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Microstructure and Mechanical Behavior of Mechanically Alloyed ODS Ni-Base Superalloy for Aerospace Gas Turbine Application

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Abstract. The gamma prime precipitation strengthening behavior and oxide dispersion strengthening behavior of mechanically alloyed oxide dispersion strengthened (ODS) Ni-base superalloys have been investigated. The most important microstructural feature affecting the elevated temperature strength of ODS alloys was found to be the grain aspect ratio. Grain aspect ratio after zone annealing was sensitively related to the primary grain size in as-extruded Ni-base superalloy. There was a suitable range of primary grain size to obtain a coarse elongated grain structure after zone annealing. The large grain aspect ratio above a critical value of about 20 resulted in an increase in stress-rupture life more than two orders of magnitude at 950°C. The size and distribution of the gamma prime precipitates were dependent on the solution heat treatment conditions. The microstructural parameters on the gamma prime precipitates significantly affected on the stress-rupture property of ODS Ni-base superalloy at the intermediate temperature range.

Keywords: gamma prime precipitates, grain aspect ratio, mechanical alloying, oxide dispersion strengthening, stress-rupture property

Introduction

It has long been recognized by aircraft engine manufacturers that the increase of turbine inlet temperature offers attractive advantages in an aircraft turbine performance and in an economy. A significant amount of improvement in turbine inlet temperature has been made possible by the advances in material capability and processing. The mechanical alloying process of Ni-base superalloys has been recognized as a novel manufacturing process for the high temperature engine components in aircraft gas turbine engines. Mechanical alloying process [1, 2] was developed around 1966 at Inco's Paul D. Merica Research Laboratory as a part of program to produce an Ni-base superalloy combining the advantage of γ' precipitation hardening for intermediate temperature ranged 700–900°C and oxide dispersion strengthening for high temperature above ≥1000°C for gas turbine engine application.

Mechanical alloying is a dry, high-energy ball milling operation that produces composite metal powders with extremely fine microstructures. Mechanically alloyed powders are consolidated by placing them in sealed cans for extrusion or hot pressing, followed by conventional hot an cold working processes. A final annealing at very high temperature is required to develop a stable, coarse grain microstructure suitable for the most demanding stress-rupture applications. The highly elongated grains are developed through secondary recrystallization by zone annealing of as-extruded, fine grained alloys. Oxide dispersion strengthened (ODS) alloys produced by MA process attract great attention as advanced high temperature materials, because they can retain useful strength up to a relatively high temperature. The elevated temperature strength of the ODS alloy is increased due to the direct strengthening of fine, uniformly dispersed and stable oxide particles by acting as barriers to dislocation motion.

In addition to this direct strengthening by oxide particles, the most important microstructural features affecting elevated temperature strength of the ODS alloy is the grain aspect ratio (GAR). The accumulation of creep damage on transverse boundaries of the ODS alloy can be controlled by an accommodation process, i.e., mutual sliding displacements between neighboring grains in the longitudinal direction. An elongated grain shape contributes to the retardation of this process and prolongation of creep life. The formation of coarse elongated grains with a high grain aspect ratio is possible during secondary recrystallization by a process of directional recrystallization or zone annealing heat treatment [3–8]. Although the exact mechanism of secondary recrystallization behavior is not fully understood up to now, the importance of GAR has been received considerable attention [9–11]. At the intermediate temperature region of 700–900°C, the strengthening of MA ODS Ni-base superalloys is obtained through the precipitation hardening by a Ni_3 (Al, Ti), which is known as γ' phase [12]. The γ' precipitates are ordered FCC ($L1_2$) structure and are coherent with the nickel-rich γ matrix having FCC structure. The size, volume fraction and distribution of the γ' precipitates are sensitively related to the heat treatment for precipitation and these parameters can significantly affect the mechanical properties of the ODS Ni-base superalloys.

The aim of this study is to show that there is a suitable range of average grain size in as-extruded materials to obtain a coarse elongated grain structure after zone annealing. Since the rupture life was closely related to the microstructure at elevated temperature, the effect of solution heat treatment temperature on the γ' precipitate size and stress rupture property of MA ODS Ni-base superalloy was investigated.

