DEFORMATION MECHANISMS AND MICROSTRUCTURAL EVOLUTION OF $\gamma + \gamma'$ DUPLEX AGGREGATES GENERATED DURING THERMOMECHANICAL PROCESSING OF NICKEL-BASE SUPERALLOYS

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Abstract

Deficiency in hot plasticity is a major limiting factor for the development of disc alloys with superior mechanical properties and temperature capabilities. The deformation behavior of a series of highly alloyed disc alloys with γ' volume fractions from 26% to 63% was experimentally and analytically investigated in this work. Flow stress data and the as-deformed structures were obtained using isothermal compression tests in which the prior thermomechanical history of experimental alloys was also taken into consideration. The results reveal that the two structural categories, the γ' dispersion and the $\gamma + \gamma'$ duplex, exhibit significant differences in deformation mechanisms. Among all of the experimental alloys, the stability of the $\gamma + \gamma'$ duplex structures increases with the increasing concentration of γ' -forming elements. The formation of the $\gamma + \gamma'$ duplex always results in a decrease in flow stress and a promotion of plasticity. In particular, the $\gamma + \gamma'$ duplex structures exhibit the ability to maintain superplasticity at a high strain rate up to 0.1 s^{-1} . The distribution of strain rate sensitivity and activation energy in the relationships with temperature and strain rate accurately identifies a specific domain within which $\gamma + \gamma'$ duplex exhibits superplasticity. Both the microstructural characterization and the mechanical interpretation confirm that the inherent characteristics of the $\gamma + \gamma'$ duplex, including flow softening, plasticity promotion and high-strain-rate superplasticity, mainly originate from a strain-induced, high-velocity $\gamma - \gamma'$ interface reaction mechanism, which is similar to that of cellular reaction or eutectoid transformation. For the alloys with up to more than 60% of γ' volume fraction, by producing a $\gamma + \gamma'$ duplex using TMP, the hot working process, including billet conversion and microstructure customization, can be achieved in a cost-effective manner on an industrial scale.

Introduction

For high-pressure compressors and turbine discs for gas turbine engines, the development of advanced materials has mainly focused on increasing the volume fraction of L_{12} -type γ' precipitates in polycrystalline nickel-base superalloys, considering the fact that elevated temperature properties are directly proportional to the γ' volume fraction over the range from 3% to 65% at temperatures from 700 °C to 980 °C [1-5]. Nevertheless, a continuous increase in the concentration of alloying elements, especially γ' -forming elements, inevitably leads to a steady deterioration in hot plasticity and makes microstructural customization more difficult during the hot working process on the highly alloyed disc alloys. In fact, the deficiency in plasticity is one of the primary limitations that impede the development of high performance disc alloys, particularly for those alloys that are produced via ingot metallurgy techniques [6, 7]. Therefore, much

attention has been paid to deformation mechanisms and the microstructural evolution during hot working so that a robust processing window can still be identified for alloys with ever-increasing γ' volume fractions [1, 2, 8, 9]. For disc alloys with moderate to high volume fractions of the γ' phase (35%~65% upon aging, more than 10% in the hot working temperature range), γ' precipitates exhibit a wide range in morphology and size distribution depending on certain processing conditions [10]. In particular, there exist two characteristic $\gamma - \gamma'$ two-phase configurations during hot working. One is a dispersion of γ' particles in the γ matrix, and the other is the equiaxed $\gamma + \gamma'$ duplex aggregates. $\gamma + \gamma'$ duplex structures are usually generated during certain thermomechanical processing (TMP) procedures, meanwhile, their extraordinary hot working plasticity and flow softening effect have long been known [11, 12]. Nonetheless, the characteristic deformation mechanism of the $\gamma + \gamma'$ duplex structure has not yet been evaluated comprehensively.

Dynamic recrystallization of the γ matrix and precipitation of the γ' phase have been extensively studied as the two major controlling processes that dominate the deformation behavior of nickel-base superalloys during hot working, and developments in TMP have also mainly focused on obtaining a fine-grained γ matrix structure. However, the grain refinement process during TMP is usually combined with a topological transformation in the $\gamma - \gamma'$ two-phase configuration, i.e., from the γ' dispersion to the $\gamma + \gamma'$ duplex. In addition, the topological transition of the two-phase relationship inevitably leads to a change in the deformation mechanisms, which has not been well elaborated [11-14]. The difference between the flow behavior of the γ' dispersion and the $\gamma + \gamma'$ duplex means that the flow stress is no longer a single-valued function of major state variables (i.e., temperature and strain rate) in the subsolvus temperature region because the topological features act as an extra variable [11, 13]. Correspondingly, the constitutive relationships for such a special circumstance have not yet been well established. Considering that a comprehensive understanding of the characteristics of $\gamma + \gamma'$ duplex structures may contribute to the development of more efficient hot working techniques for highly alloyed disc alloys, the microstructural evolution and deformation mechanism of the $\gamma + \gamma'$ duplex during hot working processes are still under further investigation.

In this research, isothermal uniaxial compression tests were conducted to obtain both flow stress data and the as-deformed structures in relation to temperature and strain rate. To ensure the generality of the results, a series of disc alloys with characteristic chemical compositions, lattice parameters, and particularly the volume fraction of γ' phase were evaluated in this study. The prior thermomechanical histories of the experimental materials were also taken into consideration as a primary variable. Based on the

experimental flow stress data, the strain rate sensitivity and apparent activation energy were derived. Further, models for describing the mechanical behavior of two-phase structures were also incorporated into the interpretations of the characteristic deformation mechanisms of the highly alloyed superalloys in γ - γ' two-phase region during the hot working process.

