Phase-Field Simulation on the Formation and Collapse Processes of the Rafted Structure in Ni-Based Superalloys

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In Ni-based superalloys, the rafted structure is known to form in the early stage of creep and to get into wavy morphology in the final stage of creep at elevated temperatures. This rafting phenomenon is essentially related to the anisotropic relaxation of the lattice mismatch between the γ and γ′ phases due to the creep strain under the external stress. In this study, in order to simulate comprehensively from the formation to collapse processes of the rafted structure by the phase-field method, a new idea that the anisotropy increases with simulation time is employed in the calculation of the elastic strain energy in alloy. This idea corresponds to the phenomenon that creep strain increases with creep time. The results are in good agreement with the microstructural change observed in practical Ni-based alloys. [doi:10.2320/matertrans.MBW200709]

1. Introduction

Ni-based superalloys consisting of the precipitated γ′ phase (L12 structure) in the γ matrix with face-centered cubic structure are applied to gas turbine materials because of their excellent mechanical properties such as creep strength at high temperatures. In aging process, the cuboidal structure are applied to gas turbine materials because of their

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In order to simulate complicated microstructures by the phase-field method using a set of field variables which change continuously in the interfaces. There have been some models of the phase-field method for simulating microstructure evolutions in Ni-Al alloys.12–14) Also, there are a few reports about the simulation of the rafting phenomena,15) but a comprehensive simulation from the formation to the collapse of the rafted structure has not been reported as long as we know.

The purpose of this study is to perform the comprehensive simulation of the change in the rafted structure corresponding to creep deformation using the phase-field method based on the elastic strain energy calculation taking account of the anisotropic relaxation of the lattice mismatch.

2. Calculation Model

In order to simulate morphological change of the γ and γ′ phases, the volume fraction of the γ′ phase, f(γ′), and three long-range order parameters si(γ′, t) (i = 1, 2, 3) which describe the four different domains in the L12 phase structure are chosen as field variables. These field variables vary spatially (r) and temporally (t). Usually, alloy composition, c(γ′), is used as a field variable,12–15) but in this study, f(γ′, t) is used instead of c(γ′, t), because f(γ′, t) is suitable to treat the multi-component system when the phase field method is applied to the practical Ni-based alloys. The temporal evolution of the field variables is given by solving the following Cahn-Hilliard and Ginzburg-Landau equations:

\[ \frac{∂f(γ', t)}{∂t} = M∇^2 \frac{δG_{sys}}{δf(γ', t)}, \]  
\[ \frac{∂s_i(γ', t)}{∂t} = -L \frac{δG_{sys}}{δs_i(γ', t)} (i = 1, 2, 3), \]
(gradient terms) \(E_{\text{surf}}\) and elastic strain energy \(E_{\text{el}}\), and it is written as

\[
G_{\text{sys}} = \int_{\mathcal{R}} \left[ G_{\text{f}}(f, s_i) + E_{\text{surf}}(f, s_i) + E_{\text{el}}(f, s_i) \right] d\mathbf{r}. \tag{3}
\]

In this study, the Gibbs free energy curve of \((\gamma + \gamma')\) two-phase region is expressed by the chemical free energy function proposed by Koyama.\(^\text{16}\)

\[
G_r = 16W_1f^2(1-f)^2 + W_2(f - s_1s_2s_3)^2, \tag{4}
\]

where \(W_1\) and \(W_2\) are the coefficients determined by the Gibbs energy calculation based on the sub-lattice model using the thermodynamic database. The first term in eq. \(4\) is the double-well function which assures the separation of \(\gamma\) and \(\gamma'\) phases. The second term assures the ordering of the \(\gamma\) phase and the local minima of \(G_r\) appear at \((s_1, s_2, s_3) = (1, 1, 1), (1, -1, -1), (-1, 1, -1)\) and \((-1, -1, 1)\), because \(f = 1\) where the \(\gamma'\) phase exists.\(^\text{12}\) These four cases correspond to the four different domains in the \(L1_2\) structure. Figure 1 shows the calculated Gibbs energy curve of \((\gamma + \gamma')\) two-phase region in Ni-Al alloy at 1273 K based on the two sub-lattice model. The values of \(W_1\) and \(W_2\) can be obtained as the values shown in Fig. 1. In a multi-component system, Gibbs energy curve of the \((\gamma + \gamma')\) two-phase region gives \(W_1\) and \(W_2\) values in a similar manner used in Ni-Al system.

