaptive meshing. Thus, the finest part of the mesh has elements with size $\Delta x = 2.93\text{ nm}$. The interface thickness extracted from 3D-APT experiment is about 5 nm , the Δx we used in our 3D simulations is approximately the same quantity. However, we have compared our 3D simulations with 2D simulations that could use a much finer mesh of element size down to $\Delta x = 0.9375\text{ nm}$. Based on this comparison, we concluded that the resolution used for the 3D simulations is fine enough to yield meaningful results. The initial time step Δt is set to 1 % of the nominal unit time, and the time step is allowed to grow by 5 % per step. We utilize the cubic symmetry of the system and arrange the precipitates such that the mirror boundaries of the computational domain are along the symmetry axes of the arrangement. In this way only one-eighth of the system is simulated and mirror boundary conditions are applied to the symmetry planes. Following Jokisaari et al. [17], we use symmetry to impose boundary conditions on the displacements that eliminate rigid body translations. Specifically, the displacements are pinned on the (100), (010), and (001) symmetry planes, respectively, so that the displacements on the three symmetry planes are zero, and zero normal-derivative boundary conditions are applied to other variables. [We confirmed that the mirror boundary conditions do not alter the results by comparing the results of a 2D simulation performed with mirror boundary conditions to one which modeled the entire precipitate and matrix without mirror boundary conditions.](#)

The system of partial differential equations (PDEs) is composed of the evolution of the order parameter and composition variable (Eqs. (16) and (17)), the constraint equations required for the KKS model (Eqs. (3) and (4)) and the time-independent mechanical equilibrium equation. To avoid prohibitively expensive fourth-order derivative operators in 3D, we split the fourth-order diffusion equation into two second-order equations [40, 41], such that Eqs. (17) and (19) are solved separately with two different nonlinear variables c and μ_c . The complete set of governing equations is then given by

$$\begin{cases} \frac{\partial \eta}{\partial t} = -L \left(\frac{\partial f_{\text{chemical}}}{\partial \eta} + \omega \frac{dg}{d\eta} + \frac{\partial f_{\text{elas}}}{\partial \eta} - \nabla \cdot (\kappa \nabla \eta) \right) \end{cases} \quad (26)$$

$$\begin{cases} \frac{\partial c}{\partial t} = \nabla \cdot (M \nabla \mu_c) \end{cases} \quad (27)$$

$$\begin{cases} \mu_c = \frac{\partial f_{\text{chemical}}}{\partial c} = \frac{df_{\gamma}}{dc_{\gamma}} \end{cases} \quad (28)$$

$$\begin{cases} \frac{df_{\gamma}}{dc_{\gamma}} = \frac{df_{\gamma'}}{dc_{\gamma'}} \end{cases} \quad (29)$$

$$\begin{cases} c = (1 - h(\eta))c_{\gamma} + h(\eta)c_{\gamma'} \end{cases} \quad (30)$$

$$\begin{cases} \nabla \cdot \sigma_{ij} = 0 \end{cases} \quad (31)$$

We use ordinary derivatives for the chemical free energy densities of γ and γ' phases as they depend only on the composition of γ and γ' , respectively.

We next outline how this system of equations can be solved via the finite element method (FEM). In the FEM weak formulation a variable X is expanded in FEM shape functions

ϕ_n , $X = \sum_n X_n \phi_n$ with X_n expansion coefficients, and we construct a residual equation (set to zero) for each equation. An accurate Jacobian matrix is then computed for an efficient Newton solve. The full list of residual and Jacobian terms are shown in the Appendix A. Part of the derivation of these terms can be found in documentation for the MOOSE KKS model [42]; the rest, which is related to the displacement variables and elastic energy density, is provided in Appendix B.

4. Results and discussion

In this section, we discuss the equilibrium shapes of Co-Al-W γ' precipitates and how the parameters influence the morphology. We will here focus only on single precipitates.

4.1. Morphology evolution

We first examine the morphology evolution of the γ' precipitate in samples No. 3, No. 4 and No. 5 among the 11 samples (see Table 1). We pick these samples because they are not only representative but also have reached an equilibrium state (defined as the change of the total energy over consecutive time steps is less than 0.001 aJ) before the precipitate grows too big and interacts with the system boundary. For reference, the total energy of the system is about -4.3×10^9 aJ and Δt before the equilibrium state is around 1 s. The modeled precipitate evolution over time is shown in Figure 6 for these three samples. These times are in arbitrary units and the displayed times serve the purpose of a relative comparison of precipitate morphology evolution under different sets of parameters. Note that the precipitates in the initial configurations have jagged edges because the mesh adaptivity is not employed at $t = 0$. During the first time step the adaptive mesh refinement is engaged and smooths out the precipitate edges. The precipitates grow in both samples No. 3 and No. 4. However, their growth processes are not the same, and the final equilibrium shapes of the precipitates also differ. In sample No. 3, the precipitate does not grow much in size, and the shape-change occurs in the beginning of the evolution process. The corners of the precipitates become sharper, and the precipitate eventually evolves into a sharp cubic shape at $t = 20$; the precipitate then maintains this cubic shape as its size increases with time. In sample No. 4, the size of the precipitate increases significantly even at the very early stages of evolution. At $t = 10$, the precipitate still has a spherical shape but it has grown significantly larger compared to its initial size. Subsequently, the precipitate keeps growing and it starts to form corners. Finally, it stops growing in size and becomes more and more cuboidal, although it does not grow into a cubic shape with sharp corners like sample No. 3. The main difference of the parameters in these two set samples are the misfit strain and initial radius: sample No. 3 has a larger misfit strain 0.852 % compared to the 0.265 % misfit strain for sample No. 4, so the elastic energy is greater than that in sample No. 4 which results in a more cuboidal shape. The precipitate in sample No. 3 also has a much larger initial radius and it is of interest to determine whether or not the initial radius of the precipitate has a large effect on the final size and shape of the precipitate. In contrast, the evolution of sample No. 5 is strikingly different. Apart from the highest misfit strain, it has a larger initial composition c_0 which indicates that the chemical free energy favors the γ matrix phase. As a result, the precipitate dissolves into the matrix in this case. The remarkably different morphology evolutions that are the result of seemingly minor differences

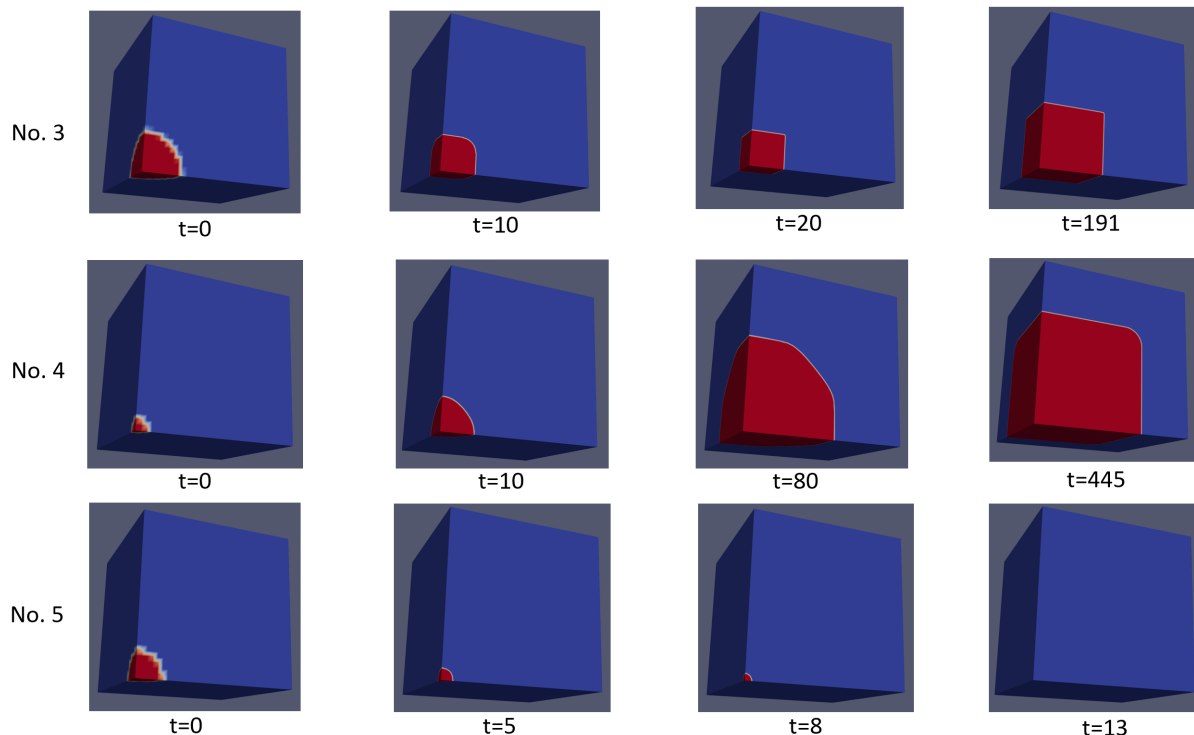


Figure 6: The morphology evolution of the precipitate with three different sets of parameters. All times are in arbitrary units and the displayed times serve the purpose of a relative comparison of precipitate morphology evolution under different sets of parameters. At $t = 0$, the precipitates have jagged edges because the mesh adaptivity is not engaged at $t = 0$ and the mesh at the interface has low resolution. Immediately when the time-evolution starts the mesh adaptivity is engaged and the precipitate shape and interface become smooth. The last column shows the final equilibrium shape of the precipitates. For sample No. 3, the precipitate evolves to a cuboid with sharp corners, while for sample No. 4 the cuboid has rounded corners and grows larger. In contrast, for sample No. 5 the precipitate dissolves.

in parameters indicate a sensitive dependence on at least some of the input parameters. This motivates us to perform a more detailed sensitivity analysis.