Experimental

Materials and specimen preparation

The chemical compositions of experimental alloys are listed in Table I, and the corresponding major characteristic parameters of these alloys are listed in Table II. In Tables I and II, the alloys are sorted according to the increasing volume fraction of γ' precipitates. Alloy GH4586A is derived from René 41, which is characterized by a high concentration of solid-solution alloying elements and exhibits the lowest amount of γ' precipitates among the experimental alloys. Alloy GH4742 and GH4065A exhibit almost the same volume fraction of γ' phase, while many other characteristic parameters, such as the titanium/aluminum ratio, γ' solvus and γ/γ' mismatch, are quite different. In particular, alloy GH4065A can be considered as a cast/wrought version of powder metallurgy René 88 DT and FGH4096, both the microstructures and the mechanical properties of these three alloys are actually identical [15]. The concentration of major alloying elements in alloy GH4975 is very similar to that in Mar-M200 and K465, which results in an extraordinarily high γ' volume fraction (more than 60% after aging at 760 °C) and a high equilibrium γ' solvus (1205 °C) for disc alloys.

All experimental alloys were taken from commercial scale billets that were produced via the standard cast/wrought method, and the conversion of triple melting (VIM+ESR+VAR) ingots (508 mm in diameter for GH4586A, GH4742 and GH4065A, 180 mm in diameter for GH4975) was conducted on an open-die forging press. A series of TMP procedures was introduced to customize the initial microstructures of the experimental alloys. All billets with a gage of 100 mm in diameter and 200 mm in length underwent a two-stage pre-processing procedure listed in Table III (i.e., one of procedures I ~ III), and cylindrical specimens with a gage of 15 mm in diameter and 30 mm in length were taken from these pancakes for isothermal compression experiments on a testing machine. The actual pre-processing procedure for a given specimen was selected according to the

deformation temperature T_f during the final isothermal compression test on the testing machine. If the specimens were prepared for supersolvus compression tests (i.e., at a testing temperature T_f equal to or higher than the equilibrium γ' solvus T_s), the billet from which the specimens were taken was processed via procedure I illustrated in Table III. When specimens were prepared for subsolvus compression tests (i.e., at a testing temperature T_f lower than the equilibrium γ' solvus T_s), two billets were separately prepared via procedures II and III (see Table III), thus duplicate specimens with different initial microstructures were obtained.

Isothermal compression tests

The isothermal uniaxial compression tests were conducted on an MTS 810 servo-hydraulic testing machine. The use of sintered Si_3N_4 -ZrO₂ ceramic platens and a resistance furnace provides strictly isothermal conditions for uniaxial compression tests up to 1200 °C. Based on the load and displacement data, which were recorded with a high sampling frequency, the stress-strain curves at various temperatures between 950 °C to 1200 °C and strain rates from 10^{-4} s^{-1} to 1 s^{-1} could be calculated. For each and every subsolvus temperature, duplicate specimens that were respectively prepared via procedure II and III were tested under exactly the same deformation conditions. Therefore the influence of prior thermomechanical history on deformation behavior could be taken into consideration. To obtain the strain rate sensitivity m in a more accurate way, a strain-rate change test was conducted at each of the deformation temperatures, based on which the change in the flow stress for one order of magnitude change in strain rate could be determined. All specimens were *in situ* preheated on the testing machine to the deformation temperature T_f at first, and soaked at T_f for 20 min prior to uniaxial compression. The specimens were water quenched immediately after the deformation to preserve the as-deformed microstructures.

Microstructural characterization methods

Microstructural characterization of the as-deformed specimens was performed using optical microscopy, X-ray diffraction, scanning and transmission electron microscopy. Optical observation was conducted on a LEICA MEF4A microscope. The microstructures were evaluated at higher resolution using a JSM-7800F scanning electron microscope (SEM) equipped with an Oxford Nordlys MAX electron backscattered diffraction (EBSD)

Table I. Chemical compositions of experimental alloys (mass fraction %)

Alloys	C	Co	Cr	W	Mo	Al	Ti	Nb	Fe	Ni
GH4586A	0.049	11.68	18.09	3.05	8.11	1.65	3.31	—	0.05	Bal.
GH4742	0.052	10.40	14.15	—	5.03	2.51	2.56	2.62	0.53	Bal.
GH4065A	0.011	12.98	15.93	4.02	4.03	2.12	3.78	0.72	1.01	Bal.
GH4975	0.115	15.58	7.96	10.22	1.18	5.01	2.49	1.66	0.10	Bal.

Table II. Characteristic parameters of experimental alloys

Alloys	Equilibrium γ' solvus (°C)	γ/γ' misfit at 760 °C (%)	Ti/Al ratio	γ' volume fraction (%)	
				760 °C	$T_{\gamma' \text{ solvus}} - 100$ °C
GH4586A	1072	−0.18	2.0	26	15
GH4742	1103	+0.11	1.0	40	18
GH4065A	1119	−0.02	1.8	42	19
GH4975	1205	−0.27	0.5	63	29

Table III. Prior thermomechanical histories of experimental alloys

Procedures ^a	Multi-cycle upsetting and drawing ^b		Isothermal forging ^c		
	Cycle numbers (<i>n</i>)	T_n (°C)	T_h (°C)	T_d (°C)	Cooling conditions
I	2	T_s	—	T_s	Oil quenching
II	2	T_s	T_s	T_f	Oil quenching
III	3~6	$T_s - (n-1)\Delta T$	T_f	T_f	Oil quenching

* T_n — Annealing temperature prior to the number n cycle of upsetting-drawing.

