3D-FEM calculations of rafting in Ni-base superalloys based on high temperature elastic and lattice parameters

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Abstract

The elastic constants of γ and γ' phases, the component phases of Ni-base superalloys, these belong to 1st, 2nd, and 3rd generation, have been measured at 900°C. Three dimensional FEM calculations have been conducted based on these measured elastic constants and lattice misfit. The elastic constants have been different between generations of superalloys, depending mainly on the amount of Re. The predicted raft structure have been different among the alloys, and it also depends on the introduction of interfacial dislocations.

Introduction

The high-temperature strength of Ni-base superalloys is affected by the structure evolution. A lamella structure often forms under actual use, and the phenomenon is called "rafting". The rafting is controlled by the difference of lattice parameters between γ and γ' phases (lattice misfit), the elastic constants of them, the diffusion rate of alloying elements, etc.^[1]

To understand the structure evolution and predict the high-temperature strength of the alloys, many studies have been conducted on the relationship between the creep strength and the parameter described above.^[1-6]

Elastic constants are important parameters in the design of gas turbines; therefore, many measurements of them in Ni-base superalloys have been conducted. On the other hand, only a few measurements of the elastic constants have been conducted on γ and γ' phases, which are the component phases of Ni-base superalloys.^[7,8] Many model calculations, such as those with a finite element method (FEM), micro mechanics, and phase field method, have been conducted to explain the mechanism of rafting

in Ni-base superalloys.^[3-6] However, almost all of them have been conducted without measuring the elastic constants of multicomponent superalloys. Instead, data extrapolated from some binary alloy systems have been used, and the mechanisms of rafting have been discussed. Because of the lack of measured data, a comparison of their contribution to the rafting mechanism of generations of single-crystal (SC) superalloys was impossible, as was the creep strength.

In this paper, the elastic constants of γ and γ' phases will be given. Measurements were conducted on 1st, 2nd, and 3rd generation SC superalloys. A comparison among them will be made, and the origin of their systematic differences found will be discussed. Three-dimensional (3D) FEM calculations based on the measured elastic constants and the lattice misfits were conducted, and a comparison of the driving force of rafting between the generations of superalloys will be shown.

Experimental procedure

 γ Single-phase alloys and γ' single-phase alloys were prepared for the measurement. The single-phase alloys were designed to have the same chemical composition as the γ phases and the γ' phases of the 1st, 2nd, and 3rd generation SC superalloys. TMS-26,^[9] TMS-82+,^[10] and TMS-75,^[11] which contain 0wt.%, 2.4wt.%, and 5wt.% Re, respectively, were used as model alloys. The chemical compositions of the single-phase alloys shown in Table 1 and Fig. 1 were designated with an alloy design program developed at NIMS, Japan,^[12] so that they would be in equilibrium at 900°C. Single-crystal rectangular parallelepipeda with all surfaces parallel to the (001) were prepared and used for the elastic constant measurement.

Alloy	Phase	Chemical composition (at.%)									
		Ni	Co	Cr	Mo	W	Al	Ti	Та	Hf	Re
TMS-26	γ	bal.	5.41	2.02	0.58	3.03	17.46	-	-	-	-
	γ'	bal.	14.60	12.99	2.29	5.41	3.56	-	-	-	-
TMS-82+	γ	bal.	4.80	1.62	0.56	2.36	17.62	0.88	3.16	0.05	0.15
	γ'	bal.	13.05	11.28	2.04	3.76	4.35	0.14	0.68	-	1.78
TMS-75	γ	bal.	7.77	1.02	0.59	1.88	19.00	-	2.97	0.05	0.34
	γ'	bal.	19.84	7.39	2.33	2.22	5.82	-	0.66	0.01	3.64

Table 1. Chemical composition of the tested single-phase alloys.



Fig. 1. The amount of alloying elements in the single-phase alloys.

The rectangular parallelepiped resonance (RPR) method,^[13] which is a kind of resonant ultrasound spectroscopy (RUS) method,^[14] was utilized for the measurement; with this method, all of the elastic parameters can be obtained from one small singlecrystal specimen. The elastic constants can be determined by measuring the size, weight, and resonant frequency of the specimen.^[15] A narrow temperature distribution is also expected with the use of one small specimen. By setting the specimen inside the vacuum furnace, resonant frequencies were measured, and the elastic constants at 900°C were obtained (Fig. 2).



