PHASE-FIELD MODELING OF γ' PRECIPITATION IN MULTI-COMPONENT NI-BASE SUPERALLOYS

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Abstract

To simulate γ' precipitation in multi-component Ni-base superalloys using the phase-field method, the Gibbs free energy of the γ and γ' phases was linked to the CALPHAD method. The free energy curve and phase equilibrium compositions in this work showed good agreement with results obtained using Thermo-Calc calculation, indicating that the model was successfully coupled with the CALPHAD method. Using this coupling method, the γ' precipitation in a quaternary Ni-Al-Re-Ta and a third-generation Ni-base superalloy TMS-75 systems were simulated. These one-dimensional simulations demonstrated the diffusion-controlled γ' precipitation under local equilibrium with precipitation interactions. According to our atom probe microanalyses, their simulated composition evolutions were reasonable. Such a microstructure modeling technique can contribute to alloy design, process design and even lifetime assessment of high-temperature turbine blades currently in service.

Introduction

Modeling the microstructure evolution of Ni-base superalloys is of considerable importance for alloy design, process design and lifetime assessment of turbine components, e.g. in a virtual turbine [1]. Phase-field simulation has been a powerful tool to predict microstructure evolution in Ni-Al systems [2-8]. Nevertheless, few reports have described its γ' precipitation in multi-component Ni-base superalloys, even though recent practical Ni-base superalloys incorporate over 10 elements, such as Al, Cr, Mo, Re, and Ta [9-11]. The addition of refractory elements, especially Re, confers solid solution strengthening at high temperature. However, the small diffusivity of such refractory elements renders it difficult to predict the elemental distributions in γ and γ' phases and the size distribution of γ' precipitates at the thermal equilibrium state. Therefore, to enable prediction of microstructure morphology in multi-component superalloys, development of a kinetic-modeling technique for multi-component systems is of considerable importance. Diffusion Controlled Transformation (DICTRA) software has been used to predict the growth rate of a simple-shaped precipitate for the multi-component systems in one-dimensional calculation [12]. However, accompanying the interaction of precipitates across the γ phase is difficult because the interface energies, such as inter-phase domain boundary (IPB) and anti-phase boundary (APB) energies, are not taken into account in the DICTRA. On the other

hand, the phase-field model is more attractive to predict complicated morphology evolution because of the characteristics of the so-called *diffuse interface model*.

One reason for the multi-component difficulty in phase-field modeling is the complexity of deriving the chemical-free-energy surface in the multi-component system, especially for order/disorder phase transformation. In phase-field modeling, the chemical free energy is an important factor that determines the driving force and path of the microstructure evolution. As for a binary Ni-Al superalloy system, some descriptions of the chemical free energy function have been reported, e.g., using the Landau-type polynomial form, the KKS model, and a link to the CALPHAD method [2-8]. However, few descriptions of the multi-component Ni-base superalloy have been proposed [13,14].

In this study, a new phase-field modeling technique that can address γ' precipitation in multi-component superalloys was developed. This was achieved using the single function formalism of γ/γ' Gibbs free energy directly in the CALPHAD method for the chemical free energy in the phase-field method. Using this coupling method, the γ' precipitation for a quaternary Ni-Al-Ta-Re superalloy and a third-generation Ni-base superalloy TMS-75 (Ni-6Al-12Co-3Cr-2Mo-5Re-6Ta-6W-0.1Hf, wt%) were simulated in one dimension. The temporal evolution of element compositions during the growth and coalescence of γ' precipitates was discussed using results of atom probe analyses.

Modeling

A thermodynamic description of the γ (fcc-Al) and γ' ($L1_2$) phases in the binary Ni-Al system was obtained by Ansara et al. using a four-sublattice model [15]. The Gibbs energy of the γ' phase was described by separating it into disordered and ordered contributions in a single function to allow independent evaluation of both γ and γ' phase parameters. Zhu et al. [5] proposed a model directly linking a phase-field model with existing thermodynamic databases using the CALPHAD method for the binary Ni-Al system.