T_s — Equilibrium γ' solvus of certain experimental alloys.

T_h — Preliminary annealing temperature prior to isothermal forging.

T_d — Actual deformation temperatures of the billets during isothermal forging on hydraulic forging press.

T_f — Deformation temperature that predetermined for a certain specimen during compression on the MTS testing machine.

ΔT — 20~35 °C for the certain alloys.

a — As-forged billets with a gage of 100 mm in diameter and 200 mm in length underwent multi-cycle upsetting and drawing at first, then were isothermally forged into pancake on which the specimens were machined.

b — During every single cycle of upsetting and drawing, the billets were uniaxially compressed to 30% engineering strain at first and then were forged into original dimensions on an open-die forging press.

c — Billets were preliminarily annealed at temperature T_h for 4 hr and then isothermally forged at T_d to 70% engineering strain. All billets were soaked at T_d for 2 hr prior to forging, and were oil quenched immediately after deformation.

detector and an X-Max 50 energy dispersive spectroscopy (EDS) detector. The samples for the EBSD analysis were prepared by an Ar-ion beam cross-section polisher (JEOL IB-09020CP). Because the crystallographic structures and lattice parameters of the two phases are very similar, it is difficult for the EBSD indexing system to distinguish γ' precipitates from the γ matrix. Therefore, a synchronized EDS analysis of major alloying elements was conducted during the EBSD analysis to identify the γ' precipitates according to the chemical composition data. The dislocation configurations were analyzed using a FEI Tecnai G² F20 transmission electron microscope (TEM) operated at 200 kV.

Results and discussion

Microstructural characteristics and flow stress data

The prior thermomechanical history greatly influences the microstructures of the experimental alloys. As a result, the specimens that came from pre-processing procedures I ~ III (illustrated in Table III) exhibit significant discrepancies in their deformation behavior in the subsolvus temperature region. Furthermore, instead of the respective morphologies, size distributions and particularly volume fractions of the two phases, the transition of the topological relationship between the γ' precipitates and the γ grains is combined with a significant change in deformation mechanisms, and therefore results in an extra complexity in the flow behavior. The γ' dispersion and the $\gamma+\gamma'$ duplex are the two major $\gamma-\gamma'$ two-phase configurations obtained in experimental alloys, as exemplified in Figure 1(a) and 1(b), respectively, which are intrinsically different from each other in their two-phase topological relationship.

Pre-processing procedures I and II yield a coarse-grained γ matrix in all experimental alloys. During supersolvus compression tests, the microstructural evolution is generally dominated by dynamic recrystallization and produces coarse-grained equiaxed microstructures. Meanwhile, in the subsolvus temperature range, γ' precipitates play a significant role in the deformation process. All specimens produced by procedure II exhibit coarse-grained γ grains combined with fine, highly dispersed γ' precipitates, as shown in Figure 1(a). Nonetheless, a dispersion of γ' particles in a γ matrix is not the only type of two-phase structures that exists.

Figure 1. Initial microstructures (optical and SEM images) of alloy GH4065A for isothermal compression tests at 1075 °C produced using TMP illustrated in Table III. (a) Coarse-grained γ' dispersive structure produced using procedure II, (b) fine-grained $\gamma+\gamma'$ duplex structure produced using procedure III.

Depending on the proper thermomechanical processing techniques (procedure III in Table III, for instance), a special $\gamma+\gamma'$ duplex structure can be produced, as shown in Figure 1(b). The formation of a $\gamma+\gamma'$ duplex is almost always combined with a drastic reduction in the grain size of the γ matrix (the average grain diameter is usually less than 10 μm) and an evident coalescence of γ' precipitates (extra-large γ' precipitates tend to lose coherency with the γ matrix). The as-deformed $\gamma+\gamma'$ duplex structures of alloy GH4065A are shown in Figure 2. In comparison with Figure 1(b), the EBSD images reveal that the as-deformed microstructures maintain equiaxed and randomly oriented, as shown in Figure 2(a) and 2(b), i.e., no obvious texture developed during deformation.

Under certain combinations of temperature and strain rate, $\gamma+\gamma'$ duplex structures exhibit a strain-independent steady state, i.e., they present no significant differences compared with the initial microstructures. At the certain temperatures close to the γ' solvus, the $\gamma+\gamma'$ duplex can maintain this ability at a high strain rate up to 0.1 s^{-1} . In general, the ability to maintain microstructural stability (unaffected grain morphology and size distribution), whatever the applied strain, implies the occurrence of superplasticity. However, the standard grain-boundary sliding mechanism is indeed very unlikely under an applied strain rate from 0.01 s^{-1} to 0.1 s^{-1} . Therefore, there must be other mechanisms involved in the superplastic deformation at high strain rates. Noticeably, the as-