Experimental procedures

Preparation of alloy

An oxide dispersion strengthened Ni-base superalloy, which is designated as Alloy 92 [13], was prepared by Inco Alloys International Inc. The elemental powders and master alloy powders were mechanically alloyed in an attritor. The mechanically alloyed powders were packed into mild steel cans, then the cans were evacuated and sealed. The sealed cans were extruded into bars with 1.8 cm in diameter at 1175°C with extrusion ratio of about 17 : 1. The nominal composition of the ODS Ni-base superalloy (Alloy 92) is listed Table 1. The

Table 1. The nominal compositions of an experimental Alloy 92 (unit: wt.%).

	Ni	Cr	Al	W	Ta	Mo	Co	Ti	Zr	B	C	Y_2O_3	Re
Alloy 92	Bal.	8	6.5	6	3	1.5	5	1	0.15	0.01	0.05	1.1	3

transmission electron microscope was observed to characterize the as-extruded microstructure of ODS Ni-base superalloy. TEM specimens were electropolished in a Struers Tenupole twin jet electropolisher operating at 30 V in a 9 : 1 mixture of ethanol and perchlolic acid at −50°C.

Zone annealing for grain size control

Zone annealing treatments were conducted at hot zone temperature of 1300°C with a furnace travel speed of 5.5 cm/h. The zone annealed bars were surface ground to reveal the secondary recrystallized grain structure. In order to enhance the secondary recrystallization response, the effect of preannealing treatment before the zone annealing have been investigated by TEM and optical microscope. The preannealing was conducted at gamma prime dissolution temperature of 1135°C for 40 min. The primary grain sizes were obtained by measuring the profile diameter of about 200 grains on TEM photographs.

The specimens for stress-rupture test were machined with a gage diameter of 4.5 mm and gage length of 20 mm. The gamma prime heat treatments were performed by three steps as 1/2 h/1280°C/AC + 2 h/950°C/AC + 24 h/850°C/AC. The stress-rupture tests were conducted at 950°C under constant load in air with the tensile axis parallel to the extrusion direction.

Heat treatment for gamma prime precipitation

The extruded bars were zone annealed at hot zone temperature of 1300°C with a furnace travel speed of 9 cm/h and followed by three-stage heat treatment for the precipitation of γ' phase. The solvus temperature of γ' is extended to 1070–1170°C and the solidus line is reported as 1330–1390°C in the Alloy 92 [14]. The superalloy was solution treated at three different conditions. The superalloy was solution treated at two different temperatures of 1232°C and 1280°C for fixed time of 30 min. While, the solution treatment time was varied from 30 min to 2 h at a temperature of 1280°C, then followed by two stages aging treatments (950°C/2 h/air cooling (AC) + 850°C/24 h/AC). The detail processes are as follows:

(1) HTC1: 1232°C/0.5 h/air cooling (AC) + 950°C/2 h/AC + 850°C/24 h/AC
(2) HTC2: 1280°C/0.5 h/AC + 950°C/2 h/AC + 850°C/24 h/AC
(3) HTC3: 1280°C/2 h/AC + 950°C/2 h/AC + 850°C/24 h/AC

Stress-rupture tests

The specimens for stress-rupture test were machined with a gage diameter of 4.5 mm and gage lengths of 20 mm after the three stages heat treatments. The stress-rupture tests were conducted at 760°C with the tensile axis parallel to the extrusion direction. The γ' precipitates were observed by the scanning electron microscope (SEM) and transmission electron microscope (TEM). The specimens for observation of the microstructure by SEM were electropolished in a mixture of 17 ml of H_2O, 2 ml HNO_3 and 1 ml of CH_3COOH.

Results and discussion

Microstructure and grain aspect ratio

It is well known that GAR values in secondary recrystallized material can be changed with sample geometry and zone annealing conditions. In this study the relationship between GAR after zone annealing and the primary grain size before zone annealing was investigated. Typical TEM micrographs of the as-extruded sample were given in figure 1. The microstructure was characterized as fine equiaxed grains. The average grain size was measured approximately ranged 0.3–0.6 μm. To improve the high temperature mechanical properties, the as-extruded, fine grained microstructure needs to be transformed into a coarse grain structure. This is possible with zone annealing during which secondary recrystallization occurs. The optical micrographs after zone annealing treatment were given in figure 2. It is well known that the high temperature strength is closely related to the shape of the grains, and the creep strength increases linearly with increasing grain aspect ratio (GAR), which is the ratio of grain length, L, to grain width, W. The GAR should be large, preferably greater than 20, to increase high temperature creep-rupture resistance.