In Jokisaari et al. [17] it was shown that the equilibrium shape is determined by the interplay of the interfacial energy and the elastic energy, which were the only two energy components in that work. In contrast, here we also include the chemical energy in our phase field model, as well as conserved diffusion kinetics of the composition field, and find that the competition between all three energies influences the morphology evolution and equilibrium shape of precipitate. The chemical energy depends mainly on the initial composition c_0 , the interfacial energy depends on the barrier height ω of the double-well potential and the gradient energy coefficient κ , and the misfit strain is an important parameter that controls the elastic energy. (The elastic energy also depends on the elastic stiffness but it has been shown that this dependence is much weaker than on the misfit strain. [17]) Changing certain parameters within experimental range gives rise to large changes in morphology evolution.

4.2. Sensitivity analysis

In order to have a better understanding of how the equilibrium size and shape of the precipitate depend on the system parameters, we carry out a sensitivity analysis on parameters c_0 , ω , κ , ϵ_{misfit} , R_0 and L . Such an analysis can help correlate modeling and experiments by providing bounds on parameters based on observed modeled and experimental behavior. It also can provide guidance for further development of Co-based superalloys by providing insight into which parameters have the largest effect on the morphology evolution. First of all, we can effectively reduce the dimension of the parameter space: we find that if restricted in a reasonable range, R_0 and L have little influence on the precipitate size or shape, so these parameters can be ignored in the sensitivity analysis. Figure 7 shows the equilibrium configuration of the precipitate with different R_0 and L while keeping the other parameters the same as those for sample No. 1. In order to accurately represent size differences, we measured the volume of precipitates in equilibrium by integrating the order parameter η over the entire volume. It can be clearly seen in the first four subfigures of Fig. 7 that with R_0 ranging from 23.98 nm to 191.9 nm, the equilibrium size and shape of the precipitate stay the same (with a maximum volume change of 1.8 %), due to the main factor that influences the equilibrium size being the initial composition of Co in the alloy. The precipitates will grow to the expected equilibrium size no matter what the initial size of precipitates is. The factors that influence the equilibrium shape are a little more complicated and will be discussed later. Similarly, the last five subfigures (bottom row) of Fig. 7 shows the equilibrium configuration of the precipitate with different L while keeping the other parameters the same as those for sample No. 1. The equilibrium precipitate stays the same (with a maximum volume change of 0.87 %) for L within the range of $25 \text{ nm}^3 \text{ s}^{-1} \text{ aJ}^{-1}$ to $200 \text{ nm}^3 \text{ s}^{-1} \text{ aJ}^{-1}$, where the system is in a diffusion-controlled regime and the morphology evolution is rather insensitive to L . However, when L increases to $400 \text{ nm}^3 \text{ s}^{-1} \text{ aJ}^{-1}$, the morphology of the precipitate begins to change, because the kinetic coefficient is now large enough that the evolution of the non-conserved order parameter η is much more rapid than that of the composition field c . This leads to a spatial distortion of the chemical potential density in the $\langle 110 \rangle$ directions, which drives a non-physical dendritic-like precipitate elongation in these directions, and is not compatible with experimental observations of Co-based superalloys.

As long as we keep the magnitude of L in a range of $25 \text{ nm}^3 \text{ s}^{-1} \text{ aJ}^{-1}$ to $200 \text{ nm}^3 \text{ s}^{-1} \text{ aJ}^{-1}$, L will not have an appreciable influence on the equilibrium size or shape of the precipitate and we avoid non-physical effects driven by a large L .

If we fix all other parameters and only allow c_0 to vary in sample No. 1, we find that a small initial composition field c_0 results in a final pure γ' phase, while a large c_0 will cause the γ' phase precipitate to dissolve, as shown in Fig. 8 (Row 1). We observe that c_0 has a strong influence on the bulk chemical free energy density, and a large c_0 will then result in the prevalence of the phase favored by the bulk chemical free energy density. Moreover, the composition c is still in the two-phase region and there could be a shift in the phase diagram caused by stress and interfacial energy. This result agrees with the finding in Pyczak et al. [30] and Wang et al. [18], where a higher volume fraction of γ' precipitates is found with increasing W composition while the Al composition stays the same, i.e., decreasing c_0 . Next,

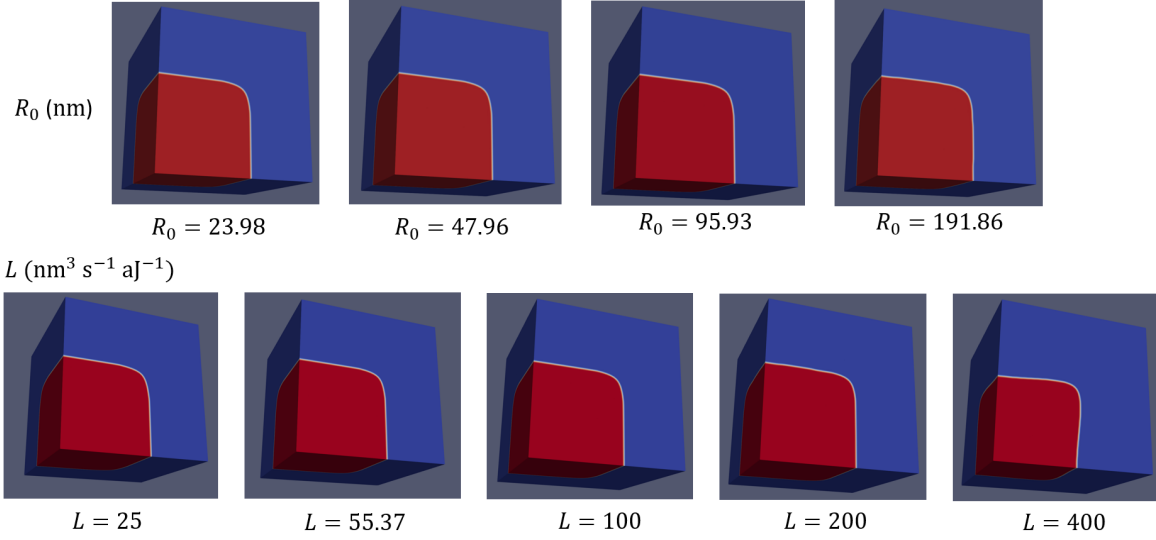


Figure 7: The equilibrium configuration of precipitates with different initial radius R_0 (nm) (top row), and different kinetic coefficients L ($\text{nm}^3 \text{s}^{-1} \text{aJ}^{-1}$) (bottom row). The other parameters are the same as those for sample No. 1. The volume for precipitates with different R_0 are 7.6482×10^7 , 7.6495×10^7 , 7.6472×10^7 and 7.5108×10^7 , respectively. The volume for precipitates with different L are 7.6774×10^7 , 7.6472×10^7 , 7.6513×10^7 , 7.6106×10^7 and 5.7913×10^7 , respectively.

we fix all parameters except for the barrier ω of the double-well potential in sample No. 1. We use ω_1 to represent the original ω value before variation. The final precipitates for different values of ω are shown in Fig. 8 (Row 2). We see in this figure that ω does not have as much influence as c_0 on the equilibrium size of the precipitate when ω is within the range from $0.5\omega_1$ to $2\omega_1$. However, the precipitate will tend to stay in its original size when ω becomes large, i.e., $4\omega_1$, as indicated in the final panel of the second row in Fig. 8. The different volume fractions are a result of the Gibbs-Thomson effect (interfacial energy) generating a shift in the phase diagram. This result agrees very well with the findings from Vorontsov et al. [43]. From Eq. (24) we can see that a large ω generally results in a small interfacial width. Vorontsov et al. [43] found that compared to the average interfacial width of Co-7Al-7W-20Ni (1.250 nm) and Co-10Al-5W-2Ta (2.139 nm), Co-7Al-7W has a much smaller interfacial width (0.460 nm) and the change of its precipitate size is much smaller compared to the other two alloys when the aging time increases. Next, we only allow κ to change and fix all other parameters in sample No. 1; the corresponding equilibrium configurations are shown in Fig. 8 (Row 3). Similar to ω , κ does not have a strong influence on the precipitate size. Small κ results in a sharply cuboidal precipitate shape because as κ increases, the interfacial energy grows and becomes dominant in determining the precipitate shape, and this drives the shape to be more spherical in order to minimize the interfacial energy. Finally, we change ϵ_{misfit} and keep the rest of the parameters in sample No. 1 unchanged. The result is shown in Fig. 8 (Row 4). Contrary to κ , small ϵ_{misfit} results in spherical precipitate shape while large ϵ_{misfit} results in a more cuboidal shape. This is because with increasing ϵ_{misfit} , the elastic energy becomes dominant in determining the precipitate shape, which drives a cuboidal shape. In Ref. [43], the Co-10Al-5W-2Ta superalloy has the highest ϵ_{misfit} (0.67 %) and the precipitate shape of it is the most cuboidal. The precipitate shape of the Co-

7Al-7W-20Ni superalloy that has a small ϵ_{misfit} (0.35 %) is not that spherical though, which indicates that the precipitate shape is a complicated property and can depend on many parameters, not solely on ϵ_{misfit} . In addition, when ϵ_{misfit} becomes really large, a shift in the phase diagram could occur, and the precipitate will dissolve into the matrix to minimize the total energy as the cost in elastic energy to maintain a precipitate becomes prohibitive.