deformed $\gamma+\gamma'$ duplex structure presents a high density of $\Sigma 3$ twin boundaries, as shown in Figure 2(c). Meanwhile, compared with the corresponding phase map, as shown in Figure 2(d), twin boundaries can be found not only in the γ grains but also in the γ' precipitates. Furthermore, twin boundaries are frequently observed to terminate at phase boundaries, as well as grain boundaries. Figure 2 indicates that twin reactions play a role in the microstructural evolution. In fact, these $\Sigma 3$ boundaries' nucleation and growth processes combine with the dynamic migration of the grain boundaries, and particularly, the $\gamma-\gamma'$ phase boundaries [14]. During deformation, driven by stored energy, high-angle interfaces, including grain boundaries and $\gamma-\gamma'$ phase boundaries, locally reorganize to accommodate the plastic strain. Therefore, the migration of boundaries can dynamically annihilate the dislocations while simultaneously maintaining the grain size and shape. The $\gamma-\gamma'$ phase transformation may provide the extra driving force for boundary migration, the mechanism of which is inherently similar to that of a cellular reaction or eutectoid transformation. The indicated areas in Figure 2 and Figure 3 present a special two-phase configuration (similar to cellular structures that usually form during discontinuous precipitations) as a result of boundary reactions. In the as-deformed $\gamma+\gamma'$ duplex structures, multi-generations of twin boundaries, including the well-developed twins in relatively large grains that distort during the subsequent deformation and the newly developed twins near triple junctions, can be observed at the same time.

Figure 2. EBSD images of the as-deformed microstructure of alloy GH4065A. The specimen with initial microstructures shown in figure 1(b) was isothermally compressed at 1075°C, 0.01 s^{-1} to 70% engineering strain. Band contrast map (a) and corresponding IPF image (b) present a fine-grained, equiaxed $\gamma+\gamma'$ duplex structure. (c) Higher magnification image of the indicated area in (a), the grain boundaries are superimposed on the band contrast map, the black lines represent high angle boundaries ($>15^\circ$) and the red lines represent $\Sigma 3$ twin boundaries (5° tolerance). (d) Corresponding phase map of (c) obtained using synchronized EDS analysis.

TEM observations also reveal that the dislocation-free areas are more likely to be found adjacent to γ - γ' phase boundaries. Whenever the dislocation density increases in a local area, such as shown in Figure 3, an impingement process is triggered by the stored energy (defects density) gradient. As shown in Figure 3(a), dislocation tangles accumulate at twin boundaries in the well-developed grain A, which is surrounded by the dislocation-free areas generated by interface migration [11, 16]. The formation of new twin boundaries on triple junctions can be observed in Figure 3(b), and the migrating interfaces leave the dislocation free areas behind, as shown in Figure 3(b ~ d). Therefore, strain-induced γ - γ' phase interface reaction, as a high velocity deformation mechanism, provides the γ + γ' duplex structures with the ability to superplastically deform at a high strain rate up to 0.1 s^{-1} . In this research, well-developed γ + γ' duplex structures were obtained in alloys GH4742, GH4065A and GH4975 but have not yet been observed in alloy GH4586A. Considering that alloy GH4586A exhibits the lowest volume fraction of γ' precipitates, the proper processing parameters for the generation of γ + γ' duplex must have shifted to a lower temperature and strain rate domain, which is far beyond that in this research.

The flow behavior of the experimental alloys is closely related to their prior thermomechanical histories. Because the alloys have already reached their steady state in both microstructures and flow stresses during the multi-cycle pre-processing listed in Table III, the flow curves derived from experimental data generally reach a steady state after a small plastic strain during isothermal compression tests, while some specimens exhibit a yield drop at the very beginning of the plastic deformation.

Figure 3. TEM images of as-deformed γ + γ' duplex structures of alloy GH4065A, the blue arrows indicate $\Sigma 3$ twin boundaries and the red arrows indicate the migrating interfaces. (a) The γ grain A, which exhibits higher dislocation density than that of neighboring areas, is impinged by surrounding γ grains and γ' precipitates. (b ~ d) Enclosed areas in (a), the migrating interfaces, including both γ grain boundaries and γ - γ' phase boundaries, leave the dislocation free areas behind.

The steady state flow stress data of specimens in relation to the temperature and strain rate are presented in Figure 4. For all experimental alloys, the temperature sensitivity of the flow stress in the subsolvus temperature region is remarkably higher than that in the supersolvus temperature region. Importantly, the flow stress of the γ + γ' duplex structures is significantly lower than that of the γ' dispersion under otherwise identical conditions. In a certain subsolvus temperature range, the flow stress of the γ + γ' duplex is only slightly higher than that in the single-phase region at the same strain rate. In fact, the γ' dispersion and the γ + γ' duplex reach the upper and lower limits of the flow stress in the two-phase region, respectively.

Mechanical interpretations of deformation behavior

The strain rate sensitivity parameter m is defined as a function of the logarithmic values of stress and strain rate as:

$$m = \left. \frac{\partial \ln \sigma}{\partial \ln \dot{\epsilon}} \right|_T \quad (1)$$

where σ and $\dot{\epsilon}$ are the flow stress and the strain rate for a given value of temperature T , respectively, and the apparent activation energy Q at a constant strain rate $\dot{\epsilon}$ can be expressed in the form:

$$Q = \left. \frac{R}{m} \frac{\partial \ln \sigma}{\partial (1/T)} \right|_{\dot{\epsilon}} \quad (2)$$