For the same GAR, the coarser elongated grains are more desirable than the finer elongated grains since there are less transverse grain boundary areas initiating creep cracks. This means that the fine-grained structure of figure 2(b) need to be changed into the coarse grained structure of figure 2(a) to increase the high temperature capability. The driving force for primary recrystallization is the reduction of dislocation line energy, whereas the driving force for secondary recrystallization is the reduction of grain boundary energy. Therefore, it is assumed that the difference in secondary recrystallization behavior of this sample results from the difference in the primary gain size of as-extruded materials. Transmission electron micrographs showed that there was a difference in the average grain size, which was 0.50 μm and 0.35 μm in the as-extruded condition, respectively. The average grain size can be changed by the normal grain growth prior to zone annealing. This is called preannealing because it is conducted before the zone annealing treatment. The upper temperature of preannealing is limited below the critical secondary recrystallization temperature, and the

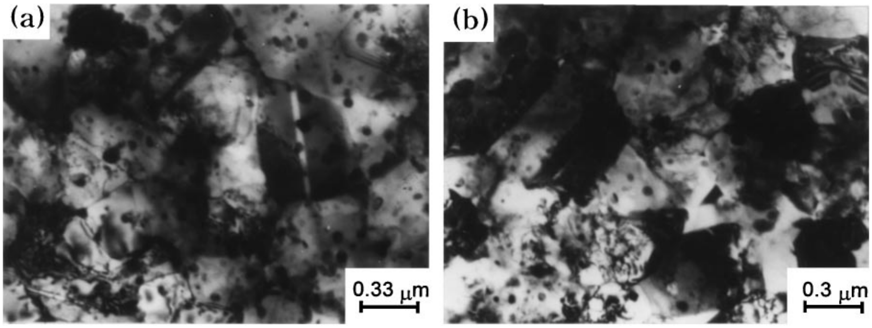

Figure 1. Bright field electron micrographs of Alloy 92 in the extruded condition: (a) Alloy 92A (0.5 μm), (b) Alloy 92B (0.35 μm).

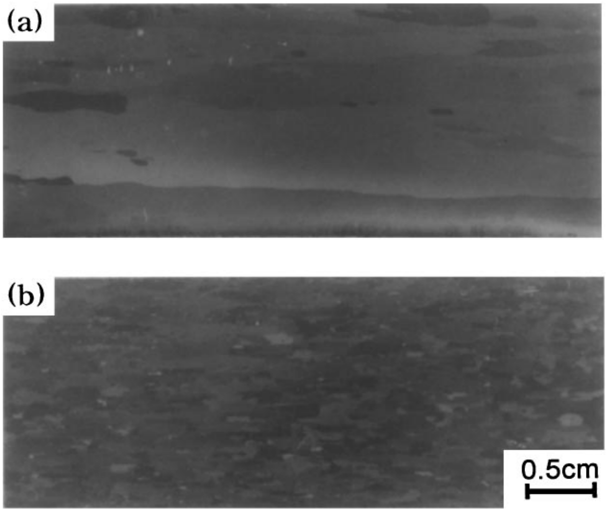

Figure 2. Optical micrographs of zone annealed sample with maximum hot zone temperature of 1290°C, furnace travel speed of 5.5 cm/h, (a) Alloy 92A (0.5 μm), (b) Alloy 92B (0.35 μm).

lower temperature is bound to the gamma prime solvus temperature because the grain boundary mobility is severely restricted.

From the above consideration, the preannealing temperature was determined 1135°C within the temperature range for gamma prime dissolution. The gamma prime dissolution temperatures were ranged from 1050°C to 1170°C as reported in the literature [10]. TEM investigations as shown in figures 3(a)–(c) had shown that there was a normal grain growth