In this study, the interfacial energy is employed as the gradient energy of the field variables, that is

\[
E_{\text{surf}} = \kappa_f (\nabla f)^2 + \frac{1}{2} \kappa_s [|\nabla s_1|^2 + |\nabla s_2|^2 + |\nabla s_3|^2], \tag{5}
\]

where \(\kappa_f\) and \(\kappa_s\) are the gradient energy coefficients.\(^\text{17}\) Since accurate data on experimental interfacial energy is not available even in Ni-Al binary alloy, the gradient energy coefficients are determined so that the calculation results can reproduce the microstructure of the practical Ni-Al alloys.

The elastic strain energy arising from the lattice misfit between the \(\gamma\) and \(\gamma'\) phases is estimated based on the micromechanics.\(^\text{18,19}\) The elastic strain is expressed as

\[
\varepsilon_{ij}^0(r, t) = \varepsilon_{ij}^1(r, t) - \varepsilon_{ij}^0(r, t), \tag{6}
\]

where \(\varepsilon_{ij}^1\) and \(\varepsilon_{ij}^0\) represent the constrained strain and eigenstrain, respectively. Here, the eigenstrain is regarded as the liner function of \(f(r, t)\) and expressed as

\[
\varepsilon_{ij}^0(r, t) = \varepsilon_{ij}^0[f(r, t) - f_0], \tag{7}
\]

where \(\varepsilon_{ij}^0\) represents the total misfit strain (eigenstrain) and \(f_0\) is the average volume fraction of the \(\gamma'\) phase. In the elastic equilibrium condition, the elastic strain is written as

\[
\varepsilon_{ij}^1(r, t) = \frac{1}{2} C_{ijkl} k_{ij} k_{kl} \tag{8}
\]

Here, \(C_{ijkl}\) is the elastic-constant tensor and is assumed to be elastically anisotropic. \(k\) is the unit vector of the reciprocal-space vector \(k\), \(\varepsilon_{ij}^1[k, t]\) and \(\varepsilon_{ij}^0[k, t]\) are the Fourier transformation of \(\varepsilon_{ij}^1(r, t)\) and \(\varepsilon_{ij}^0(r, t)\), respectively, and \(\Omega_{\gamma'}^{-1}(k)\) is given by \(\Omega_{\gamma'}^{-1}(k) = C_{ijkl} k_i k_j\). The elastic strain energy is then given by

\[
E_{\text{el}} = \frac{1}{2} \sum_{ij} [\varepsilon_{ij}^1(r, t) - \varepsilon_{ij}^0(r, t)][\varepsilon_{ij}^r(r, t) - \varepsilon_{ij}^0(r, t)]. \tag{9}
\]

The misfit strain between \(\gamma\) and \(\gamma'\) phases is given by

\[
\varepsilon^T = \begin{pmatrix} \varepsilon_0 & 0 & 0 \\ 0 & \varepsilon_0 & 0 \\ 0 & 0 & \varepsilon_0 \end{pmatrix}, \tag{10}
\]

and

\[
\varepsilon_0 = \frac{a_{\gamma'} - a_{\gamma}}{a_{\gamma'}}. \tag{11}
\]

Here, \(a_{\gamma'}\) and \(a_{\gamma}\) are the lattice parameters of the \(\gamma\) and \(\gamma'\) phases, respectively.