This method was extended to multi-component systems using the four-sublattice model. The site fraction of each alloying element i ($i = \text{Al, Co, Cr, Mo, Ni, Re, Ta and W}$) on each sublattice s is denoted as $y_i^{(s)}$. The molar Gibbs energy for the γ and γ' phases is expressed using the following equations [15,16]:

$$G^{fcc,or,L1_2} = \sum_i c_i G_i^{fcc} + RT \sum_i c_i \ln c_i + \sum_i \sum_{j>i} c_i c_j \sum_{n=0}^m L_{i,j}^{fcc} (c_i - c_j)^n + \sum_i \sum_{j>i} \sum_{k>j} c_i c_j c_k L_{i,j,k}^{fcc} + \Delta G^{ord}(y_i^{(s)}), \quad (1)$$

$$\Delta G^{ord}(y_i^{(s)}) = \Delta G^{L1_2}(y_i^{(s)}) - \Delta G^{L1_2}(y_i^{(s)} = c_i), \quad (2)$$

$$\Delta G^{L1_2}(y_i^{(s)}) = \sum_i \sum_j \sum_k \sum_l y_i^{(1)} y_j^{(2)} y_k^{(3)} y_l^{(4)} \Delta G_{ijkl}^{L1_2} + \frac{RT}{4} \sum_s \sum_i y_i^{(s)} \ln(y_i^{(s)}) + \sum_s \sum_i \sum_{j>i} y_i^{(s)} y_j^{(s)} \sum_{n=0}^l L_{i,j}^{L1_2} (y_i^{(s)} - y_j^{(s)})^n, \quad (3)$$

where i, j, k and l can be Al, Co, Cr, Mo, Re, Ta, W and Ni on any of the sublattices. c_i is a mole-fraction of element i , R is the gas constant and T is the temperature. G_i^{fcc} is a molar Gibbs energy of element i with the structure of fcc. The first four terms of Eq. (1) correspond to the Gibbs energy of the disordered γ phase; $\Delta G^{ord}(y_i^{(s)})$ is the ordering energy of the γ' phase as a function of the site fractions. $\Delta G_{ijkl}^{L1_2}$ was expressed in terms of the interchange bond energies (e.g., U_{ij} between i and j atoms), as

$$\Delta G_{ijkl}^{L1_2} = U_{ij} + U_{ik} + U_{il} + U_{jk} + U_{jl} + U_{kl} + a_{ijkl}, \quad (4)$$

where a_{ijkl} is a correction variable for some compounds [16]. All variables in Eqs. (1), (3), and (4) are obtainable from the CALPHAD-type assessment [15-18]. Blavette et al. demonstrated by an atom probe study that the partitioning behavior between the Ni-site and the Al-site in the γ' phase depends greatly on the alloying elements [19]. Therefore, the order parameter of each element was newly introduced for the multi-component system. According to the Landau-Lifshitz rule [5, 20, 21], the order parameter of four interpenetrating simple cubic sublattices had three components that can be represented by the site fraction in each sublattice. The order parameters and the composition for each element are defined as

$$\eta_i^1 = (y_i^{(1)} - y_i^{(2)} - y_i^{(3)} + y_i^{(4)})/4c_i, \quad (5a)$$

$$\eta_i^2 = (y_i^{(1)} - y_i^{(2)} + y_i^{(3)} - y_i^{(4)})/4c_i, \quad (5b)$$

$$\eta_i^3 = (y_i^{(1)} + y_i^{(2)} - y_i^{(3)} - y_i^{(4)})/4c_i, \quad (5c)$$

$$c_i = (y_i^{(1)} + y_i^{(2)} + y_i^{(3)} + y_i^{(4)})/4, \quad (5d)$$

where η_i^j ($i=1, 2, 3, j=Al, Co, Cr, Mo, Re, Ta, W$), c_j ($j=Al, Co, Cr, Mo, Re, Ta, W$) respectively indicate the order parameter and the composition fields. By combining Eqs. (1)-(5), the molar Gibbs energy of the γ and γ' phases for the multi-component system can be described using the variables of the order parameter

and composition fields for elements Al, Co, Cr, Mo, Re, Ta, W and Ni.