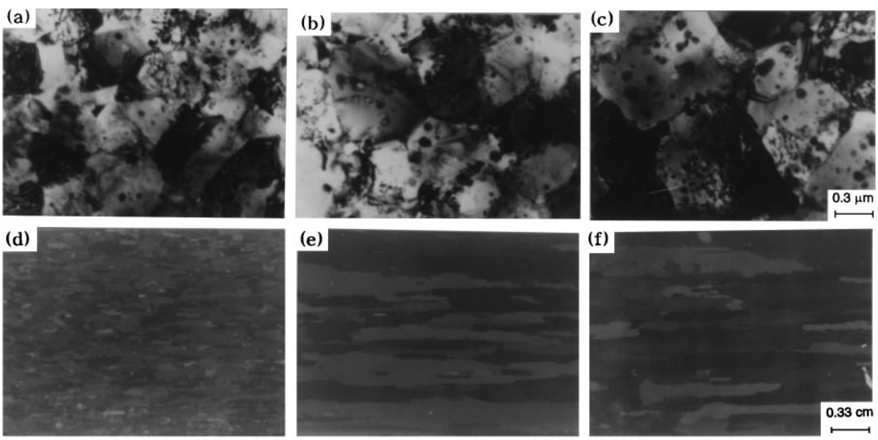

Figure 3. TEM micrographs of Alloy 92 show the difference of primary grain size: (a) $D = 0.35$ μm, (b) $D = 0.47$ μm, (c) $D = 0.61$ μm, and (d), (e), (f) are optical micrographs of these specimens after zone annealing treatment at 1290°C, 5.5 cm/h respectively.

during preannealing. Average grain sizes of Alloy 92B were changed from 0.35 μm in as-extruded materials to 0.47 μm or 0.61 μm after preannealing for 40 or 240 min, respectively. The optical micrographs of zone annealed Alloy 92 showed the enhanced grain structure after preannealing heat treatment as shown in figures 3(e) and (f). The coarsest elongated grain structure was obtained when preannealing time was 40 min as shown in figure 3(e). In the case of preannealing for 240 min, the GAR value was lower than that of 40 min annealed sample. These results confirmed that the difference in secondary recrystallization behavior of ODS alloy was resulted from the difference in the primary grain size. These results showed that the preannealing heat treatment prior to zone annealing could increase the grain aspect ratio after zone annealing. It is possible to establish the direct relationship between GAR after zone annealing and the primary grain size of as-extruded materials. From the above microstructural consideration, it is considered that there was a suitable range of primary grain size for producing coarse elongated grain structure after zone annealing as summarized in figure 4.

It has been found that the optimum primary grain size for producing a coarse elongated structure is ranged 0.45–0.55 μm. When the primary grain size was less than 0.45 μm, a coarse elongated structure was not developed during the secondary recrystallization. However, the preannealed fine grained alloy produced the desirable coarse elongated structure after zone annealing because the preannealing heat treatment was able to produced a suitable primary grain size for a coarse elongated grain structure. Furthermore, the application of preannealing heat treatment can induce more homogeneous deformation across the transverse section of the as-extruded bars.

The results on the preannealing heat treatment indicate that the desirable coarse elongated structure could be obtained from a very fine grained ODS superalloy. These confirmed that

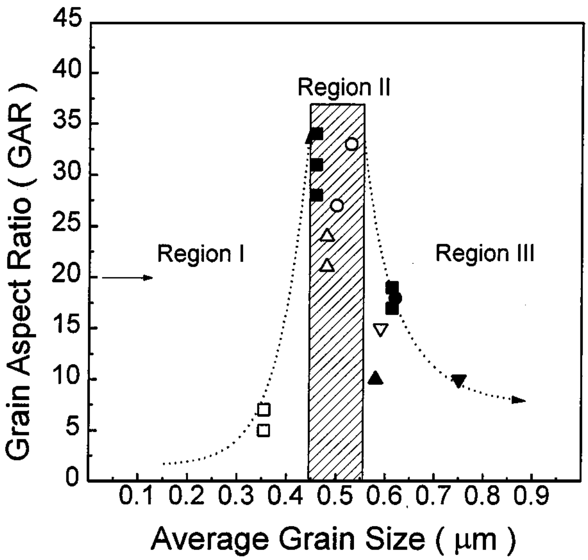

Figure 4. Variation of grain aspect ratio after zone annealing as a function of primary grain size (open symbol: as-extruded and closed symbol: preannealed stated).