Fig. 2. Schematic drawing of an elastic constant measurement with the RPR method. The single-crystal specimen is vibrated from the bottom by sweeping a broad frequency range, and the resonance frequency of the specimen is measured.

The 3D-FEM calculation was conducted with the original program. As the FEM method is already well established, the factors related to the accuracy of the calculation are explained below. The element used is a tetrahedron. First, equivalent nodal forces in the top of the tetrahedron were calculated from the stress in the element. The stiffness equations of the total system were derived as follows. First, the nodal forces from the adjoining elements were summed up, then the equations of force equilibrium were resolved.

To simplify the calculation, the unit cell is established. It consists of a hexahedron precipitate phase, γ' , and the cubic parent phase, γ (Fig. 3). The total system presumes that this unit cell is repeated infinitely in three dimensions. Only 1/8th of the unit cell are used in the calculation in consideration of the symmetry of the unit cell. The boundary plane is assumed to be a flat plane under the coherency strain and the load of the external force. Moreover, the total volume of the unit cell is assumed to be constant. The γ phase is assumed to consist of a cube with 1.0 micrometer in length, and the size of the γ' phase, without rafting, is 0.871 micrometer in length for TMS-26, and 0.857 micrometer in length for TMS-82+ and TMS-75. The size of the γ' phase reflects the real volume fraction of the precipitate phase at 900°C in the alloys simulated. It is 66% for TMS-26, and 63% for TMS-82+ and TMS-75. When the raft structure formation was examined, these lengths were changed while keeping the volume fraction constant. The enthalpy change for raft structure formation is given by the expression

$$\Delta F = \Delta E_{costrn} + \Delta E_{intf} + (1/2)(\Delta Y \sigma^2 V / Y_c Y_r).$$
(1)

Each term in the above expression corresponds to the coherency strain, the interface energy, and the external stress. The external stress term contains strain energy induced by external stress and potential energy.^[17]



Fig. 3. The unit cell used to simulate the microstructure of Ni-base superalloys. A 1/8th of the unit cell is used for the FEM calculation.

The calculation is divided into two steps. First, the lattice misfit is given, and the coherency strain field energy is calculated. Next, finite displacement is given in the direction of the tensile axis, and enthalpy is calculated. By taking the enthalpy of the cubic γ and γ' phases as origin, the enthalpy change of expression (1) for the raft structure formation is estimated for the γ' with an arbitrary configuration. As a parameter to show the shape change, the aspect ratio of the γ' phase for x-axis is defined as

 $\rho_{xp} = (p_{lx} - p_{l0})/(p_{lc} - p_{l0}),$

where p_{l0} and p_{lx} represents the length of the precipitate before and after the rafting, respectively, and p_{lc} represents the length of the unit cell. When two adjacent γ' phases are connected, the raft structure formation is completed. In this calculation, the aspect ratio at this condition is defined as 1.0/0.871=1.148 for TMS-26, and 1.0/0.857=1.167 for TMS-82+ and TMS-75. The measured elastic constants and the lattice misfits were attributed to the calculations. The lattice misfit of the alloys with coherent γ/γ' interface is used in the calculation. The use of them with relaxed γ/γ' interface is recomended, but it is often difficult to measure because tetragonal close packed (TCP) phase often appears while the specimen is annealed to obtain relaxed γ/γ' interface. Anyway, it will not affect the results when we make comparison of the alloys these belong to different generations. The target temperature of the calculation was 900°C.

Results and discussion

i) High temperature elastic constants

The measured elastic constants are summarized in Table 2. The lattice misfit, δ , is also shown in the table. The Young's modulus along the <100> direction and the anisotropy factor, A, both calculated from the elastic constants, are shown in Figures 4 and 5, respectively. These two parameters clearly show dependence on the amount of refractory elements, i.e. Mo, W, Ta, and Re. At the same time, in the γ' phase, these parameters show dependence on the generation of alloys.

The generation dependence described above can be explained as below. In higher-generation superalloys, the amount of alloyed Re is higher compared with lower generation superalloys. In these alloys, the amount of other refractory elements becomes lower to prevent the formation of tetragonal close packed (TCP) phases. In other words, Re is added in the expense of other refractory elements in higher-generation superalloys as shown in Fig. 1. Re shows a strong tendency to partition into the γ phase as shown filled markers in Fig. 1. This results in the decrease of refractory elements in the γ' phase, as shown in Figures 1, 4, and 5. The decrease of refractory elements in the γ' phase results in generation dependence of the Young's modulus and anisotropy factor. On the other hand, the amount of refractory elements does not change drastically in the γ phase, as shown in Figures 1, 4, and 5 because decreased Mo, W, and Ta are substituted by Re, resulting in no generation dependence of the Young's modulus and anisotropy factor.