The total stress-free chemical free energy F_{sys}^{multi} in the multi-component system includes the local free energy density and the interface energy, given as [22]

$$F_{sys}^{multi} = \int_V \left(\frac{1}{V_m} G^{fcc,or,L1_2} + \frac{\alpha}{2} \sum_j (\nabla c_j)^2 + \frac{\beta}{2} \sum_j \sum_{i=1}^3 (\nabla \eta_i^j)^2 \right) dV, \quad (6)$$

where V_m is the molar volume, which was assumed to be constant. In addition, α and β respectively signify the gradient energy coefficients for the compositions and order parameters. For this study, to retain simplicity, the composition-dependent elastic energy stemming from the elastic inhomogeneity is not included.

The temporal evolution of the element compositions and order parameters are obtainable by solving the following equations:

$$\frac{\partial c_i}{\partial t} = \sum_j \nabla \cdot \left(\tilde{M}_{ij} \nabla \frac{\delta F}{\delta c_j} \right), \quad (7)$$

$$i = j = Al, Co, Cr, Mo, Re, Ta, W,$$

$$\frac{\partial \eta_i^j}{\partial t} = -L \frac{\delta F}{\delta \eta_i^j}, \quad (8)$$

$$i = Al, Co, Cr, Mo, Re, Ta, W, j = 1, 2, 3$$

where $\tilde{M}_{ij}$ is the so-called chemical mobility with Ni as the dependent species, and L is the structural relaxation coefficient. The chemical mobility $\tilde{M}_{ij}$ for the γ phase is related to the atomic mobilities of the constituent elements, as

$$\tilde{M}_{ij} = \sum_l (\delta_{il} - c_i)(\delta_{jl} - c_j) c_l M_l^{\gamma}, \quad (9)$$

where δ_{il} and δ_{jl} are the Kronecker's delta and M_l^{γ} is obtainable from the mobility database [23, 24]. The chemical mobility for the γ' phase was assumed to be identical to that of the γ phase. As for parameter L , it was determined as $\tilde{M}_{AlAl}=L a_0/16$ [25], such that the interface migration velocity was mainly controlled by the diffusion mobility, because the structural relaxation was much faster than the diffusion, with lattice parameter $a_0=3.35 \times 10^{-10} m$. Assuming the gradient energy coefficient α to be zero, $\beta = 3.75 \times 10^{-16} J \cdot m^2 / mol$ for Ni-Al-Re-Ta system, and $\beta = 1.13 \times 10^{-15} J \cdot m^2 / mol$ for TMS-75 were chosen by fitting the interfacial energy to $10 mJ/m^2$, which was close to the experimental and simulated values reported in a Ni-Al system [7,26,27], although very few accurate experimental interface energy data for binary and multi-component Ni-base superalloy systems are available. The assumption that the composition gradient coefficient is zero was a simplification introduced by Vaithyanathan et al. [4]. A Semi-Implicit-Fourier-Spectral-

Method [28] was used for numerical analyses with a periodical boundary condition. The grid number and grid size were set respectively to 256 and 0.5 nm. At the nucleation stage, small cells of 3 nm diameter in the field were transformed to γ' phases with the values of equilibrium order parameters after uniform random values were generated and compared to a certain value.