As a result, we can reduce our parameter set in the sensitivity analysis to only four parameters: c_0 , ω , κ , ϵ_{misfit} . In order to quantitatively measure how strong the influences are, we fit a function of these input parameters and their cross terms to the equilibrium precipitate size (r_{mean}) and shape (k_{cubic}) after normalization, respectively, as shown in Eqs. (33) and (34). Here, r_{mean} is defined as the average distance from the interface ($0.3 < \phi < 0.7$) to the center of the precipitate²; k_{cubic} is defined as

$$k_{\text{cubic}} = 1 - \left| \frac{1}{\sqrt{3}} \frac{r_{\text{max}}}{r_{\text{min}}} - 1 \right|, \quad (32)$$

where r_{max} and r_{min} are the maximum and minimum distances from the interface to the center of the precipitate. The constant factor $1/\sqrt{3}$ is chosen so that k_{cubic} of a cube is unity. Any particle which is not cubic will have k_{cubic} less than 1. For example, a spherical precipitate that has $r_{\text{max}}/r_{\text{min}}$ ratio of 1 will result in a k_{cubic} equal to 0.58 and a dendritic precipitate that has $r_{\text{max}}/r_{\text{min}}$ ratio of 2 will result in a k_{cubic} equal to 0.85. Thus, the sensitivity analysis yields the following equations relating the precipitate mean size r_{mean} and shape k_{cubic} :

$$\begin{aligned} \bar{r}_{\text{mean}} = & a_1 \bar{c}_0 + a_2 \bar{\omega} + a_3 \bar{\kappa} + a_4 \bar{\epsilon}_{\text{misfit}} + a_5 \bar{c}_0 \bar{\omega} + a_6 \bar{c}_0 \bar{\kappa} \\ & + a_7 \bar{c}_0 \bar{\epsilon}_{\text{misfit}} + a_8 \bar{\omega} \bar{\kappa} + a_9 \bar{\omega} \bar{\epsilon}_{\text{misfit}} + a_{10} \bar{\kappa} \bar{\epsilon}_{\text{misfit}}, \end{aligned} \quad (33)$$

$$\begin{aligned} \bar{k}_{\text{cubic}} = & b_1 \bar{c}_0 + b_2 \bar{\omega} + b_3 \bar{\kappa} + b_4 \bar{\epsilon}_{\text{misfit}} + b_5 \bar{c}_0 \bar{\omega} + b_6 \bar{c}_0 \bar{\kappa} \\ & + b_7 \bar{c}_0 \bar{\epsilon}_{\text{misfit}} + b_8 \bar{\omega} \bar{\kappa} + b_9 \bar{\omega} \bar{\epsilon}_{\text{misfit}} + b_{10} \bar{\kappa} \bar{\epsilon}_{\text{misfit}}, \end{aligned} \quad (34)$$

with the coefficients a_i and b_i ($i = 1, 2, \dots, 10$) of Eqs. (33) and (34) given in Table 2. The bar notation is used for normalized quantities. ~~Instead of ω and κ we can express the sensitivity in terms of interfacial energy σ and interfacial width λ using Eq. 23 and 24, as in Eq. 35 and 36. The coefficients are illustrated in Fig. 9 to give us a clearer view of how these parameters influence the precipitate size and shape.~~

$$\begin{aligned} \bar{r}_{\text{mean}} = & -0.312 \bar{c}_0 - 0.467 \frac{\bar{\sigma}}{\bar{\lambda}} + 0.0286 \sqrt{\bar{\sigma} \bar{\lambda}} - 0.0494 \bar{\epsilon}_{\text{misfit}} - 2.07 c_0 \frac{\bar{\sigma}}{\bar{\lambda}} - 0.114 c_0 \sqrt{\bar{\sigma} \bar{\lambda}} \\ & - 0.268 \bar{c}_0 \bar{\epsilon}_{\text{misfit}} + 1.16 \sqrt{\frac{\bar{\sigma}^3}{\bar{\lambda}}} - 1.08 \epsilon_{\text{misfit}} \frac{\bar{\sigma}}{\bar{\lambda}} + 0.0909 \bar{\epsilon}_{\text{misfit}} \sqrt{\bar{\sigma} \bar{\lambda}}, \end{aligned} \quad (35)$$

² r_{mean} can also be defined as the spherical equivalent radius, which will result in a similar fitting result as shown in Appendix C.

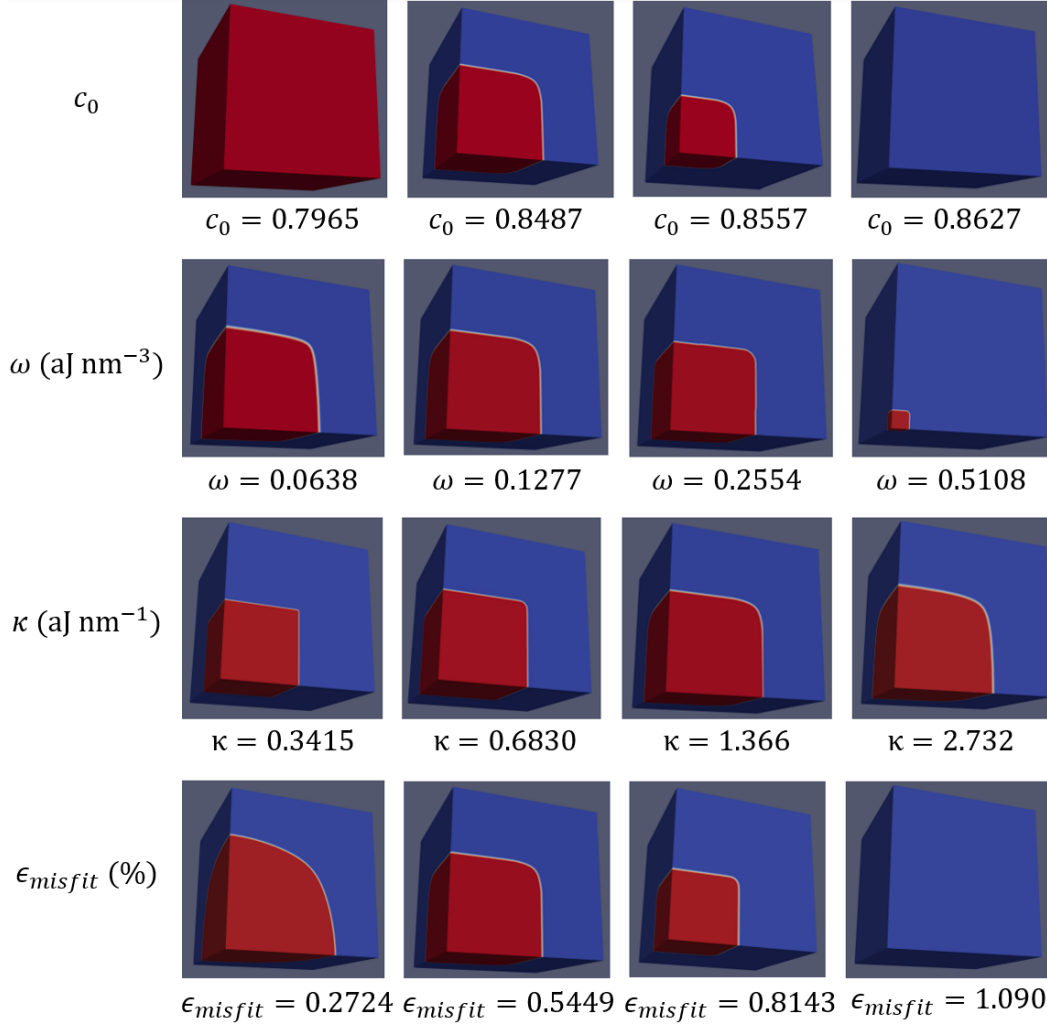


Figure 8: (Row 1) The equilibrium configurations of the precipitates for different initial composition c_0 of Co in the alloy. The volume of the precipitates are 4.2881×10^8 , 7.6472×10^7 , 1.8602×10^7 , and 0, respectively. (Row 2) The equilibrium configurations of the precipitates for different barrier height ω . The volume of the precipitates are 7.7976×10^7 , 7.6472×10^7 , 5.6098×10^7 , and 4.2074×10^5 , respectively. (Row 3) The equilibrium configurations of the precipitate for different gradient energy coefficients κ . The volume of the precipitates are 4.6178×10^7 , 7.0858×10^7 , 7.6472×10^7 , and 7.5077×10^7 , respectively. (Row 4) The equilibrium configurations of the precipitate for different misfit strain ϵ_{misfit} . The volume of the precipitates are 8.8410×10^7 , 7.6472×10^7 , 3.5234×10^7 , and 0, respectively. In Rows 1 - 4, the parameters other than c_0 (Row 1), ω (Row 2), κ (Row 3), and ϵ_{misfit} (Row 4) are kept the same as those for sample No. 1.