where R is the gas constant. Based on the data obtained in strain-rate change tests, the strain rate sensitivity m can be calculated according to equation (1), and according to equation (2), Q can be derived from the flow stress data shown in Figure 4. Distribution patterns of m and Q obtained using an interpolation method reveal that all experimental alloys exhibit similar configurations of m and Q when their microstructures can be categorized into the same topological type in the two-phase region (i.e., the γ' dispersion or γ + γ' duplex). In comparison, the transformation of γ - γ' two-phase topologies always result in a major change in the distribution patterns of m and Q . Actually, for the alloys GH4742, GH4065A and GH4975, the two different types of m and Q distribution patterns can be obtained in the two-phase region depending on the actual topological relationship between the γ' precipitates and the γ matrix. Distributions of m and Q for alloy GH4065A are presented in Figure 5. For the γ' dispersion, as shown in Figure 5(a), m is relatively low, ranging from 0.18~0.29, and the value in the single-phase region I is slightly higher than that in the two-phase region J. In particular, for the γ + γ' duplex structure, a remarkably high m region K appears in the two-phase region J, as shown in Figure 5(b). In region K, m exhibits a high value ranging from 0.40 to 0.55, while in regions J_1 and J_2 , m remains close to that of γ' dispersion. Actually, the high m region K defines a domain of temperatures and strain rates within which the γ + γ' duplex structures exhibit superplasticity.

Considering that the calculating Q values this way result in a continuous range of Q values and therefore does not distinguish regimes of the discrete mechanisms, the apparent Q values are compared with the literature values for commonly accepted mechanisms to identify what mechanisms might be occurring in what range. As shown in Figure 5(c) and 5(d), the relatively low Q value from 330 to 420 $\text{kJ} \cdot \text{mol}^{-1}$ appears in single-phase region

Figure 4. Temperature and strain rate dependence of the steady state flow stress obtained during compression tests: (a) GH4586A, (b) GH4742, (c) GH4065A, (d) GH4975. The γ' dispersion and the $\gamma+\gamma'$ duplex exhibit a significant discrepancy in their flow behaviors.

U, which is approximated to that of self-diffusion. This result means that the deformation behavior in the single-phase region is similar to that of a typical solid solution. For the γ' dispersion, the Q value in the two-phase region V, given in Figure 5(c), is $500 \text{ kJ} \cdot \text{mol}^{-1}$ and higher. This result indicates that the dislocation mechanism has been significantly affected by the dispersed particles of γ' , as shown in the insets of Figure 5(a). In the two-phase region and between U and V, there exists a high Q region W, in which the value is up to more than $2000 \text{ kJ} \cdot \text{mol}^{-1}$, see Figure 5(c). For the $\gamma+\gamma'$ duplex structures, the distribution patterns of Q significantly differ from that of the γ' dispersion. As shown in Figure 5(d), three distinct regions X, Y and Z appear instead of region W, while region V in low temperature range still exists. In particular, the Q value ranges from 40 to $230 \text{ kJ} \cdot \text{mol}^{-1}$ in region Y, which is close to that of boundary diffusion and are much lower than that of bulk diffusion. This result means that some type of boundary reaction is indeed the mechanism that dominates the flow behavior in region Y, which is consistent with the result of microscopic investigations (see Figure 3). As the rate controlling mechanism, boundary diffusion provides a much higher migration velocity for alloying elements and can therefore

efficiently accommodate plastic deformation at a higher strain rate. As shown in Figure 5(d), region Y extends to high strain rate region in a certain temperature range, which suggests that the $\gamma+\gamma'$ duplex structures possess the ability to undergo superplastic deformation at an extraordinarily high strain rate up to 0.1 s^{-1} . Comparing Figure 5(b) and 5(d), m can be found to reach the peak value exactly at the transition area between regions Y and Z, this phenomenon is analyzed in detail later. The differences in the distribution patterns of m and Q indicate that the $\gamma+\gamma'$ duplex significantly differs from the γ' dispersion not only in their microstructural characteristics and flow stress level, but also in their controlling mechanisms of plastic deformation.

There are many mechanical models that have been well developed to predict the deformation behavior of the two-phase material based on the properties of the individual phases [17, 18]. The two particular ones are based on the strain rate or alternatively the flow stress being uniform during deformation, which are schematically illustrated in Figure 6(a) and 6(b), respectively. In particular, the uniform strain rate model, as seen in Figure 6(a), predicts the upper limit of the flow stress, which

can sometimes be reached by that of the γ' dispersion, while the uniform stress model predicts the lower limit of the flow stress, which is close to that of the $\gamma+\gamma'$ duplex, as shown in Figure 6(b).

Consider the two-phase structures of γ' precipitates (volume fraction f) and the polycrystalline γ matrix (volume fraction $1-f$). When we assume that $\dot{\epsilon} = \dot{\epsilon}_{\gamma} = \dot{\epsilon}_{\gamma'}$, where $\dot{\epsilon}$ is the prescribed overall strain rate as illustrated in Figure 6(a), the overall flow stress σ is defined as the volume average of the local stresses, and can be expressed as [18]:

$$\sigma = f\sigma_{\gamma'} + (1-f)\sigma_{\gamma} \quad (3)$$

the strain rate sensitivity m of the two-phase structure is then given by [17]:

$$m = \frac{f\sigma_{\gamma'}m_{\gamma'} + (1-f)\sigma_{\gamma}m_{\gamma}}{f\sigma_{\gamma'} + (1-f)\sigma_{\gamma}} \quad (4)$$