The Young's modulus along the <100> direction of γ' phases previously reported is shown in Fig.6. The NASAIR 100 is a 1st generation superalloy.^[7] In the case of CMSX-4, a typical 2nd generation superalloy, no W, Ta, Mo, nor Re is alloyed on the tested γ' single-phase alloy reported in order to achieve hightemperature stability.^[8] As the previously reported alloys are poorly alloyed ones, the generation dependence is not clear in this figure. On the other hand, the chemical composition dependence is conserved. For a better explanation of the chemical composition dependence on the elastic constants, a more precise analysis on the effect of all alloying elements is required.

Alloy	Phase	<i>C</i> ₁₁ (GPa)	<i>C</i> ₁₂ (GPa)	<i>C</i> ₄₄ (GPa)	$\delta(\%)$	
TMS 26	γ	224	166	97.7	0.10	
11013-20	γ'	218	218 146		-0.19	
TMS 82+	γ	208	154	93.5	0.16	
11013-02	γ'	205	138	101	-0.10	
TMS 75	γ	212	153	98.4	-0.15	
11013-73	γ'	206	140	101		

(2)

Table 2 Elastic constants of the component phases of Ni-base superalloys at 900°C. Lattice misfits of the alloys at 900°C, δ , are also shown.



Fig. 4. The Young's modulus along the <100> direction at 900°C is plotted against the total amount of refractory elements, i.e. W, Ta, Mo, and Re. The solid line is a guide for the eyes.



Fig. 5. The anisotropy factor, A, at 900°C is plotted against the total amount of refractory elements, i.e. W, Ta, Mo, and Re. The solid line is a guide for the eyes.



Fig. 6. The Young's modulus along the <100> direction of the γ' phases of superalloys at 900°C, which appeared in previous reports,^[7, 8] are plotted against the total amount of refractory elements, i.e. W, Ta, Mo, and Re. The solid line is a guide for the eyes.

ii) Distribution of the equivalent true stress

The results of FEM calculations without external stress are shown in Fig. 7. On the figure, one fourth of the plane A in Fig. 3 is shown. The results show good agreement, qualitatively, with the 2D-FEM calculation of CMSX-3 conducted by Pollock et al.,^[3] who used estimated elastic constants. The equivalent true stress inside the γ' precipitate is almost similar in all the alloys calculated. On the other hand, the equivalent true stress at the γ channel is not similar. This is clearly shown in Fig. 8, which shows one fourth of the plane B in Fig. 3. The equivalent true stress is the largest in the TMS-26, a 1st generation superalloy, and the smallest in TMS-82+,

a 2^{nd} generation superalloy and TMS-75, a 3^{rd} generation superalloy.

Once external stress is applied, the equivalent true stress inside the γ' precipitate becomes different among the alloys. In the case of tensile stress, as shown in Fig. 9, it is the largest in TMS-26 and the smallest in TMS-82+ and TMS-75. The equivalent true stress at the γ channel parallel to the tensile axis becomes smaller. On the other hand, the equivalent true stress at the γ channel normal to the tensile axis becomes larger as shown in Fig. 10. The difference of the equivalent true stress between the γ channel parallel to the tensile axis and normal to it is the largest in TMS-26 and the smallest in TMS-82+ and TMS-75.

The difference of the equivalent true stress inside the γ' precipitate among the alloys while externally stressed is governed by the difference of the elastic constants between the alloys. The equivalent true stress depends on the elastic constant, lattice misfit, and external stress. Without external stress and with lattice misfit, the equivalent true stress inside the γ' precipitate is almost similar in all the alloys. The effect of lattice misfit to the difference of the equivalent true stress distribution among the alloys should be the same with or without the external stress. The external tensile stresses applied for TMS-26, TMS-82+, and TMS-75 were 147.36MPa, 137.56MPa, and 140.62MPa, respectively. The difference of the external tensile stress applied on the alloys is less than 8%, much smaller than the difference of the equivalent true stress distribution inside the γ' precipitate among the alloys. It is concluded that the difference is due to the difference of the elastic constants among these alloys.