Experimental Procedures

For this study, TMS-75 alloy, a third-generation Ni-base single crystal superalloy developed by the National Institute for Materials Science (NIMS) Japan, was investigated using atom-probe analysis in this study. Its nominal composition is Ni-6Al-12Co-3Cr-2Mo-5Re-6Ta-6W-0.1Hf in weight %, whereas Ni-13.7Al-12.6Co-3.6Cr-1.3Mo-1.7Re-2.1Ta-2.0W-0.01Hf in atomic % [10,11]. Bar-shaped samples (10mm diameter, 120mm length) were cast into single crystal form by directional solidification with the longitudinal axis within 5° from the [100] pole in the standard stereographic triangle. One bar sample was solution treated for 1 h at 1300°C , then by 4 h at 1320°C in the argon gas; then air-cooled. The solution-treated bar sample was then machined into two plate-type samples, which were $20\text{mm} \times 9\text{mm} \times 2\text{mm}$. Subsequently, they were aged at 1100°C for 15 min or 4 h in air. They were then water quenched.

To avoid preparing atom-probe tips with the oxidized microstructure, the cross-section microstructure of the heat-treated plate-type sample was observed using scanning electron microscopy (SEM). The γ' depletion and microstructure inhomogeneity in a sample that was heat-treated at 1100°C for 4 h were observed in the range of maximum $30\text{ }\mu\text{m}$ from the surface. Square rods of approximately $0.2\text{mm} \times 0.2\text{mm} \times 10\text{mm}$ were cut from the bulk of the heat-treated sample without the oxidized microstructure. These rods were electropolished with 25% perchloric acid and 75% acetic acid to sharp needle-shaped specimens initially, with subsequent treatment in 2% perchloric acid in butoxyethanol. Atom-probe analyses were performed at a tip temperature of about 50K under ultrahigh vacuum ($<1 \times 10^{-8}\text{Pa}$) with a pulse fraction of 0.2 and a pulse repetition rate of 1500 Hz.

Results and Discussion

Phase-Field Simulation of the Ni-Al-Re-Ta System

Figure 1 presents a comparison of the Gibbs energy using the equilibrium order parameters obtained under the condition of $\partial G / \partial \eta_i^j = 0$ ($i=\text{Al, Ta, Re, } j=1, 2, 3$), with the result obtained using the Thermo-Calc calculation of the total Gibbs free energy. According to the result, the free energy of the γ' phase was lower than that of the γ phase from 0.148 to 0.18 of C_{Al} . The free energy curve linking to the CALPHAD method agreed well

with that of the Thermo-Calc calculation, which indicates that the free energy surface in the multi-component system was successfully derived by linkage to the CALPHAD method. Therefore, the driving force and path of the microstructure evolution are obtainable in the phase-field method according to the CALPHAD method, even in a complex higher-component system.

Using the free energy linking to the CALPHAD method, a one-dimensional simulation for the γ' precipitation of a Ni-16.5at% Al-1at%Re-2at%Ta superalloy at 1100°C was carried out as shown in Fig. 2. Ta and Re were rich in the γ' and γ phases, respectively. After nuclei formed, the precipitates were supersaturated with Al of 0.19, as presented in Fig. 2(b). Fig. 2(c) shows that the supersaturation then decreased continuously. The local equilibrium of Re at the γ/γ' interface, as demonstrated by results of atom probe studies [29, 30], was confirmed in Fig. 2(c). In Figs. 2(b) and 2(c), precipitate B is closer to C than to A. However, precipitates A and B subsequently merged and decreased the interface energy, as presented in Fig. 2(d) because the domain of precipitate A was identical to that of B, whereas those of B and C differed, not forming an APB. The APB energy was usually high: twice as large as the IPB energy.

Regarding the coalescence of precipitates A and B, after two spikes of Re, denoted by D and E, came near each other, a narrow γ channel formed with a supersaturated Re composition, which piled up as a result of D and E, thereby prompting a higher Re spike, presented as F in Fig. 2(d). Moreover, the small diffusivity of Re made the spike F decrease slowly in the γ' phase. The phase compositions then gradually reached the equilibrium composition. The spike of the element composition accompanied by the precipitate coalescence, such as that shown by F in Fig. 2(d), was probably common for alloying elements, although the diffusion rate of the spike depended largely on the element diffusivity.