Therefore, the value of m can be regarded as the average of the individual strain rate sensitivities $m_{\gamma'}$ and m_{γ} , which are weighted by the volume fraction of each phase. The overall apparent activation energy Q can be derived from equations (2 ~ 4), and expressed as the sum of two terms [17]:

$$Q = \frac{f\sigma_{\gamma'}m_{\gamma'}Q_{\gamma'} + (1-f)\sigma_{\gamma}m_{\gamma}Q_{\gamma}}{f\sigma_{\gamma'}m_{\gamma'} + (1-f)\sigma_{\gamma}m_{\gamma}} + RT^2 \frac{(\sigma_{\gamma} - \sigma_{\gamma'})}{m\sigma} \frac{df}{dT} \quad (5)$$

where the second term involves the difference between the flow stresses of the two phases at a given temperature multiplied by the first derivative of f with respect to the temperature. If we assume that $\tau = \tau_{\gamma} = \tau_{\gamma'}$, where τ is the overall flow stress, as illustrated in Figure 6(b), the overall strain rate $\dot{\epsilon}$ is the volume average of the local strain rates:

Figure 5. Strain rate sensitivity m (a, b) and apparent activation energy Q (c, d) of alloy GH4065A. The value of m and Q are single-valued function of deformation conditions (temperature and strain rate) in the supersolvus temperature range (region I or U), while in the subsolvus temperature range, the γ' dispersion (a, c) and the $\gamma+\gamma'$ duplex (b, d) exhibit completely different distribution. The insets present the as-deformed structures (optical and TEM images) corresponding to the indicated deformation conditions (1025 °C and 0.001 s⁻¹).

$$\dot{\epsilon} = f\dot{\epsilon}_{\gamma'} + (1-f)\dot{\epsilon}_{\gamma} \quad (6)$$

here the strain rate sensitivity m of the aggregate is determined in the form:

$$m = \frac{f\dot{\epsilon}_{\gamma'} + (1-f)\dot{\epsilon}_{\gamma}}{f\dot{\epsilon}_{\gamma'}m_{\gamma'} + (1-f)\dot{\epsilon}_{\gamma}m_{\gamma}} \quad (7)$$

The overall apparent activation energy Q is again the sum of two terms [17, 18]:

$$Q = \frac{f\dot{\epsilon}_{\gamma'}Q_{\gamma'} + (1-f)\dot{\epsilon}_{\gamma}Q_{\gamma}}{f\dot{\epsilon}_{\gamma'} + (1-f)\dot{\epsilon}_{\gamma}} + RT^2 \frac{(\dot{\epsilon}_{\gamma'} - \dot{\epsilon}_{\gamma})}{\dot{\epsilon}} \frac{df}{dT} \quad (8)$$

where the second term involves the first derivative of the volume fraction f with respect to the temperature and the difference between the strain rates of the two phases. In equation (5) and (8), the sign of the additional term then depends on the product of the temperature dependence of the volume fraction f and the difference between the flow stresses (or alternatively, the strain rates) of the two phases at a given temperature within the subsolvus temperature range. Because f is the volume fraction of the low temperature phase γ' , it decreases with increasing temperature in the two-phase region (i.e., $df/dT < 0$ is always satisfied). When the flow stress of the high temperature phase γ is lower than that of the low temperature phase γ' over the entire two-phase region, the uniform strain rate model directly leads to $\sigma_{\gamma} < \sigma_{\gamma'}$, while $\dot{\epsilon}_{\gamma'} < \dot{\epsilon}_{\gamma}$ is valid for the uniform stress model. As a result, the second right-hand side term in equations (5) and (8) is always positive (an exceptional case of equation (8) is discussed later), and therefore, the overall apparent activation energy Q is higher than the weighted average of $Q_{\gamma'}$ and Q_{γ} and can even be higher than either $Q_{\gamma'}$ or Q_{γ} . Although there is not, in fact, a single-phase region of the γ' phase at lower temperature region in nickel-base superalloys, the virtual flow behavior of the single γ' phase can still be obtained by extrapolation from the experimental data according to equations (3 ~ 8). Specifically, the uniform strain rate model predicts a rapid increase in Q with the increase in temperature up to more than $2000 \text{ kJ} \cdot \text{mol}^{-1}$ adjacent to the γ - γ' transition temperature (γ' solvus) of all experimental alloys. This result is in accordance with that in Figure 5(c), which is derived from the experimental data. Therefore, the calculations confirm that the flow behavior of the γ' dispersion can be well described by the uniform strain rate model in most cases. In comparison, the uniform stress model only predicts a moderate influence of temperature on Q at the γ' solvus, which agrees with the experimental data for the $\gamma+\gamma'$ duplex structures near the γ' solvus, such as region X in Figure 5(d).

The presence of region Y in Figure 5(d) can also be predicted by the uniform stress model when plastic strain is accommodated by the migration of high-angle interfaces (particularly the γ' - γ phase boundaries), as shown in Figure 3 and insets of Figure 5(d). Consider a microscopic local volume dV that a phase interface has just swept across. Apparently, $dV = dV_{\gamma'} = dV_{\gamma}$ is satisfied, which leads directly to $f\dot{\epsilon}_{\gamma'} = (1-f)\dot{\epsilon}_{\gamma}$, where $\dot{\epsilon}_{\gamma'}$ and $\dot{\epsilon}_{\gamma}$ are equivalent strain rate of the γ' and γ phases, respectively. Given

that the volume fraction of the γ' phase is far less than that of the γ matrix ($f < 0.5$) for all experimental alloys in the experimental temperature range (see Table II), it is probable that the assumption $\dot{\epsilon}_{\gamma'} < \dot{\epsilon}_{\gamma}$ is no longer satisfied while $\sigma_{\gamma} < \sigma_{\gamma'}$ is still valid [18, 19]. Only under such circumstance can equation (8) predict an overall Q value lower than either $Q_{\gamma'}$ and Q_{γ} because the sign of second term on the right side is now negative, this interpretation is independent of the microstructural characterization.