Table 2: The fitted coefficient values for the precipitate size and shape function used in Eqs. (33) and (34).

a_i	coef. value	a_i	coef. value	b_i	coef. value	b_i	coef. value
a_1	-0.312	a_6	-0.0797	b_1	-0.0484	b_6	0.433
a_2	-0.0529	a_7	-0.268	b_2	0.326	b_7	-0.281
a_3	0.0200	a_8	0.0921	b_3	-0.0784	b_8	-0.374
a_4	-0.0494	a_9	-0.122	b_4	0.129	b_9	0.595
a_5	-0.235	a_{10}	0.0636	b_5	-0.370	b_{10}	-0.322

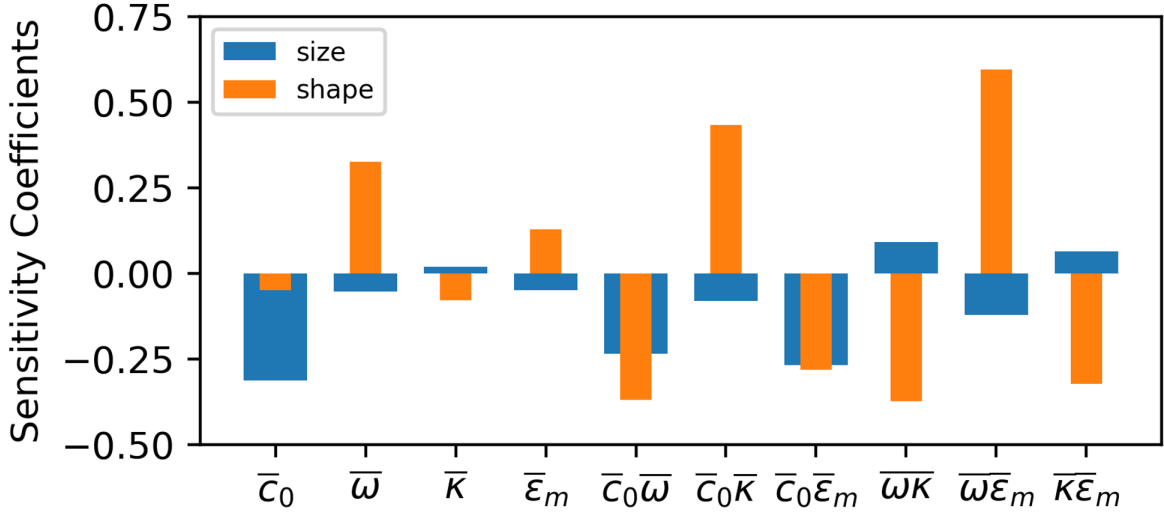


Figure 9: The sensitivity coefficients for the precipitate size and shape.

$$\begin{aligned}
\bar{k}_{\text{cubic}} = & -0.0484\bar{c}_0 + 2.87\frac{\bar{\sigma}}{\bar{\lambda}} - 0.112\sqrt{\bar{\sigma}\bar{\lambda}} + 0.129\bar{\epsilon}_{\text{misfit}} - 3.26c_0\frac{\bar{\sigma}}{\bar{\lambda}} + 0.619c_0\sqrt{\bar{\sigma}\bar{\lambda}} \\
& - 0.281\bar{c}_0\bar{\epsilon}_{\text{misfit}} - 4.72\sqrt{\frac{\bar{\sigma}^3}{\bar{\lambda}}} + 5.25\epsilon_{\text{misfit}}\frac{\bar{\sigma}}{\bar{\lambda}} - 0.460\bar{\epsilon}_{\text{misfit}}\sqrt{\bar{\sigma}\bar{\lambda}}.
\end{aligned} \tag{36}$$

Figure 9 gives a pictorial representation of the coefficients in Eq. 33 and 34, and the shape- and size-sensitivity to the parameters. The figure clearly shows an intricate and non-trivial sensitivity of the shape and size to the parameters. First of all, c_0 has a large negative sensitivity coefficient, and it plays an important role in determining the precipitate size. This tells us that the composition has to be carefully determined in making these alloys as even a small increase in the Co composition could cause the γ' precipitates to decrease in the size and even dissolve in the γ matrix. Similarly, a small decrease in the Co composition may lead to overgrowth of the γ' precipitates, which is detrimental to the yield strength of the superalloy. This can be interpreted in terms of the alloy composition and the equilibrium volume fraction of γ' phase. In our case, since we have only one particle, the size of the particle is representative of the equilibrium volume fraction. This is just a manifestation of mass conservation between the precipitate and matrix. However, the composition of the precipitate and matrix can be a function of the misfit strain, so the equilibrium volume fraction (or particle size) can vary with strain. The factors that could influence the precipitate shape are more complicated. We first look at the single terms. The composition field c_0 has a much weaker influence on the precipitate shape compared to its influence on the precipitate size. It is easy to understand that κ has a (small) negative sensitivity coefficient. As mentioned above, κ is the gradient energy coefficient, and therefore an increase in κ will magnify the influence of the interfacial energy and it will be energetically more costly in terms of interfacial energy for the precipitates to evolve to cuboidal shapes. In contrast, ϵ_{misfit} has a positive sensitivity coefficient since increasing the misfit strain will make the elastic energy drive the precipitate shape more cuboidal. The cross terms all have

significant impacts on the precipitate shape. However, since we have established that c_0 is restricted to a small range in order to achieve a reasonable precipitate size, we will here not discuss cross terms in c_0 . Recall that according to Eq. (23), the product $\omega\kappa$ governs the magnitude of the interfacial energy, which is the reason why this cross term has a large negative sensitivity coefficient: when this cross term grows, so does the interfacial energy, which favors a more spherical shape. The cross term $\omega\epsilon_{\text{misfit}}$ has the largest positive sensitivity coefficient, which means that the interaction between a large misfit and a large barrier height (which has an impact on the interfacial energy, and thus influences the balance between the interfacial and elastic energies) will make the precipitate more likely to become cuboidal. This analysis gives us some useful insights. For example, a key parameter to drive and maintain a cuboidal shape, important for yield strength, is the misfit strain. This suggests that elements that increase the misfit strain and are energetically favored in the precipitate γ' phase could be added in order to obtain cuboidal precipitates and enhance the strength of the superalloy. When the temperature increases, thermal motion tends to decrease the misfit strain [30]. A potential path to maintaining cuboidal shape is then to decrease the interfacial energy between the γ and γ' phase in order to maintain alloy strength. Future work will further refine this sensitivity analysis as well as uncertainty quantifications.

5. Conclusion

We have built a realistic quantitative phase field model using thermodynamics from a CALPHAD database as well as other experimental or first-principles data to study the driving forces for the precipitate morphology evolution and shape instabilities on Co-based γ/γ' superalloys. Our work gives insight into how the competition between chemical, interfacial, and elastic energies influences the morphology evolution and equilibrium shape of precipitates. Our study also shows how changing certain parameters within reasonable bounds gives rise to large changes in the morphology evolution. A central part of our work is a sensitivity analysis to study quantitatively how the system parameters influence the size and shape of the precipitate. We found that the initial radius of the precipitate and the kinetic coefficients have little influence on the equilibrium size or shape of the precipitate. The initial composition of Co in the alloy has the strongest influence on the equilibrium size of the precipitate, while the cross term of the barrier height and misfit strain have a large influence on the shape of the precipitate. We hope to give some insights to the experimental design as well as guidance for lower-scale modeling, such as density functional theory (DFT). The composition of Co needs to be carefully determined to achieve a reasonable size of γ' precipitates and thus the desired yield strength. A small increase in the Co composition could cause the γ' precipitates to decrease in the and even dissolve in the γ matrix. Similarly, a small decrease in the Co composition may lead to overgrowth of the γ' precipitates, which is also detrimental to the yield strength of the superalloy. Furthermore, great attention needs to be paid when adding elements to the superalloy. For example, Cr is often added to the Co-based superalloy to enhance its oxidation resistance. However, it partitions strongly to the γ phase which results in a decrease of the misfit strain and hindrance of the γ' coarsening behavior. To make up for the loss of alloy strength due to the decreasing misfit strain, one could add other elements like Ni, which lowers the interfacial energy ([43]) and maintains the driving force for coarsening and the cuboidal shape of the precipitates from a different

aspect. The barrier height ω is, at least in principle, accessible through a combination of interfacial width and interfacial energy and, by combining results from 3D-APT and/or DFT, better estimates and bounds on ω can be achieved. Conversely, the sensitivity analysis can give guidance to what the bounds on ω , and therefore on interfacial width and energy, need to be in order to maintain the desired microstructural evolution.