The difference of the equivalent true stress between the γ channel parallel to the tensile axis and normal to it is affected by the difference of the elastic constants and lattice misfit among the alloys. Note that this difference is a key piece of data to explain the dislocation penetration anisotropy into the γ channel normal and parallel to the stressed axis. ^[3, 17] At the same time the distribution of equivalent true stress on the γ channel, as shown in Fig. 10, is a key piece of data to explain the mechanism of dislocation penetration into the γ channel, that will be discussed elsewhere, which cannot be obtained with 2D-calculations.



Fig. 7. Distribution of the equivalent true stress on plane A of Fig. 3 is plotted. External stress is not applied.



Fig. 8. Distribution of the equivalent true stress on plane B of Fig. 3 is plotted. External stress is not applied.





Fig. 9. Distribution of the equivalent true stress on plane A of Fig. 3 is plotted. External stress is applied.



Fig. 10. Distribution of the equivalent true stress on plane B of Fig. 3 is plotted. External stress is applied.

iii) Rafting without interfacial dislocation network

In this section, raft structure formation without the formation of dislocation network at the γ - γ' interface will be discussed. In some papers, the rafting tendency is explained from the viewpoint of the stress distribution in a cubic precipitate model. Actually, this way to explain it is risky, as the stress distribution changes with the structure evolution. The free energy change accompanied with the structure evolution should be compared to explain the driving force for the raft structure formation. 3D-calculation is required to obtain the free energy. It is impossible to obtain the free energy of the cubic or rafted structure, which has a 3Ddistribution of the equivalent true stress, as shown in section ii, from 2D-calculations.

The rafting tendency of the alloys was examined by calculating the change of the enthalpy density accompanied with the change of the aspect ratio of the γ' hexahedron under stress. The results are shown in Fig. 11. In this calculation, an interfacial dislocation network is not introduced. The plate-like rafted structure is called "plate", and the rod-like rafted structure is called "rod" in the figure and hereafter. The enthalpy density of the cubic precipitate is taken as the origin (zero in the figure), and the difference from it, ΔF in eq. (1), is plotted against the aspect ratio, which is defined in eq. (2). TMS-26 and TMS-75 show a tendency to form a plate parallel to the tensile direction, and TMS-82+ shows a tendency to form a rod parallel to the tensile direction. ΔF is the smallest in TMS-26 and the largest in TMS-75. In other words, TMS-26 has the largest driving force for structure evolution, and TMS-75 the smallest.

In the case of TMS-26, which shows the largest driving force, ΔF for plate normal to the tensile axis and parallel to it is almost similar. The difference between them is a contribution of the external tensile stress as described below. ΔF is sum of two terms; morphology change term, first two terms in eq. (1), and external stress term, the third term in eq. (1). The morphology change term, the enthalpy density change accompanied with the morphology change for plate normal to the tensile axis and parallel to it, must be the same without the external tensile stress because without external stress, they show the same stress distribution. The external stress term is schematically explained in Fig. 12. The figure shows the enthalpy density change accompanied with applying external stress to cubic, plate normal to the tensile axis, and parallel to it. When we apply external stress to them, plate normal to the tensile axis can be treated as Reuss model, and plate parallel to it can be treated as Voigt model. In general, the Reuss model gives us the smallest macroscopic Young's modulus, and the Voigt model the largest. Taking potential energy of the external stress into account, the enthalpy density change is the largest in plate normal to the tensile axis, and the smallest in plate parallel to it. ^[17] If we take the cubic structure as basis, the enthalpy density becomes larger in plate parallel to the tensile axis, and smaller in plate normal to it as shown in Fig. 12. It is concluded that the difference of ΔF between plate normal to the tensile axis and parallel to it in Fig. 11 results from the external stress, and the major part of the driving force for rafting results from the morphology change itself in the case of TMS-26.

Nevertheless, in the case of TMS-26, the ΔF of plate parallel to the tensile axis is smaller than that of plate normal to the tensile axis. It is opposite to what is predicted by the explanation described above. It suggests that the Reuss model does not give us smaller macroscopic Young's modulus than the Voigt model, which can be occurred by the effect of Poisson's ratio. ^[18] The difference of most stable structure among the alloys we calculated and the difference between our results and the result of the 3D micro-mechanics calculation conducted by Ichitsubo et al.^[17] may result from this effect of Poisson's ratio. Precise explanation of the effect of Poisson's ratio on the Ni-base superalloys will be shown in a separate paper.