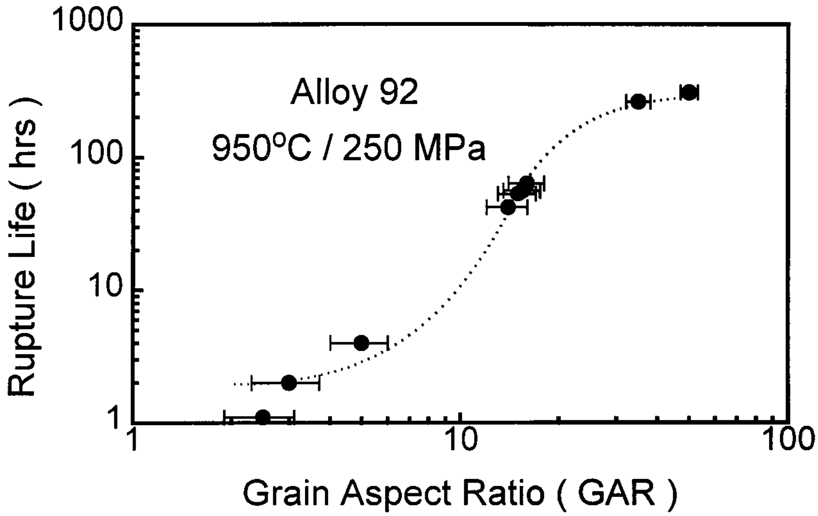

Figure 5. Stress-rupture life as function of grain aspect ratio (GAR) for Alloy 92B at 950°C, 250 MPa.

the grain aspect ratio after zone annealing heat treatment was closely related with the primary average grain size of as-extruded alloy. According to Jongenburger Singer [11], the growth rate was dependent on the primary grain size ($G \propto 1/L$), and the nucleation rate was inversely dependent on the third power of primary grain size ($Nv \propto 1/L^3$). This means that the nucleation rate is more sensitively changed with primary grain size than that of the growth rate. So, if the primary grain size is controlled appropriately, the nucleation rate could reduced more than the growth rate. Figure 5 shows the variation of rupture life as a function of GAR in the longitudinal direction. As the GAR increasing from about 5 to 45, rupture lives increased from about 3 h to about 300 h. The increase in rupture life is resulted from the change in fracture mode as shown in figure 6. At low GAR, intergranular fracture by the pull-out of individual grains was predominant, whereas the transgranular

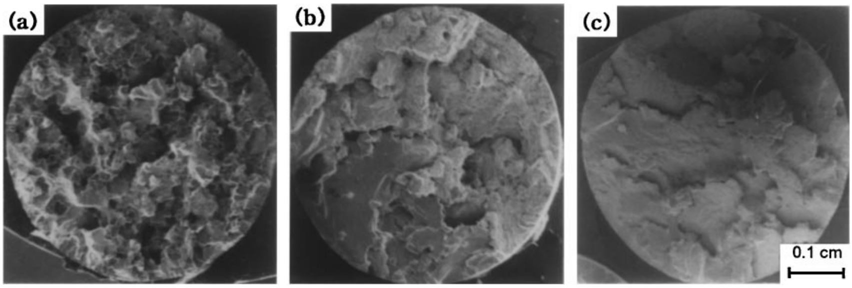

Figure 6. SEM micrographs showing the fracture surface of specimen with a various GAR: (a) GAR $\approx$ 2, (b) GAR $\approx$ 15, (c) GAR $\approx$ 45, fracture morphology changed from intergranular (a), to transgranular (c) as a GAR value increasing.

fracture was predominant at high GAR. These results showed that the high temperature stress-rupture properties were sensitively dependent on the grain aspect ratio after zone annealing treatment, and the GAR is closely related to primary grain size of as-extruded materials.

Gamma prime precipitates and stress rupture property

To make the ODS Ni-base superalloy suitable for use at high temperature, the as-extruded fine-grained microstructure needs to be transformed into an elongated coarse-grained microstructure. The elongated coarse-grained microstructure can be obtained by the secondary recrystallization through gradient annealing. For most MA ODS superalloys, the recrystallization is performed under the condition of temperature gradient as shown in figure 7, which causes the formation of elongated large grains with high grain aspect ratio (GAR). The microstructure of the zone annealed specimen in figure 2 showed a coarse elongated microstructure developed during the gradient annealing treatment.

The specimens were furnace cooled to room temperature after passing the peak temperature zone passed during zone annealing as illustrated in figure 1. During the furnace cooling, the gamma prime precipitates were precipitated in gamma matrix with the compositions as analyzed in Table 2.