Figure 1 Comparison of free energy obtained in this work with the calculated result obtained using Thermo-Calc software.

Figure 2 Temporal evolution of Al, Re, and Ta composition profiles in a Ni-16.5at% Al-1at%Re-2at%Ta superalloy at 1100°C.

		Co	Cr	Mo	W	Al	Ta	Re	V _f
Calculation (this work).	γ'	6.3	1.3	0.4	0.7	18.9	3.5	0.3	0.51
	γ	19.1	6.0	2.2	3.4	8.4	0.4	3.1	
Observation by Yokokawa et al.[11].	γ'	10.0	1.8	0.7	1.6	16.9	2.7	0.4	0.55 [10]
	γ	18.2	6.4	1.9	2.2	8.9	1.0	3.1	

Table 1 Comparison of the predicted and the observed γ and γ' phase compositions of TMS-75 at 1100°C.

In Fig. 2(f), the phase compositions at $x=10\text{nm}$ in the γ' phase and at $x=115\text{nm}$ in the γ phase were Ni-18.4at%Al-0.472at%Re-2.87at%Ta and Ni-13.8at%Al-1.70at%Re-0.792at%Ta, respectively. These values agreed well with the equilibrium compositions calculated using Thermo-Calc software, which were Ni-18.5at%Al-0.450at%Re-2.86at%Ta for the γ' phase and Ni-13.8at%Al-1.77at%Re-0.792at%Ta for the γ phase. Consequently, the present model demonstrated γ' precipitation with the growth and interaction of precipitates in the quaternary system, by coupling with thermodynamic description in the CALPHAD method. In the next section, a practical alloy, TMS-75, is simulated.

Phase-Field Simulation and Atom Probe Analysis of TMS-75

A third-generation Ni-base superalloy, TMS-75 contains Al, Co, Cr, Mo, Re, Ta, W, and Hf. Its nominal composition is Ni-6Al-12Co-3Cr-2Mo-5Re-6Ta-6W-0.1Hf in weight %, and Ni-13.7Al-12.6Co-3.6Cr-1.3Mo-1.7Re-2.1Ta-2.0W-0.01Hf in atomic % [10,11]. For simplification, Hf was excluded from present calculations. At first, γ' volume fraction, γ and γ' phase compositions were compared between the calculated prediction and experimental observations as shown in Table 1. According to Murakumo et al.[10], the observed γ' volume fraction of TMS-75 was about 0.55 at 1100°C . On the other hand, the present CALPHAD predicted 0.51 of the volume fraction for this alloy. Yokokawa et al. [11] measured the γ and γ' phase compositions using an electron probe x-ray microanalyzer (EPMA), followed by water quenching after heat treatment at 1100°C as shown in Table 1. Both the predicted and measured compositions and partitioning behavior show good agreement, even though the phase-equilibrium information for higher component systems are determined from the extrapolation of binary, ternary and quaternary systems in the CALPHAD method. The observed γ' composition of Co is slightly larger than the predicted one. W is preferred to partition into much more γ phase in the calculated result. In this alloy, Al and Ta were rich in the γ' phases, whereas Co, Cr, Mo, Re, and W were rich in the γ phase.

To investigate the temporal evolution of alloying element compositions experimentally, microstructural observation and atom-probe microanalyses of TMS-75 were performed. Figure 3 shows microstructures of TMS-75 after heat treatment at 1100°C for 15 min (a) or 4 h (b). In the alloy heat treated for 15 min, fine spherical γ' precipitates were observed because the growth of γ' phases at this stage was mainly controlled by the increasing isotropic interfacial energy. In the alloy that had been heat treated for 4 h depicted in Fig. 3(b), the morphology of grown γ' precipitates was aligned cuboidally along a certain orientation.