Fig. 11. The enthalpy density change accompanied with the structure evolution, rafting, is plotted against the aspect ratio defined in eq. (2). An interfacial dislocation network is not introduced. The dotted line represents results on TMS-26, a 1^{st} generation superalloy. The phantom line represents results on TMS-82+, a 2^{nd} generation superalloy. The solid line represents results on TMS-75, a 3^{rd} generation superalloy.



Fig. 12. The enthalpy density change accompanied with applying external stress to cubic precipitate, plate normal to the tensile axis, and parallel to it. The relative enthalpy density from cubic precipitate is schematically shown. ΔY is a difference of Young's modulus between cubic and rafted structure appeared in eq. (1).

ix) The effect of interfacial stress relaxation anisotropy

It is often emphasized that the rafting tendency is ruled by the stress relaxation on the γ - γ' interface normal or parallel to the tensile direction which arise from the formation of dislocation network at these interfaces.^[16, 17] The difference of the driving force for rafting with and without this relaxation is shown in Fig. 13. When the γ - γ' interface normal to the tensile direction is relaxed, the ΔF of the plate normal to the tensile direction becomes smallest and the ΔF of the plate parallel to the tensile direction becomes slightly larger.

On the other hand, when the γ - γ' interface parallel to the tensile direction is relaxed, the plate parallel to the tensile direction kept most stable. Actually, it can not occur on alloys with negative lattice misfit under tensile creep because dislocations prefer to penetrate into the γ channel normal to the tensile direction than into the γ channel parallel to the tensile direction.^[19]



Relative aspect ratio of the precipitate

The relationship described above is plotted in the broad range of stress relaxation in Fig. 14. In the figure, ΔF is plotted against the degree of interfacial stress relaxation, in other words, the density of interfacial dislocation network normal to the tensile direction. With or without the external stress, ΔF were almost similar.

The driving force for raft structure formation is the enthalpy density difference between the cubic precipitate and plate at given degree of relaxation as shown in the figure. As expected, the driving force for raft structure formation normal to the tensile axis at completely relaxed state is larger than that without relaxation. At the same time, the driving force for raft structure formation parallel to the tensile axis decreased with the increase of degree of interfacial stress relaxation. It is concluded that these two effects described above accelerate the formation of raft structure normal to the tensile direction when the γ - γ' interface normal to the tensile direction is relaxed.



Fig. 13. The enthalpy density change with the structure evolution, rafting, is plotted against the aspect ratio defined in eq. (2). The interfacial dislocation network is introduced. The calculation was conducted on TMS-26, a 1st generation superalloy. The dotted line represents results without interfacial stress relaxation, the phantom line represents results for 30% of the interfacial stress normal to the tensile axis is relaxed, and solid line represents results for 30% of the interfacial stress parallel to the tensile axis is relaxed.

Fig. 14. The relationship between the interfacial stress relaxation anisotropy and enthalpy density difference, ΔF . The γ - γ' interface normal to the tensile direction is relaxed. In horizontal axis, zero means that the interface is not relaxed at all and one means that the interfacial stress normal to the tensile axis is completely relaxed. The calculation was conducted on TMS-26, a 1st generation superalloy.

Conclusion

The elastic constant of γ and γ' phases on 1^{st} , 2^{nd} and 3^{rd} generation single-crystal Ni-base superalloys at 900°C was measured. The Young's modulus and anisotropy factor showed generation dependence, which can be explained as an alloying effect of Re.

3D-FEM calculations were conducted based on the measured elastic constants and the lattice misfit. The calculations were conducted on the models with coherent γ - γ' interface and that with semi-coherent one. In the case of coherent models,

- 1. The expected raft structure depends on individual alloys and not depends on the generation of alloys. It may be the effect of Poisson's ratio.
- 2. The driving force for raft structure formation is the highest in 1st generation superalloy, TMS-26, and the lowest in 3rd generation superalloy, TMS-75.

In the case of partially relaxed (semi-coherent) models,

- 3. When the γ - γ' interface normal to the tensile direction was relaxed, the raft structure normal to the tensile axis formed. It formed because the driving force for raft structure formation normal to the tensile axis increased with the degree of interfacial stress relaxation increased, and the driving force for raft structure formation parallel to the tensile axis decreased with the degree of interfacial stress relaxation increased.
- 4. The plate-like rafted structure parallel to the tensile direction became the most stable when the γ - γ' interface parallel to the tensile direction is relaxed. Actually, it can not occur on alloys with negative lattice misfit under tensile creep condition because dislocations prefer to penetrate into the γ channel normal to the tensile direction than into the γ channel parallel to the tensile direction.

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