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Appendix A. List of Residual and Jacobian terms

The weighted integral residual projection is constructed using test function ψ_m and applying the divergence theorem to reduce the derivative order. [We include the elastic energy terms in our derivation which are not included in the MOOSE document \[42\].](#) Thus Eq. (26) yields

$$R_\eta = \left(\frac{\partial \eta}{\partial t}, \psi_m \right) + (L\kappa \nabla \eta, \nabla \psi_m) + \left(-L \frac{dh}{d\eta} \left(f_\gamma - f_{\gamma'} - \frac{df_\gamma}{dc_\gamma} (c_\gamma - c_{\gamma'}) \right) + L\omega \frac{dg}{d\eta} + L \frac{\partial f_{\text{elas}}}{\partial \eta}, \psi_m \right) \quad (\text{A.1})$$

with zero flux on all boundaries. Here, (A, B) denotes the standard $\mathcal{L}^2$ inner product of A and B in a continuous Hilbert space. The Jacobian for variable η is:

$$\left\{ \begin{array}{l} \text{OnDiag } J_\eta^\eta = \left(\frac{(\partial \eta / \partial t)}{\partial \eta} \phi_n, \psi_m \right) + (L\kappa \nabla \phi_n, \nabla \psi_m) \\ \quad + \left(-L \frac{d^2 h}{d\eta^2} (f_\gamma - f_{\gamma'}) \phi_n + L\omega \frac{d^2 g}{d\eta^2} \phi_n + L \frac{\partial^2 f_{\text{elas}}}{\partial \eta^2} \phi_n, \psi_m \right) \\ \quad + \left(L \frac{d^2 h}{d\eta^2} \frac{df_\gamma}{dc_\gamma} (c_\gamma - c_{\gamma'}) \phi_n, \psi_m \right) \\ \text{OffDiag } J_\eta^{c_\gamma} = \left(-L \frac{dh}{d\eta} \frac{df_\gamma}{dc_\gamma} \phi_n, \psi_m \right) + \left(L \frac{dh}{d\eta} \left(\frac{d^2 f_\gamma}{dc_\gamma^2} (c_\gamma - c_{\gamma'}) + \frac{df_\gamma}{dc_\gamma} \right) \phi_n, \psi_m \right) \\ \text{OffDiag } J_\eta^{c_{\gamma'}} = \left(L \frac{dh}{d\eta} \frac{df_{\gamma'}}{dc_{\gamma'}} \phi_n, \psi_m \right) + \left(-L \frac{dh}{d\eta} \frac{df_\gamma}{dc_\gamma} \phi_n, \psi_m \right) \\ \text{OffDiag } J_\eta^{u_{i'n}} = \left(L \frac{\partial^2 f_{\text{elas}}}{\partial \eta \partial u_{i'n}}, \psi_m \right) = \left(LC_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{\text{el}}}{\partial \eta} + L \frac{\partial C_{i'jkl}}{\partial \eta} \frac{\partial \phi_n^{(i')}}{\partial x_j} \epsilon_{kl}^{\text{el}}, \psi_m \right) \end{array} \right. \quad (\text{A.2})$$

The notations for the displacement variables are set to be i' in order to be consistent with the derivation in the Appendix. The residual for the variable μ_c is derived from Eq. (27) as

$$R_{\mu_c} = \left(\frac{\partial c}{\partial t}, \psi_m \right) + (M \nabla \mu_c, \nabla \psi_m), \quad (\text{A.3})$$

and the Jacobian for μ_c is:

$$\begin{cases} \text{OnDiag } J_{\mu_c}^{\mu_c} = (M \nabla \phi_n, \nabla \psi_m) \\ \text{OffDiag } J_{\mu_c}^c = \left(\frac{\partial(\partial c / \partial t)}{\partial c} \phi_n, \psi_m \right) \end{cases} \quad (\text{A.4})$$

The residual for variable c derived from Eq. (28) is

$$R_c = \left(\frac{\partial f_\gamma}{\partial c_\gamma} - \mu_c, \psi_m \right), \quad (\text{A.5})$$

with a Jacobian for c as

$$\begin{cases} \text{OnDiag } J_c^c = 0 \\ \text{OffDiag } J_c^{c_\gamma} = \left(\frac{d^2 f_\gamma}{dc_\gamma^2} \phi_n, \psi_m \right) \\ \text{OffDiag } J_c^{\mu_c} = (-\phi_n, \psi_m) \end{cases} \quad (\text{A.6})$$

The residual for variable c_γ derived from Eq. (29) is

$$R_{c_\gamma} = \left(\frac{df_\gamma}{dc_\gamma} - \frac{df_{\gamma'}}{dc_{\gamma'}}, \psi_m \right) \quad (\text{A.7})$$

with the corresponding Jacobian for c_γ

$$\begin{cases} \text{OnDiag } J_{c_\gamma}^{c_\gamma} = \left(\frac{d^2 f_\gamma}{dc_\gamma^2} \phi_n, \psi_m \right) \\ \text{OffDiag } J_{c_\gamma}^{c_{\gamma'}} = \left(-\frac{d^2 f_{\gamma'}}{dc_{\gamma'}^2} \phi_n, \psi_m \right). \end{cases} \quad (\text{A.8})$$

The residual for variable $c_{\gamma'}$ derived from Eq. (30) and is

$$R_{c_{\gamma'}} = ((1 - h(\eta))c_\gamma + h(\eta)c_{\gamma'} - c, \psi_m), \quad (\text{A.9})$$

with a Jacobian for $c_{\gamma'}$

$$\begin{cases} \text{OnDiag } J_{c_{\gamma'}}^{c_{\gamma'}} = (h(\eta)\phi_n, \psi_m) \\ \text{OffDiag } J_{c_{\gamma'}}^{c_\gamma} = ((1 - h(\eta))\phi_n, \psi_m) \\ \text{OffDiag } J_{c_{\gamma'}}^c = (-\phi_n, \psi_m) \\ \text{OffDiag } J_{c_{\gamma'}}^\eta = \left(\frac{dh}{d\eta}(c_{\gamma'} - c_\gamma)\phi_n, \psi_m \right). \end{cases} \quad (\text{A.10})$$

Finally, the residual for the displacement variables u_i is derived from Eq. (31) as

$$R_{u_i} = \left(\sum_j \sigma_{ij}, \nabla \psi_m \right), \quad (\text{A.11})$$

with a Jacobian for u_i

$$\left\{ \begin{array}{l} \text{OnDiag } J_{u_i}^{u_i} = \left(\sum_j \frac{\partial \sigma_{ij}}{\partial u_i} \phi_n^{(i)}, \nabla \psi_m \right) \\ \text{OffDiag } J_{u_i}^\eta = \left(\sum_j \frac{\partial \sigma_{ij}}{\partial \eta} \phi_n, \nabla \psi_m \right) \\ \text{OffDiag } J_{u_i}^{u_{i''}} = \left(\sum_j \frac{\partial \sigma_{ij}}{\partial u_{i''}} \phi_n^{(i'')}, \nabla \psi_m \right) \quad (i'' \neq i). \end{array} \right. \quad (\text{A.12})$$

Here $u_{i''}$ means the other two displacement variables other than u_i ($i'' \neq i$).

Appendix B. Derivation of the Residual and Jacobian terms

This appendix shows the derivation of the elastic energy related Residual/Jacobian terms in Eqs. (A.1), (A.2) and Residual/Jacobian for displacement variables in Eqs. (A.11), (A.12). We have derived the entire Jacobian matrix in addition to the diagonal terms contained in the MOOSE document [42].

$$f_{\text{elas}} = \frac{1}{2} \sigma_{ij} \epsilon_{ij}^{el} \quad (\text{B.1})$$

$$\sigma_{ij} = C_{ijkl} \epsilon_{kl}^{el} \quad (\text{B.2})$$

$$C_{ijkl} = C_{ijkl}^\gamma [1 - h(\eta)] + C_{ijkl}^{\gamma'} h(\eta) \quad (\text{B.3})$$

$$\frac{\partial C_{ijkl}}{\partial \eta} = -C_{ijkl}^\gamma \frac{dh}{d\eta} + C_{ijkl}^{\gamma'} \frac{dh}{d\eta} \quad (\text{B.4})$$

$$\epsilon_{ij}^{el} = \epsilon_{ij}^{tot} - \epsilon_{ij}^0 \quad (\text{B.5})$$

$$\epsilon_{ij}^{tot} = \frac{1}{2} \left[\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right] \quad (\text{B.6})$$

$$\epsilon_{ij}^0 = \epsilon^{\text{misfit}} h(\eta) \quad (\text{B.7})$$

$$\frac{\partial \epsilon_{ij}^{el}}{\partial \eta} = -\frac{\partial \epsilon_{ij}^0}{\partial \eta} = -\epsilon^{\text{misfit}} \frac{dh}{d\eta} = \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \quad (\text{B.8})$$

$$\frac{\partial f_{\text{elas}}}{\partial \eta} = \frac{1}{2} \frac{\partial (C_{ijkl} \epsilon_{kl}^{el} \epsilon_{ij}^{el})}{\partial \eta} = \frac{1}{2} \left(\frac{\partial C_{ijkl}}{\partial \eta} \epsilon_{kl}^{el} \epsilon_{ij}^{el} + C_{ijkl} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \epsilon_{ij}^{el} + C_{ijkl} \epsilon_{kl}^{el} \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} \right) \quad (\text{B.9})$$