The analysis based on the mechanical models exactly confirms that the γ - γ' boundary reaction indeed plays a significant role in plastic deformation in region Y. Actually, the models illustrated in Figure 6(a) and 6(b) can be generalized to any case in which the two different deformation mechanisms are involved at the same time, i.e., any region that deformed based on the same mechanism can be assumed as one phase. Therefore, the material can be defined as a two-phase aggregate as an alternative way of assuming that the deformation mechanism of one phase is a dislocation reaction and the other phase is a boundary reaction. Apparently, there exists a transition of mechanisms between region Y and V in Figure 5(d), and then, the mechanical model immediately predicts the presence of region Z, which is similar to the region W between U and V in Figure 5(c). This result also means that both dislocation and boundary reactions play a role in the transition region from Y to Z, which is confirmed by the microstructural characterization. Notably, m of the $\gamma+\gamma'$ duplex always reach its peak value in this transition region.

Figure 6. Schematic of mechanical models for two-phase structures: the uniform strain rate model (a) and uniform stress model (b) are topologically equivalent to the flow behavior of the γ' dispersion and $\gamma+\gamma'$ duplex, respectively.

In comparison, alloy GH4975 exhibits the highest volume fraction of γ' precipitates among all experimental alloys. At 1050°C, there still exists a high γ' volume fraction of 37% in the as-deformed structures, as shown in Figure 7(a) and 7(b). The distribution patterns of m and Q of alloy GH4975 are shown in Figure 7(c ~ f). A high m region K in Figure 7(e) exhibits a value up to more than 0.6, which is the highest among all experimental alloys. The distribution of the Q value in Figure 7(f) is similar to that in Figure 5(d), while the only exception is that the regions Y and Z exhibit an inverse sequence in temperature, compared with the configuration in Figure 5(d). According to the uniform stress model, this inversion between regions Y and Z indicates that the flow stress of γ grains is higher than that of the γ' precipitates ($\sigma_\gamma > \sigma_{\gamma'}$). Comparing all experimental alloys, the γ' volume fraction can be identified as the primary factor that dominates the microstructural characteristics and deformation behavior. The $\gamma+\gamma'$ duplex aggregate is a thermodynamically stable structure, and the stability of the $\gamma+\gamma'$ duplex structures increases with the increasing γ' volume fraction. During multi-cycle TMP in the two-phase region, the γ' dispersion tends to transform spontaneously into a $\gamma+\gamma'$ duplex. Actually, the fine-grained microstructures of highly alloyed disc alloys obtained using TMP are almost always a $\gamma+\gamma'$ duplex [11, 12]. Furthermore, with the increasing γ' volume fraction, region K expands correspondingly, and the maximum m value increases. In particular, in a certain subsolvus temperature range, the high m region K extends to the high strain rate region up to the level of 0.1 s^{-1} , which means that the $\gamma+\gamma'$ duplex

structures acquire the ability to superplastically deform at high strain rate levels. This behavior of the $\gamma+\gamma'$ duplex structures means that for those highly alloyed superalloys, the industrial scale hot working processes, such as billet conversion and grain refinement, can be achieved on an open die forging press while the isothermal conditions are no longer indispensable.

Conclusions

The γ' dispersion and the $\gamma+\gamma'$ duplex are the two structural categories that usually appear in nickel-base superalloys during TMP. The two structures are intrinsically different from each other in their $\gamma-\gamma'$ two-phase topological relationship, and the stability of $\gamma+\gamma'$ duplex structures increases with the increasing concentration of γ' -forming elements. During multi-cycle TMP in the two-phase region, the γ' dispersion tends to spontaneously transform into a $\gamma+\gamma'$ duplex. Under most circumstances, the fine-grained microstructures of highly alloyed disc alloys obtained using TMP are actually the $\gamma+\gamma'$ duplex structures.

Instead of the respective morphologies and size distributions of the two phases, the topological relationship between the γ' precipitates and γ grains is instead the primary factor that dominates the deformation behavior in the two-phase region. The formation of a $\gamma+\gamma'$ duplex almost always results in a decrease in the flow stress and a promotion of plasticity. In particular, $\gamma+\gamma'$ duplex structures exhibit the ability to maintain superplasticity at

Figure 7. (a, b): The as-deformed γ' dispersion (a) and $\gamma+\gamma'$ duplex (b) of alloy GH4975 (at 1050 °C and 0.001 s^{-1}). (c ~ f): The strain rate sensitivity m and apparent activation energy Q of alloy GH4975 in the two-phase region, (c, d) the γ' dispersion, (e, f) the $\gamma+\gamma'$ duplex.

a high strain rate up to 0.1 s^{-1} . The distribution of the strain rate sensitivity m and the activation energy Q in relationship with temperature and strain rate accurately identifies a specific domain within which the $\gamma+\gamma'$ duplex exhibits superplasticity.