The large irregular γ' precipitates with an average cube size of about 1.8 μm were observed as shown in figure 8. It is reported that the stress-rupture resistance at 760°C was obtained by the fine precipitates less than 1 μm [15–17]. This indicates that solution treatment temperature needs to be high enough to dissolve the preexisting coarse gamma

Table 2. The chemical composition of matrix and gamma prime precipitates of Alloy 92 (unit: wt.%).

	Ni	Al	W	Mo	Ti	Cr	Co	Others
Matrix	66	5.8	6.0	1.8	1.0	7.8	4.9	7.1
Precipitates (γ')	70	12.2	7.6	0.9	1.6	2.3	2.8	3.0

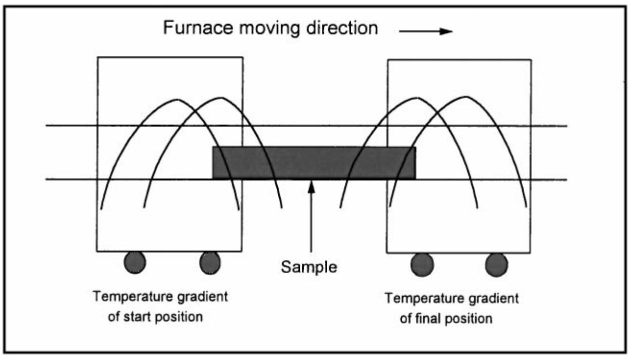

Figure 7. Schematic illustration of the zone annealing heat treatment process.

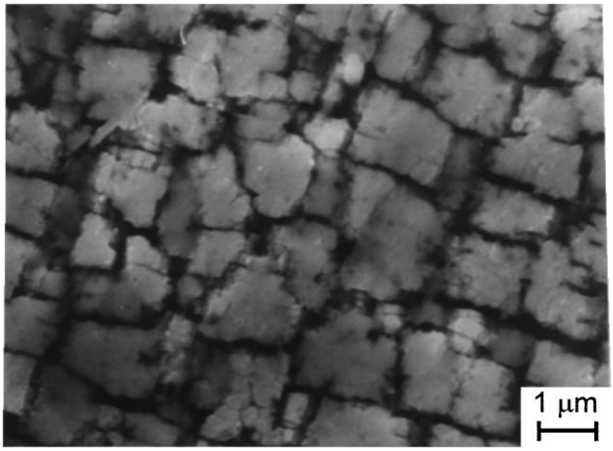

Figure 8. SEM micrograph showing coarse γ' precipitates formed during the furnace cooling after zone annealing heat treatment (1300°C, 9 cm/h).

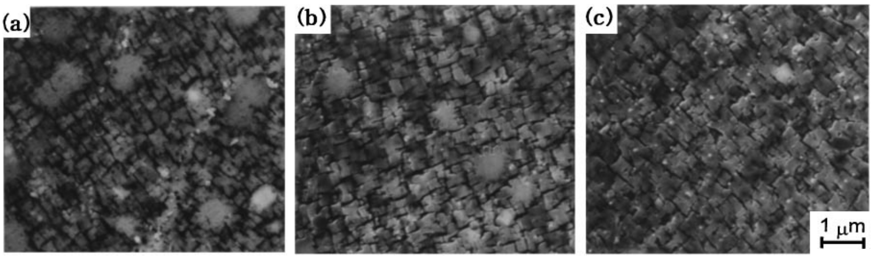

Figure 9. SEM micrographs show the variation of γ' particle size with solution heat treatment: (a) 0.5 h/1232°C [HTC1], (b) 0.5 h/1280°C [HTC2] and (c) 2 h/1280°C [HTC3].

prime so that it can be reprecipitated into a finer form during cooling. The microstructure of Alloy 92 after three different solution treatments were observed and the morphologies of gamma prime precipitates after gamma prime heat treatments were shown in figure 9. The HTC1 treatment resulted in a precipitation of cuboidal shape fine gamma prime particles of about 0.30 μm in cube size in addition to the unresolved coarse residual gamma prime of about 0.90 μm. The HTC2 treatment showed a similar morphology with HTC1 treatment, while the average size of gamma prime precipitates was about 0.41 μm and the number of unresolved coarse residual gamma prime was much reduced compared to HTC1 (figure 9(b)). The HTC3 treatment resulted in a uniform cuboidal γ' precipitates of about 0.53 μm without any unresolved coarse gamma prime precipitates (figure 9(c)). In order to compare the high temperature creep resistance, the superalloy heat treated with three different conditions were stress-rupture tested at 760°C. Figure 10 shows the variation of 100 h rupture strength of the superalloy as a function of the gamma prime precipitates size. The highest stress-rupture strength was obtained at the cuboidal γ' size of about 0.41 μm.