In this study, the plastic deformation observed in creep experiments are introduced into the matrix with the misfit strain by \(\varepsilon_p\) along the [001] tensile axis.\(^\text{11}\) In this case, the lateral directions undergo a plastic strain of \(-\varepsilon_p/2\) and hence the misfit strain of the the \(\gamma'\) phase induced by the creep deformation is expressed as

\[
\varepsilon^p = \begin{pmatrix} \varepsilon_p/2 & 0 & 0 \\ 0 & \varepsilon_p/2 & 0 \\ 0 & 0 & -\varepsilon_p \end{pmatrix}. \tag{12}
\]

As a result, when the misfit strain and the creep deformation-induced misfit strain (creep strain) exist, the total misfit strain (eigenstrain) \(\varepsilon^*\) is expressed as the sum of \(\varepsilon^T\) and \(\varepsilon^p\), that is

\[
\varepsilon^* = \varepsilon^T + \varepsilon^p. \tag{13}
\]

3. Results and Discussion

3.1 Elastic strain energy

The elastic constant of the \(\gamma\) phase is different from that of the \(\gamma'\) phase, but elastic strain energy caused by the difference
in the elastic constant is negligible when the creep strain exists. In this study, therefore, the same elastic constants as pure Ni, which are \( c_{11} = 250.8 \text{ GPa}, \quad c_{12} = 150.0 \text{ GPa} \) and \( c_{44} = 123.5 \text{ GPa} \) were used for the two phases in the calculation of the elastic strain energy. Figure 2 shows change in the elastic strain energy with the shape change of the \( \gamma' \) phase as a function of \( 1 - c/a \), where the shape of the \( \gamma' \) phase is represented by \( a \times a \times c \) in dimensions. In this calculation, \( \epsilon_0 = -0.0133 \) was used as the misfit strain and the volume fraction of the \( \gamma' \) phase was set to be 0.5. In Fig. 2, the calculation results are shown in three cases, i.e., the creep strain \( \epsilon_p \) of the \( \gamma' \) phase along the [001] direction are 0 ( ), 0.0266 ( ) and 0.0466 ( ), respectively. Here, the axial ratio \( c/a \) of the rod-shape and the disc-shape of the \( \gamma' \) phases are assumed to be 1.414 and 0.5, respectively, when the volume fraction of the \( \gamma' \) phase was set to be 0.5. Generally, the elastic strain energy increases when the creep strain is introduced, and hence each of the three cases in Fig. 2 cannot be compared with each other. In other words, each case predicts the stable shape of the \( \gamma' \) phase at a creep strain. When \( \epsilon_p = 0 \), the eigenstrain has spherical symmetry and the value of the elastic strain energy is independent of the shape change of the \( \gamma' \) phase. However, as the creep strain \( \epsilon_p \) increases, the spherical symmetry of the eigenstrain changes into tetragonal symmetry. As a result, the elastic strain energy decreases with rafting, though it increases for the rod-shape structure; see the case of \( \epsilon_p = 0.0266 \) in Fig. 2. Thus, the anisotropic relaxation of the lattice mismatch due to the creep strain reduces the elastic strain energy, resulting in the change in the shape of the \( \gamma' \) phase from the cuboidal structure to the rafted structure. Here, if the creep deformation proceeds until \( \epsilon_{11}' = \epsilon_{22}' \) becomes a positive value, i.e. if \( \epsilon_p \) exceeds 0.0266 in this case, a remarkable result is obtained concerning the stable shape of the \( \gamma' \) phase. Figure 3 shows the change in the elastic strain energy at \( 1 - c/a = 0.5 \) with increasing the creep strain gradually. From the figure, it can be said that the rafted structure becomes stable as the creep strain increases, but it becomes unstable when \( \epsilon_p \) exceeds 0.0266, i.e. when \( \epsilon_{11}' = \epsilon_{22}' \) becomes a positive value. Figure 4 shows the relation between the stable shape of the \( \gamma' \) phase and the ratio of \( t_1 = \epsilon_{11}'/\epsilon_{22}' \). When the creep strain exists and \( t_1 \) is between 0 and 1, the (001) raft is the stable structure and the rafting phenomena occurs. However, if the creep strain increases and \( t_1 \) becomes less than 0, i.e. if \( \epsilon_{11}' = \epsilon_{22}' \) becomes a positive value and the stable orientation of the raft plane tilts, which leads to the destabilization of the rafted structure.

Based on the analysis of the elastic strain energy as shown in Figs. 2, 3 and 4, the formation and collapse processes of the rafted structure can be understood. Actually there has been a report that the stable orientation of the lamellar plane changes as the creep deformation proceeds.