Figure 4 shows the composition profiles obtained using atom-probe analyses for these alloys: heat-treated for 15 min (a) or 4 h

(b). In the alloy that was heat treated for 15 min, small γ' precipitates, where the Al mole fraction is about 0.2, were easily detected. At the γ/γ' interface denoted by I, the local equilibrium was significantly observed, especially for Re. In addition, Al and Ta in a narrow γ channel next to small precipitates shown by arrows G and H in Fig. 4 (a) were depleted. On the other hand, Co and Cr and other refractory elements such as Mo, Re, and W supersaturated in its γ phase. In Fig. 4(b), the local equilibrium was still observable at the interfaces indicated by arrows of J and K because that the γ' phase remained under the growth process at this stage, i.e. the system had not reached a thermodynamic equilibrium state. These phenomena were simulated using the phase-field method.

Figure 5 shows snapshots of simulated temporal evolution of alloying element composition profiles in TMS-75 at 1100°C . At the initial stage after nucleation, Al in a narrow γ channel shown by an arrow L in Fig. 5(a) was depleted. The γ formers, such as Co, Cr, Mo, Re, and W, supersaturated in the γ phase. In the γ' growth process presented in Fig. 5(b), almost all elements at the interfaces of small precipitates had almost equilibrium compositions, except for Re and W. On the other hand, at the interface of γ' and large γ phases shown by an arrow M, a local equilibrium was confirmed for all elements. In Figs. 5(c) and 5(d), a small γ' phase shrank and disappeared, as shown by an arrow N. Such a so-called *Ostwald-ripening* process was observed in the simulation. Its Re profile was very characteristic, like a *spinning top* as presented in Fig. 5(c) because the Re's local equilibria at γ/γ' interfaces of both sides of the precipitate merged. Therefore, the simulated temporal evolution of composition profiles at the γ/γ' interface agreed well qualitatively with the experimental observations, considering the simplicity of its multi-component model. In general, it is difficult to detect, experimentally, such temporal evolution of compositions during γ' coalescence and Ostwald-ripening process. Such multi-component simulation is necessary to predict the microstructure evolution.

It is noteworthy that the present simulated system incorporated no composition-dependent elastic energy stemming from the elastic inhomogeneity for simplification. Such elastic energy would affect microstructure morphology such as the γ' precipitate alignment, precipitate dispersion, and precipitate shapes. Moreover, the simulated system size is smaller than the real one because of the computational scheme and model limits. Therefore, the growth rate of the γ' phase does not correspond quantitatively to experimental results. Such incorporation of elastic energy effects and increase of the length scale are our next objectives. Nevertheless, it was concluded from results of this study that the present modeling technique is very useful to design new alloys as well as process conditions.

Figure 3 SEM images of TMS-75 after heat treatment at 1100°C for 15min (a) or 4 h (b).

Figure 4 Composition profiles of TMS-75 obtained using atom-probe analyses: heat treated at 1100°C for 15min (a) or for 4 h (b).

Figure 5 Temporal evolution of alloying-element composition profiles in TMS-75 at 1100°C.

Conclusions

New multi-component phase-field modeling coupled with the CALPHAD method was developed by adopting the γ/γ' single formalism of the Gibbs free energy in the CALPHAD for the chemical free energy in the phase-field method. Using this coupling method, the γ' phase precipitation in a quaternary Ni-Al-Re-Ta system and a third-generation superalloy TMS-75 was simulated. These simulations demonstrated the local equilibrium at γ/γ' interfaces, growth of γ' precipitates, precipitate interactions such as coalescence, and the Ostwald-ripening process. The simulated temporal evolution of composition profiles was qualitatively reasonable, compared to experimental observations using atom-probe analyses. This modeling technique can contribute to alloy design, process design and lifetime assessment of high-temperature turbine blades currently in service.

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