Insert Eqs. (B.4),(B.8) into Eq. (B.9), and we get the elastic energy related residual term in Eq. (A.1):

$$\begin{aligned} \frac{\partial f_{\text{elas}}}{\partial \eta} = & \frac{1}{2} \left[-C_{ijkl}^{\gamma} \frac{dh}{d\eta} \epsilon_{kl}^{el} \epsilon_{ij}^{el} + 2C_{ijkl}^{\gamma} [1 - h(\eta)] (-\epsilon^{\text{misfit}} \frac{dh}{d\eta}) \epsilon_{ij}^{el} \right. \\ & \left. + C_{ijkl}^{\gamma'} \frac{dh}{d\eta} \epsilon_{kl}^{el} \epsilon_{ij}^{el} + 2C_{ijkl}^{\gamma'} h(\eta) (-\epsilon^{\text{misfit}} \frac{dh}{d\eta}) \epsilon_{ij}^{el} \right] \end{aligned} \quad (\text{B.10})$$

Now we look into the elastic energy related Jacobian. For the diagonal term:

$$\begin{aligned} \text{OnDiag } J_{\text{elas}}^{\eta} &= \frac{\partial}{\partial \eta_n} \left(\sum_{n'} \frac{\partial f_{\text{elas}}}{\partial \eta} \Big|_{\eta=\eta_n, \phi_{n'}, \psi_m} \right) \\ &= \left(\frac{\partial^2 f_{\text{elas}}}{\partial \eta^2} \frac{\partial \sum_{n'} \eta_{n'} \phi_{n'}}{\partial \eta_n}, \psi_m \right) = \left(\frac{\partial^2 f_{\text{elas}}}{\partial \eta^2} \phi_n, \psi_m \right) \end{aligned} \quad (\text{B.11})$$

The derivation of the off-diagonal terms are a little bit complicated:

$$\frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'n}} = \frac{\partial \epsilon_{ij}^{tot}}{\partial u_{i'n}} = \frac{1}{2} \left[\frac{\partial (\frac{\partial u_i}{\partial x_j})}{\partial u_{i'n}} + \frac{\partial (\frac{\partial u_j}{\partial x_i})}{\partial u_{i'n}} \right]. \quad (\text{B.12})$$

Then insert the expansion

$$u_i = \sum_{n'} u_{in'} \phi_{n'}^{(i)}, \quad (\text{B.13})$$

so

$$\frac{\partial u_i}{\partial x_j} = \sum_{n'} u_{in'} \frac{\partial \phi_{n'}^{(i)}}{\partial x_j}. \quad (\text{B.14})$$

Then

$$\frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'n}} = \frac{1}{2} \sum_{n'} \delta_{nn'} \left[\delta_{ii'} \frac{\partial \phi_{n'}^{(i)}}{\partial x_j} + \delta_{ji'} \frac{\partial \phi_{n'}^{(j)}}{\partial x_i} \right] = \frac{1}{2} \left[\delta_{ii'} \frac{\partial \phi_n^{(i)}}{\partial x_j} + \delta_{ji'} \frac{\partial \phi_n^{(j)}}{\partial x_i} \right]. \quad (\text{B.15})$$

$$\frac{du_i}{du_{in}} = \sum_{n'} \frac{du_{in'}}{du_{in}} \phi_{n'}^{(i)} = \phi_n^{(i)} \text{ when } n = n', \frac{du_i}{du_{i'n}} = 0 \text{ when } i \neq i'.$$

$$\frac{\partial C_{ijkl}}{\partial u_{in}} = 0, \quad \frac{\partial^2 C_{ijkl}}{\partial \eta \partial u_{in}} = 0, \quad (\text{B.16})$$

and

$$\frac{\partial^2 \epsilon_{ij}^{el}}{\partial \eta \partial u_{in'}} = 0 \quad (\text{B.17})$$

$$\frac{\partial^2 f_{\text{elas}}}{\partial \eta \partial u_{i'n}} = \frac{1}{2} \left(\frac{\partial C_{ijkl}}{\partial \eta} \frac{\partial \epsilon_{kl}^{el}}{\partial u_{i'n}} \epsilon_{ij}^{el} + \frac{\partial C_{ijkl}}{\partial \eta} \epsilon_{kl}^{el} \frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'n}} + C_{ijkl} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'n}} + C_{ijkl} \frac{\partial \epsilon_{kl}^{el}}{\partial u_{i'n}} \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} \right) \quad (\text{B.18})$$

Note that we can use the symmetries $C_{ijkl} = C_{klij} = C_{jikl} = C_{ijlk}$ in the last two terms in the above equation:

$$\begin{aligned}
& \frac{1}{2}C_{ijkl} \left[\delta_{ki'} \frac{\partial \phi_n^{(k)}}{\partial x_l} + \delta_{li'} \frac{\partial \phi_n^{(l)}}{\partial x_k} \right] \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + \frac{1}{2}C_{ijkl} \left[\delta_{ii'} \frac{\partial \phi_n^{(i)}}{\partial x_j} + \delta_{ji'} \frac{\partial \phi_n^{(j)}}{\partial x_i} \right] \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\
&= \frac{1}{2} \left[C_{ijl'i'} \frac{\partial \phi_n^{(i')}}{\partial x_l} + C_{ijk'i'} \frac{\partial \phi_n^{(i')}}{\partial x_k} \right] \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + \frac{1}{2} \left[C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} + C_{ii'kl} \frac{\partial \phi_n^{(i')}}{\partial x_i} \right] \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\
&= \frac{1}{2} \left[C_{ijl'i'} \frac{\partial \phi_n^{(i')}}{\partial x_l} + C_{ijk'i'} \frac{\partial \phi_n^{(i')}}{\partial x_k} \right] \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + \frac{1}{2} \left[C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} + C_{i'ikl} \frac{\partial \phi_n^{(i')}}{\partial x_i} \right] \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\
&= C_{ijk'i'} \frac{\partial \phi_n^{(i')}}{\partial x_k} \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\
&= C_{i'kij} \frac{\partial \phi_n^{(i')}}{\partial x_k} \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\
&= 2C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta}. \tag{B.19}
\end{aligned}$$

Note that there is no sum over the index i' ! And the summation of the first two terms is:

$$\frac{\partial C_{ijkl}}{\partial \eta} \frac{\partial \epsilon_{kl}^{el}}{\partial u_{i'n}} \epsilon_{ij}^{el} + \frac{\partial C_{ijkl}}{\partial \eta} \epsilon_{kl}^{el} \frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'n}} \tag{B.20}$$

$$= \frac{1}{2} \frac{\partial C_{ijkl}}{\partial \eta} \left[\delta_{ki'} \frac{\partial \phi_n^{(k)}}{\partial x_l} + \delta_{li'} \frac{\partial \phi_n^{(l)}}{\partial x_k} \right] \epsilon_{ij}^{el} + \frac{1}{2} \frac{\partial C_{ijkl}}{\partial \eta} \left[\delta_{ii'} \frac{\partial \phi_n^{(i)}}{\partial x_j} + \delta_{ji'} \frac{\partial \phi_n^{(j)}}{\partial x_i} \right] \epsilon_{kl}^{el} \tag{B.21}$$

$$= \frac{\partial C_{ijk'i'}}{\partial \eta} \frac{\partial \phi_n^{(i')}}{\partial x_k} \epsilon_{ij}^{el} + \frac{\partial C_{i'jkl}}{\partial \eta} \frac{\partial \phi_n^{(i')}}{\partial x_j} \epsilon_{kl}^{el} \tag{B.22}$$

$$= 2 \frac{\partial C_{i'jkl}}{\partial \eta} \frac{\partial \phi_n^{(i')}}{\partial x_j} \epsilon_{kl}^{el} \tag{B.23}$$

$$\frac{\partial^2 f_{\text{elas}}}{\partial \eta \partial u_{i'n}} = C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} + \frac{\partial C_{i'jkl}}{\partial \eta} \frac{\partial \phi_n^{(i')}}{\partial x_j} \epsilon_{kl}^{el} \tag{B.24}$$

$$\text{OffDiag} J_{\eta}^{u_{i'}} = \frac{\partial R_{\eta}}{\partial u_{i'n}} = \left(\frac{\partial^2 f_{\text{elas}}}{\partial \eta \partial u_{i'n}}, \psi_m \right) \tag{B.25}$$

Or we could derive the off-diagonal terms in a different way using chain rule:

$$\frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'}} = \frac{\partial \epsilon_{ij}^{tot}}{\partial u_{i'}} = \frac{1}{2} \left[\frac{\partial(\frac{\partial u_i}{\partial x_j})}{\partial u_{i'}} + \frac{\partial(\frac{\partial u_j}{\partial x_i})}{\partial u_{i'}} \right] = \frac{1}{2} \left[\delta_{ii'} \frac{\partial}{\partial x_j} + \delta_{ji'} \frac{\partial}{\partial x_i} \right]. \tag{B.26}$$

$$\frac{\partial^2 f_{\text{elas}}}{\partial \eta \partial u_{i'}} = \frac{1}{2} \left(\frac{\partial C_{ijkl}}{\partial \eta} \frac{\partial \epsilon_{kl}^{el}}{\partial u_{i'}} \epsilon_{ij}^{el} + \frac{\partial C_{ijkl}}{\partial \eta} \epsilon_{kl}^{el} \frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'}} + C_{ijkl} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \frac{\partial \epsilon_{ij}^{el}}{\partial u_{i'}} + C_{ijkl} \frac{\partial \epsilon_{kl}^{el}}{\partial u_{i'}} \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} \right) \tag{B.27}$$

$$\frac{\partial u_{i'}}{\partial u_{i'n}} = \sum_{n'} \frac{\partial u_{i'n'}}{\partial u_{i'n}} \phi_{n'}^{(i')} = \phi_n^{(i')} \quad (\text{B.28})$$