### 3.2 Phase-field simulation

Simulations were performed by solving the two sets of equations, eqs. (1) and (2) numerically, using the explicit method under the periodic boundary conditions. The coefficients of the chemical free energy in eq. (4) were determined as \( W_1 = 133 \) and \( W_2 = 877.1 \text{ J/mol} \) from the Gibbs energy curve at 1273 K calculated based on the thermodynamic database as shown in Fig. 1. The average volume fraction of the \( \gamma' \) phase was set to be \( f_0 = 0.5 \) and the gradient energy coefficients were chosen as \( \kappa_f = \kappa_s = 5.0 \times 10^{-15} \text{ J m}^2/\text{mol} \). In the calculation of the elastic strain energy, values \( c_{11} = 250.8 \text{ GPa}, \quad c_{12} = 150.0 \text{ GPa}, \quad c_{44} = 123.5 \text{ GPa} \) and \( \epsilon_0 = -0.0133 \) were used in this study. Time step \( \Delta t \) was selected to be 0.2 so as to maintain the stable solution.
Figure 5 shows the 2D morphological evolution of the γ' phase for $f_0 = 0.5$ at 1273 K. The γ' phase is expressed as white area. First of all, simulation was carried out using only the misfit strain without any creep strain up to $t = 5000$, assuming that the specimen was aged simply at this temperature. The result is shown in Fig. 5(a). In this case, the eigenstrain has spherical symmetry, and the cuboidal shape of the γ' phase arranged along the (100) crystallographic directions is observed due to only the anisotropic elastic interaction. After $t = 5000$, the increase of the creep strain at the constant rate $d\varepsilon_p/dt = 6.66 \times 10^{-7}$ under the [001] tensile stress is introduced by assuming that the creep strain increases during creep. This creep strain relaxes the lattice mismatch in (001) plane. Thus, during $t = 5000$–45000 (Fig. 5(a)–(e)), it is observed that the cuboidal γ' structure gradually changes into the rafted structure and the rafted structure becomes stable in terms of the elastic strain energy. When $t$ becomes more than 45000, i.e. when $\varepsilon_{11}^e \neq \varepsilon_{22}^e$ becomes a positive value, the rafted structure collapses and gets into wavy morphology as shown in Fig. 5(e)–(h). This means that the rafted structure becomes unstable. This is consistent with the fact that the rafted structure is stable when $\varepsilon_{11}^e/\varepsilon_{33}^e$ is between 0 and 1, and the rafted structure is unstable when $\varepsilon_{11}^e/\varepsilon_{33}^e$ is less than 0, as mentioned in Section 3.1.

Fig. 5 Results on two-dimensional phase-field simulation showing the change of the γ' phase for $f_0 = 0.5$ at 1273 K. (a) $t = 5000$, (b) $t = 15000$, (c) $t = 25000$, (d) $t = 35000$, (e) $t = 45000$, (f) $t = 55000$, (g) $t = 65000$ and (h) $t = 75000$. Time expressed here are reduced simulation time.
Figure 6(a) shows a microstructure obtained from the experiment of a nickel based superalloy (Ni-14.0 mol%Cr-10.8%Al-2.5%W-1.9%Ta-1.5%Ti-0.3%Mo),1) which was designed for land-base gas-turbines, crept at 1193 K for 1145 h. A part marked by a square in Fig. 6(a) is enlarged in Fig. 6(b). Figure 6(c) shows the simulation microstructure at $t = 75000$ obtained in this study. The simulation result is in good agreement with the microstructural change of practical Ni-based alloys.

4. Conclusions

The phase-field method has been adopted to simulate comprehensively from the formation to collapse process of the rafted structure in Ni-based alloys. In this work, on the basis of the elastic-plastic consideration, anisotropic relaxation of the lattice misfit between the $\gamma$ and $\gamma'$ phases due to the creep strain is introduced by increasing the creep strain $\varepsilon_P$ with simulation time in a constant rate. The microstructural evolution of the $\gamma'$ phase in the creep process is reproduced well in this simulation; the stabilization and the following destabilization of the rafted structure. This result is in good agreement with the microstructural change of practical Ni-based superalloys.

REFERENCES