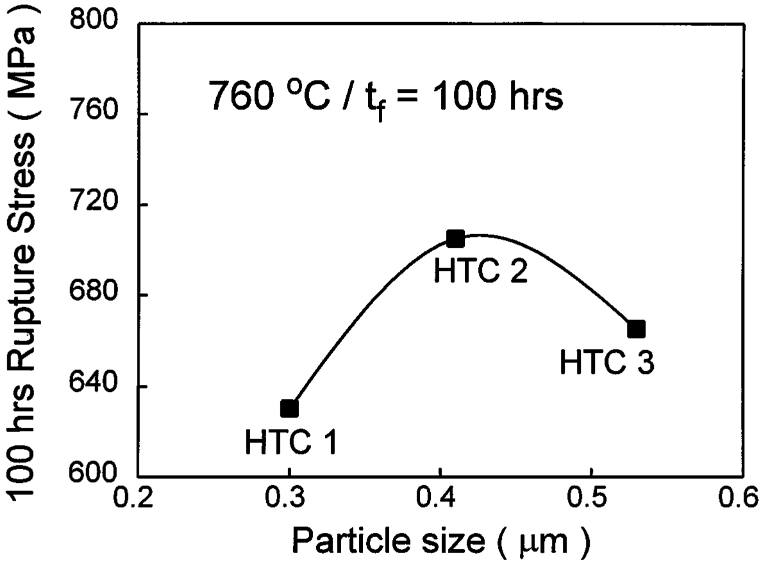

Figure 10. The variation of 100 h rupture-stress of Alloy 92 with varying gamma prime precipitate size.

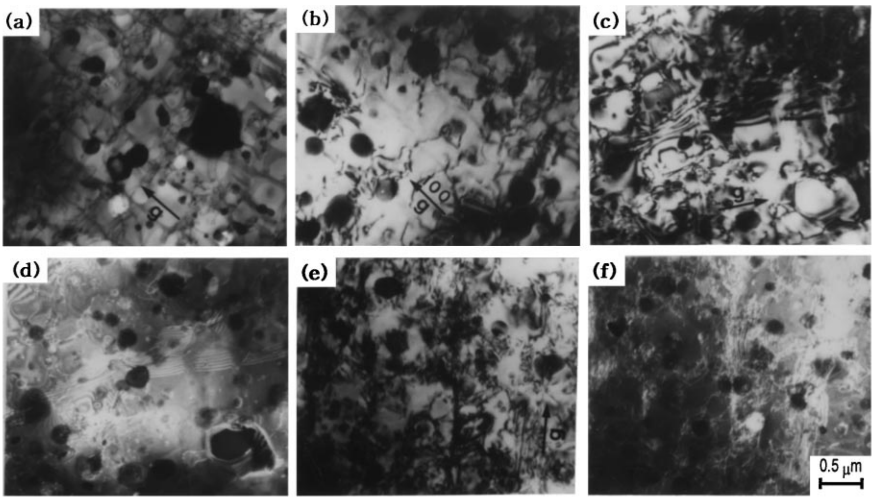

Figure 11. The transmission electron micrographs showing the dislocation structure in stress-rupture tested specimens at 760°C: (a) and (b) are bright filed image showing extensive slip through the matrix channel (HTC1), (c) is bright field image and (d) is weak beam dark field image showing the stacking faults due to γ' shearing (HTC2). (e) is bright field image and (f) is weak beam dark field image showing the dislocation tangles around the γ' precipitates (HTC3).