The last two terms in Eq. (B.27) times $\frac{\partial u_{i'}}{\partial u_{i'n}}$:

$$\begin{aligned} & \frac{1}{2} C_{ijkl} \left[\delta_{ki'} \frac{\partial \phi_n^{(i')}}{\partial x_l} + \delta_{li'} \frac{\partial \phi_n^{(i')}}{\partial x_k} \right] \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + \frac{1}{2} C_{ijkl} \left[\delta_{ii'} \frac{\partial \phi_n^{(i')}}{\partial x_j} + \delta_{jj'} \frac{\partial \phi_n^{(i')}}{\partial x_i} \right] \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\ &= \frac{1}{2} \left[C_{ijil'} \frac{\partial \phi_n^{(i')}}{\partial x_l} + C_{ijk i'} \frac{\partial \phi_n^{(i')}}{\partial x_k} \right] \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + \frac{1}{2} \left[C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} + C_{ii'kl} \frac{\partial \phi_n^{(i')}}{\partial x_i} \right] \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\ &= \frac{1}{2} \left[C_{ijli'} \frac{\partial \phi_n^{(i')}}{\partial x_l} + C_{ijk i'} \frac{\partial \phi_n^{(i')}}{\partial x_k} \right] \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + \frac{1}{2} \left[C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} + C_{i'ikl} \frac{\partial \phi_n^{(i')}}{\partial x_i} \right] \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\ &= C_{ijk i'} \frac{\partial \phi_n^{(i')}}{\partial x_k} \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\ &= C_{i'kij} \frac{\partial \phi_n^{(i')}}{\partial x_k} \frac{\partial \epsilon_{ij}^{el}}{\partial \eta} + C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} \\ &= 2C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta}. \end{aligned} \quad (\text{B.29})$$

$$\begin{aligned} \frac{\partial^2 f_{\text{elas}}}{\partial \eta \partial u_{i'n}} &= \frac{\partial^2 f_{\text{elas}}}{\partial \eta \partial u_{i'}} \frac{\partial u_{i'}}{\partial u_{i'n}} = C_{i'jkl} \frac{\partial \phi_n^{(i')}}{\partial x_j} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} + \frac{\partial C_{i'jkl}}{\partial \eta} \frac{\partial \phi_n^{(i')}}{\partial x_j} \epsilon_{kl}^{el} \\ &= \left(C_{i'jkl} \frac{\partial \epsilon_{kl}^{el}}{\partial \eta} + \frac{\partial C_{i'jkl}}{\partial \eta} \epsilon_{kl}^{el} \right) \frac{\partial \phi_n^{(i')}}{\partial x_j} \\ &= (C_{i'jkl}^\gamma [1 - h(\eta)] (-\epsilon^{\text{misfit}} \frac{dh}{d\eta}) - C_{i'jkl}^\gamma \frac{dh}{d\eta} \epsilon_{kl}^{el} \\ &\quad + C_{i'jkl}^{\gamma'} h(\eta) (-\epsilon^{\text{misfit}} \frac{dh}{d\eta}) + C_{i'jkl}^{\gamma'} \frac{dh}{d\eta} \epsilon_{kl}^{el}) \frac{\partial \phi_n^{(i')}}{\partial x_j} \end{aligned} \quad (\text{B.30})$$

The residual in Eq. (A.11) can be calculated in the following way:

$$R_{u_i} = - \int \sum_j \frac{\partial}{\partial x_j} \sigma_{ij} \cdot \psi_m dV = \int \sum_j \sigma_{ij} \cdot \frac{\partial \psi_m}{\partial x_j} dV = \left(\sum_j \sigma_{ij}, \frac{\partial \psi_m}{\partial x_j} \right) \quad (\text{B.31})$$

For terms in Eq. (A.12):

$$\begin{aligned} \text{OnDiag } J_{u_i}^{u_i} &= \frac{\partial R_{u_i}}{\partial u_{in}} = \frac{\partial}{\partial u_{in}} \left(\sum_j \sigma_{ij}, \frac{\partial \psi_m}{\partial x_j} \right) \\ &= \left(\frac{\partial \sum_j \sigma_{ij}}{\partial u_i}, \frac{\partial \sum_{n'} u_{in'} \phi_{n'}^{(i)}}{\partial u_{in}}, \frac{\partial \psi_m}{\partial x_j} \right) = \left(\sum_j \frac{\partial \sigma_{ij}}{\partial u_i} \phi_n^{(i)}, \frac{\partial \psi_m}{\partial x_j} \right) \end{aligned} \quad (\text{B.32})$$

$$\begin{aligned}
\sum_j \frac{\partial \sigma_{ij}}{\partial u_i} \phi_n^{(i)} &= \frac{1}{2} \sum_j C_{ijkl} \left[\delta_{ki} \frac{\partial \phi_n^{(i)}}{\partial x_l} + \delta_{li} \frac{\partial \phi_n^{(i)}}{\partial x_k} \right] \\
&= \frac{1}{2} \sum_j \left(C_{ijil} \frac{\partial \phi_n^{(i)}}{\partial x_l} + C_{ijkj} \frac{\partial \phi_n^{(i)}}{\partial x_k} \right) = \sum_j C_{ijkj} \frac{\partial \phi_n^{(i)}}{\partial x_k}
\end{aligned} \tag{B.33}$$

$$\begin{aligned}
\text{OffDiag } J_{u_i}^\eta &= \frac{\partial R_{u_i}}{\partial \eta_n} = \frac{\partial}{\partial \eta_n} \left(\sum_j \sigma_{ij}, \frac{\partial \psi_m}{\partial x_j} \right) \\
&= \left(\frac{\partial \sum_j \sigma_{ij}}{\partial \eta} \frac{\partial \sum_{n'} \eta_{n'} \phi_{n'}}{\partial \eta_n}, \frac{\partial \psi_m}{\partial x_j} \right) = \left(\sum_j \frac{\partial \sigma_{ij}}{\partial \eta} \phi_n, \frac{\partial \psi_m}{\partial x_j} \right)
\end{aligned} \tag{B.34}$$

$$\sum_j \frac{\partial \sigma_{ij}}{\partial \eta} \phi_n = \sum_j \frac{\partial C_{ijkl}}{\partial \eta} \epsilon_{kl} \phi_n + \sum_j C_{ijkl} \frac{\partial \epsilon_{kl}}{\partial \eta} \phi_n \tag{B.35}$$

$$\begin{aligned}
\text{OffDiag } J_{u_i}^{u_{i''}} &= \frac{\partial R_{u_i}}{\partial u_{i''n}} = \frac{\partial}{\partial u_{i''n}} \left(\sum_j \sigma_{ij}, \frac{\partial \psi_m}{\partial x_j} \right) (i'' \neq i) \\
&= \left(\frac{\partial \sum_j \sigma_{ij}}{\partial u_{i''}} \frac{\partial \sum_{n'} u_{i''n'} \phi_{n'}^{(i'')}}{\partial u_{i''n}}, \frac{\partial \psi_m}{\partial x_j} \right) = \left(\sum_j \frac{\partial \sigma_{ij}}{\partial u_{i''}} \phi_n^{(i'')}, \frac{\partial \psi_m}{\partial x_j} \right)
\end{aligned} \tag{B.36}$$

$$\begin{aligned}
\sum_j \frac{\partial \sigma_{ij}}{\partial u_{i''}} \phi_n^{(i'')} &= \frac{1}{2} \sum_j C_{ijkl} \left[\delta_{ki''} \frac{\partial \phi_n^{(i'')}}{\partial x_l} + \delta_{li''} \frac{\partial \phi_n^{(i'')}}{\partial x_k} \right] \\
&= \frac{1}{2} \sum_j \left(C_{ijji''} \frac{\partial \phi_n^{(i'')}}{\partial x_l} + C_{ijkj''} \frac{\partial \phi_n^{(i'')}}{\partial x_k} \right) \\
&= \sum_j C_{ijkj''} \frac{\partial \phi_n^{(i'')}}{\partial x_k} (i'' \neq i)
\end{aligned} \tag{B.37}$$

Appendix C. Coefficients comparison using different definition of precipitate size

When fitting the input parameters and their crossterms to the equilibrium precipitate size, one can also quantify the size of the precipitate by using the spherical equivalent radius (proportional to the cube root of the precipitate volume). The fitting results will not differ much from using the average distance from the interface to the center of the precipitate as shown in Table C.3.

References

- [1] K. Ishida, Recent progress on Co-base alloys - phase diagrams and application, Archives of Metallurgy and Materials 53 (2008) 1075–1088.