Microstructural characterization and mechanical interpretation independently confirm that the inherent characteristics of the $\gamma+\gamma'$ duplex, including flow softening and plasticity promotion, particularly the high-strain-rate superplasticity, mainly originate from a strain-induced, high-velocity $\gamma-\gamma'$ interface reaction mechanism (i.e., a strain-induced discontinuous precipitation process), which is essentially identical to that of a cellular reaction or a eutectoid transformation.

The existence of the $\gamma+\gamma'$ duplex structure opens a window for the processing of highly alloyed nickel-base superalloys. By producing a fine-grained $\gamma+\gamma'$ duplex using TMP, the hot working processes of the disc alloys with up to more than 60% of γ' volume fraction, including billet conversion and microstructure customization, can be well accomplished in a cost-effective manner on an industrial scale.

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References

1. R. C. Reed, *The Superalloys: Fundamentals and Applications* (Cambridge: Cambridge University Press, 2006), 217-236.
2. J. C. Williams, and E. A. Starke, "Progress in Structural Materials for Aerospace System," *Acta Materialia*, 51(2003), 5775-5799.
3. R. F. Decker, "The Evolution of Wrought Age-hardenable Superalloys," *JOM*, 58(2006), 32-36.
4. D. J. Crudden, A. Mottura, N. Warnken, B. Raeisinha, and R. C. Reed, "Modelling of the Influence of Alloy Composition on Flow Stress in High-Strength Nickel-based Superalloys," *Acta Materialia*, 75(2014), 356-370.
5. E. I. Galindo-Nava, L. D. Connor, and C. M. F. Rae, "On the Prediction of the Yield Stress of Unimodal and Multimodal γ' Nickel-base Superalloys," *Acta Materialia*, 98(2015), 377-390.
6. H. Monajati, M. Jahazi, S. Yue, and A. K. Taheri, "Deformation Characteristics of Isothermally Forged Udimet 720 Nickel Base Superalloy," *Metallurgical and Materials Transactions A*, 36A(2005), 895-905.
7. J. L. W. Carter, M. W. Kuper, M. D. Uchic, and M. J. Mills, "Characterization of Localized Deformation near Grain Boundaries of Superalloy René-104 at Elevated Temperature," *Materials Science & Engineering A*, 605(2014), 127-136.
8. W. Gostic, "Application of Materials and Process Modeling to the Design, Development and Sustainment of Advanced Turbine Engines," *Superalloys 2012: 12th International Symposium on Superalloys*, ed. E. S. Huron, R. C. Reed, M. C. Hardy, M. J. Mills, R. E. Montero, P. D. Portella, J. Telesman, (Hoboken, NJ: John Wiley & Sons, Inc.), 3-12.
9. M. C. Hardy, B. Zirbel, G. Shen, and R. Shankar, "Developing Damage Tolerance and Creep Resistance in a High Strength Nickel Alloy for Disc Applications," *Superalloys 2004: 10th International Symposium on Superalloys*, ed. K. A. Green, T. M. Pollock, H. Harada, T. E. Howson, R. C. Reed, J. J. Schirra, S. Walston, (Warrendale, PA: The Minerals, Metals & Materials Society), 83-90.
10. D. U. Furrer, and H. Fecht, " γ' Formation in Superalloy U720Li," *Scripta Materialia*, 40(1999), 1215-1220.
11. A. K. Koul, and J. A. Immarrigeon, "Modelling of Plastic Flow in Coarse Grained Nickel-base Superalloy Compacts under Isothermal Forging Conditions," *Acta Materialia*, 35(1987), 1791-1805.
12. V. A. Valitov, F. Z. Utyashev, and S. K. Mukhtarov, "Formation of Microcrystalline Structures and Superplastic Properties of Nickel Based Alloys," *Materials Science Forum*, 304-306(1999), 79-84.
13. W. J. Tu, and T. M. Pollock, "Deformation and Strain Storage Mechanisms during High-Temperature Compression of a Powder Metallurgy Nickel-base Superalloy," *Metallurgical and Materials Transactions A*, 41A(2010), 2002-2009.
14. N. Bozzolo, N. Souaï, and R. E. Logé, "Evolution of Microstructure and Twin Density during Thermomechanical Processing in $\gamma-\gamma'$ Nickel-based Superalloy," *Acta Materialia*, 60(2012), 5056-5066.
15. B. Zhang, G. Zhao, W. Zhang, S. Huang, and S. Chen, "Investigation of High Performance Disc Alloy GH4065 and Associated Advanced Processing Techniques," *Acta Metallurgica Sinica*, 51(2015), 1227-1234.
16. A. Porter, and B. Ralph, "The Recrystallization of Nickel-base Superalloys," *Journal of Materials Science*, 16(1981), 707-713.
17. L. Briottet, J. J. Jonas, and F. Montheillet, "A Mechanical Interpretation of the Activation Energy of High Temperature Deformation in Two Phase Materials," *Acta Materialia*, 44(1996), 1665-1672.
18. S. Ankem, H. Margolin, C. A. Greene, B. W. Neuberger, and P. G. Oberson, "Mechanical Properties of Alloys Consisting of Two Ductile Phases," *Progress in Materials Science*, 51(2006), 632-709.
19. C. C. Tasan, M. Diehl, D. Yan, C. Zambaldi, P. Shanthraj, F. Roters, and D. Raabe, "Integrated Experimental-simulation Analysis of Stress and Strain Partitioning in Multiphase Alloys," *Acta Materialia*, 81(2014), 386-400.