The typical deformation microstructures at 760°C of the Alloy 92 containing γ' precipitates with size in the range of 0.30–0.53 μm are shown in figure 11. When γ' size was about 0.41 μm, the γ' precipitates were sheared by 1/3 $\langle 112 \rangle$ type dislocations and formed the superlattice intrinsic and extrinsic stacking faults as shown in figures 11(c) and (d). The specimen having the shear deformation mode of the gamma prime showed the highest stress-rupture strength among the three different heat treatments. When the γ' size was decreased to 0.30 μm, the deformation was occurred by the extensive slip through matrix. The stress-rupture life was decreased as the γ' size decreased from 0.41 μm to 0.30 μm as shown in figure 10. As the precipitate size increased to 0.53 μm, the Orowan bowing of dislocations around the gamma prime precipitates was promoted and resulted in a decrease in stress-rupture life as shown in figure 10.

Summary

The effect of preannealing on the secondary recrystallization behavior of an ODS superalloy (Alloy 92) was investigated. Grain aspect ratio after zone annealing heat treatment was closely related to the primary grain size of the as-extruded superalloys. There was a suitable range of average grain size of as-extruded superalloys, which ranged from 0.45 μm to 0.55 μm, to obtain coarse elongated grain structure after zone annealing. When the primary grain size was less than 0.45 μm, the average grain size need to be increased to suitable range for coarse elongated grain structure by preannealing heat treatment. The high GAR above 20 could be obtained by the preannealing treatment and resulted in an increase of rupture life more than a hundred times at 950°C.

The effect of solution treatment on the γ' size and the stress-rupture property of ODS Ni-base superalloys was investigated at intermediate temperature of 760°C. The highest stress-rupture strength was obtained at a cuboidal γ' size of about 0.41 μm after solution treatment at 1280°C for 0.5 h (HTC2). The shear deformation mode of precipitate by 1/3 $\langle 112 \rangle$ type dislocations was observed when the average size of γ' was about 0.41 μm. Lower stress-rupture strength was obtained, when extensive matrix slip at 0.3 μm γ' size (HTC1) or the Orowan bowing at 0.53 μm γ' size (HTC3) were observed. These showed that the sizes of γ' precipitates were closely related to the heat treatment conditions and this parameter could significantly affect the stress-rupture property of the ODS Ni-base superalloy at intermediate temperature regime.

References

1. J.S. Benjamin, Metall. Trans. **1A**, 2943–2951 (1970).
2. J.S. Benjamin, Mechanical alloying—A perspective, in *New Materials by Mechanical Alloying Techniques*, edited by E. Arzt and L. Shultz (Duetsche Geshellschaft Metallkunde, 1988), p. 3–18.
3. Y.G. Kim and H.F. Merrick, NASA CR-15943, 1979.
4. S.K. Kang and R.C. Benn, Metall. Trans. **16A**, 1285 (1985).
5. G.H. Gessinger, Metall. Trans. **7A**, 1203 (1976).
6. R.K. Holzler and T.K. Glasglow, Metall. Trans. **13A**, 1665 (1982).
7. K. Mino, Y.G. Nakagawa, and A. Ohtomo, Metall. Trans. **18A**, (1987).

8. Y.G. Kim, L.J. Park, S.H. Hong, J.J. deBarbadillo, and J.M. Poole, Materials Science Forum **113-115**, 237–244 (1993).
9. B.A. Wilcox and A.H. Clauer, Acta Metall. **20A**, 743 (1972).
10. J.J. Stephens and W.D. Nix, Metall. Trans. **16A**, 1307 (1985).
11. C.P. Jongenburger and R.F. Singer, Recrystallization of ODS superalloy, in *New Materials by Mechanical Alloying Techniques*, edited by E. Arzt and L. Schultz (DGM, 1988), pp.157–200.
12. C.T. Sims, N.S. Stoloff, and W.C. Hagel, *Superalloy II* (John Weley & Sons, 1987), pp. 66–78.
13. R.C. Benn and P.K. Mirchandani, Dispersion strengthening by mechanical alloying, in *New Materials by Mechanical Alloying Techniques*, edited by E. Arzt and L. Schultz (DGM, 1988), pp. 19–38.
14. Development and characterization of MA ODS Ni-base turbine applications, Inco Internal Report, 1993, pp. 1–10.
15. G.E. Dieter, *Mechanical Metallurgy* (McGraw-Hill Book Company, 1986), p. 295.
16. J.L. Walter, M.R. Jackson, and C.T. Sims, Alloying, ASM, 1988, p. 125.
17. T. Khan, Recent development and potential of single crystal superalloy for advanced turbine blades, ONERA Internal Report, 1986, pp. 1–30.