Table C.3: The fitted coefficient values for the precipitate size function using the average distance from the interface to the center of the precipitate, a_i^{avg} , and the fitted coefficient values for the precipitate size function using the spherical equivalent radius, a_i^{eqv} .

a_i^{avg}	coef. value	a_i^{eqv}	coef. value	a_i^{avg}	coef. value	a_i^{eqv}	coef. value
a_1^{avg}	-0.312	a_1^{eqv}	-0.313	a_6^{avg}	-0.0797	a_6^{eqv}	-0.0829
a_2^{avg}	-0.0529	a_2^{eqv}	-0.0516	a_7^{avg}	-0.268	a_7^{eqv}	-0.270
a_3^{avg}	0.0200	a_3^{eqv}	0.0079	a_8^{avg}	0.0921	a_8^{eqv}	0.0904
a_4^{avg}	-0.0494	a_4^{eqv}	-0.0635	a_9^{avg}	-0.122	a_9^{eqv}	-0.127
a_5^{avg}	-0.235	a_5^{eqv}	0.234	a_{10}^{avg}	0.0636	a_{10}^{eqv}	0.0548

- [2] T. M. Pollock, J. Dibbern, M. Tsunekane, J. Zhu, A. Suzuki, New Co-based γ - γ' high-temperature alloys, JOM 62 (2010) 59–64.
- [3] J. Sato, T. Omori, K. Oikawa, I. Ohnuma, R. Kainuma, K. Ishida, Cobalt-base high-temperature alloys, Science 312 (2006) 90–91.
- [4] R. C. Reed, The Superalloys: Fundamentals and Applications, Cambridge University Press, 2006.
- [5] Center for Hierarchical Materials Design: CHiMaD, <https://chimad.northwestern.edu/>, 2019.
- [6] P. Wang, W. Xiong, U. R. Kattner, C. E. Campbell, E. A. Lass, O. Y. Kontsevoi, G. B. Olson, Thermodynamic re-assessment of the Al-Co-W system, CALPHAD 59 (2017) 112–130.
- [7] P. J. Bocchini, E. A. Lass, K. W. Moon, M. E. Williams, C. E. Campbell, U. R. Kattner, D. C. Dunand, D. N. Seidman, Atom-probe tomographic study of γ/γ' interfaces and compositions in an aged Co–Al–W superalloy, Scripta Materialia 68 (2013) 563–566.
- [8] W. C. Johnson, J. W. Cahn, Elastically induced shape bifurcations of inclusions, Acta Metallurgica 32 (1984) 1925–1933.
- [9] M. E. Thompson, C. S. Su, P. W. Voorhees, The equilibrium shape of a misfitting precipitate, Acta Metallurgica et Materialia 42 (1994) 2107–2122.
- [10] H.-J. Jou, P. H. Leo, J. S. Lowengrub, Microstructural evolution in inhomogeneous elastic media, Journal of Computational Physics 131 (1997) 109–148.
- [11] D. Mueller, E. Gross, 3d simulation of equilibrium morphologies of precipitates, Computational Materials Science 11 (1998) 35–44.
- [12] M. E. Thompson, P. W. Voorhees, Equilibrium particle morphologies in elastically stressed coherent solids, Acta Materialia 47 (1999) 983–996.
- [13] S. Kolling, R. Mueller, D. Gross, The influence of elastic constants on the shape of an inclusion, International Journal of Solids and Structures 40 (2003) 4399–4416.

- [14] X. Li, K. Thornton, Q. Nie, P. W. Voorhees, J. S. Lowengrub, Two-and three-dimensional equilibrium morphology of a misfitting particle and the Gibbs-Thomson effect, *Acta Materialia* 52 (2004) 5829–5843.
- [15] X. Zhao, R. Duddu, S. P. Bordas, J. Qu, Effects of elastic strain energy and interfacial stress on the equilibrium morphology of misfit particles in heterogeneous solids, *Journal of the Mechanics and Physics of Solids* 61 (2013) 1433–1445.
- [16] R. Shi, N. Ma, Y. Wang, Predicting equilibrium shape of precipitates as function of coherency state, *Acta Materialia* 60 (2012) 4172–4184.
- [17] A. M. Jokisaari, S. S. Naghavi, C. Wolverton, P. W. Voorhees, O. G. Heinonen, Predicting the morphologies of γ' precipitates in cobalt-based superalloys, *Acta Materialia* 141 (2017) 273–284.
- [18] D. Wang, Y. Li, S. Shi, X. Tong, Z. Yan, Phase-field simulation of γ' precipitates rafting and creep property of Co-base superalloys, *Materials and Design* 196 (2020) 109077.
- [19] S. G. Kim, W. T. Kim, T. Suzuki, Phase-field model for binary alloys, *Physical Review E* 60 (1999) 7186–7197.
- [20] U. R. Kattner, The CALPHAD method and its role in material and process development, *Technol. Metal. Mater. Min.* 13 (2016) 3–15.
- [21] A. T. Dinsdale, SGTE data for pure elements, *CALPHAD* 15 (1991) 317–425.
- [22] O. Redlich, A. T. Kister, Algebraic representation of thermodynamic properties and the classification of solutions, *Industrial and Engineering Chemistry* 40 (1948) 345–348.
- [23] N. Dupin, I. Ansara, B. Sundman, Thermodynamic re-assessment of the ternary system Al-Cr-Ni, *CALPHAD* 25 (2001) 279–298.
- [24] M. Hillert, The compound energy formalism, *Journal of Alloys and Compounds* 320 (2001) 161–176.
- [25] J. D. Eshelby, The determination of the elastic field of an ellipsoidal inclusion, and related problems, *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences* 241 (1957) 376–396.
- [26] C. E. Campbell, Assessment of the diffusion mobilities in the γ' and B2 phases in the Ni–Al–Cr system, *Acta Materialia* 56 (2008) 4277–4290.
- [27] J. Ågren, Diffusion in phases with several components and sublattices, *Journal of Physics and Chemistry of Solids* 43 (1982) 421–430.
- [28] K. W. Moon, C. E. Campbell, M. E. Williams, W. J. Boettinger, Diffusion in FCC Co-rich Co–Al–W alloys at 900 and 1000 °C, *Journal of Phase Equilibria and Diffusion* 37 (2016) 402–415.

- [29] J. Andersson, J. Ågren, Models for numerical treatment of multicomponent diffusion in simple phases, *Journal of Applied Physics* 72 (1992) 1350–1355.
- [30] F. Pyczak, A. Bauer, M. Göken, U. Lorenz, S. Neumeier, M. Oehring, J. Paul, N. Schell, A. Schreyer, A. Stark, F. Symanzik, The effect of tungsten content on the properties of L12-hardened Co–Al–W alloys, *Journal of Alloys and Compounds* 632 (2015) 110–115.
- [31] F. Pyczak, A. Bauer, M. Göken, S. Neumeier, U. Lorenz, M. Oehring, N. Schell, A. Schreyer, A. Stark, F. Symanzik, Plastic deformation mechanisms in a crept L12 hardened Co-base superalloy, *Materials Science and Engineering: A* 571 (2013) 13–18.
- [32] K. Tanaka, M. Ooshima, N. Tsuno, A. Sato, H. Inui, Creep deformation of single crystals of new Co-Al-W-based alloys with fcc/L12 two-phase microstructures, *Philosophical Magazine* 92 (2012) 4011–4027.
- [33] D. Gaston, J. Peterson, C. Permann, D. Andrs, A. Slaughter, J. Miller, Continuous integration for concurrent computational framework and application development, *Journal of Open Research Software* 2 (2014).
- [34] D. R. Gaston, C. J. Permann, J. W. Peterson, A. E. Slaughter, D. Andrš, Y. Wang, M. P. Short, D. M. Perez, M. R. Tonks, J. Ortensi, et al., Physics-based multiscale coupling for full core nuclear reactor simulation, *Annals of Nuclear Energy* 84 (2015) 45–54.
- [35] C. J. Permann, D. R. Gaston, D. Andrš, R. B. Carlsen, F. Kong, A. D. Lindsay, J. M. Miller, J. W. Peterson, A. E. Slaughter, R. H. Stogner, R. C. Martineau, MOOSE: Enabling massively parallel multiphysics simulation, *SoftwareX* 11 (2020) 100430.
- [36] L. K. Aagesen, D. Schwen, MARMOT Phase-Field Model for the U-Si System, Technical Report, U.S. Department of Energy Office of Scientific and Technical Information, 2016.
- [37] L. K. Aagesen, D. Schwen, K. Ahmed, M. R. Tonks, Quantifying elastic energy effects on interfacial energy in the kim-kim-suzuki phase-field model with different interpolation schemes, *Computational Materials Science* 140 (2017) 10–21.
- [38] H. Garcke, B. Nestler, B. Stinner, F. Wendler, Allen-Cahn systems with volume constraints, *Mathematical Models and Methods in Applied Sciences* 18 (2008) 1347–1381.
- [39] B. S. Kirk, J. W. Peterson, R. H. Stogner, G. F. Carey, et al., libMesh : a C++ library for parallel adaptive mesh refinement/coarsening simulations, *Engineering with Computers* 22 (2006) 237–254.
- [40] C. M. Elliott, D. A. French, F. A. Milner, A second order splitting method for the Cahn-Hilliard equation, *Numerische Mathematik* 54 (1989) 575–590.
- [41] M. R. Tonks, D. Gaston, P. C. Millett, D. Andrs, P. Talbot, An object-oriented finite element framework for multiphysics phase field simulations, *Computational Materials Science* 51 (2012) 20–29.

- [42] Derivation of the KKS system residuals and Jacobians, https://mooseframework.inl.gov/modules/phase_field/MultiPhase/KKSDerivations.html, 2019.
- [43] V. A. Vorontsov, J. S. Barnard, K. M. Rahman, H. Y. Yan, P. A. Midgley, D. Dye, Coarsening behaviour and interfacial structure of γ' precipitates in Co-Al-W based superalloys, *Acta Materialia* 120 (2016) 14–23.