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Effects of temperature and grain size on deformation and fracture in recrystallized Ni_3Al doped with boron

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The effects of temperature and grain size on the deformation and fracture behaviour of recrystallized Ni_3Al doped with boron were investigated by tensile tests at temperatures up to 973 K as a function of grain sizes from 1.6 to 105 μm. The yield stress showed a positive temperature dependence to a peak temperature in somewhat different manners depending on the grain size. For coarse-grained specimens, a rapid drop in elongation was observed with increasing temperature. The predominant fracture mode changed with temperature from the transgranular fracture of {1 1 1} cracking to brittle intergranular fracture. This embrittlement at elevated temperatures was considered to occur by a high stress concentration at grain boundaries arising from increased flow stress level and the occurrence of grain boundary sliding (GBS). In contrast, the elongation was not so markedly decreased with temperature for intermediate- and fine-grained specimens which exhibited ductile intergranular fracture and cavitation fracture, respectively, at elevated temperatures, and a slant-type fracture and cup–cone fracture, respectively, at low temperatures. The suppression of serious high-temperature embrittlement for intermediate-grained specimens was explained in terms of the slow propagation of a crack formed by GBS, owing to stress relaxation by dynamic recrystallization (DR) and plastic deformation. In the case of ultra-fine-grained specimens a large elongation was developed at elevated temperatures, which was interpreted as that the further occurrence of DR with increasing volume fraction of grain boundaries reduces the cavitation promoted by GBS, and that the limited sliding length due to extremely small grain diameter raises the stress for cavity formation.

1. Introduction

The most attractive characteristic of Ni_3Al-based intermetallic compounds is the ability to increase their strength with increasing temperature as well as their superior resistance to creep, fatigue and corrosion at elevated temperatures. From both scientific and practical interest in these anomalous and excellent high-temperature properties, extensive studies have been performed on these compounds. In particular, the development for high-temperature structural applications becomes the centre of interest since a small addition of boron to Ni_3Al leads to a remarkable improvement in room-temperature ductility [1]. Brittleness at elevated temperatures is also overcome by grain refinement using the process of powder extrusion [2] or recrystallization [3, 4]. Moreover, superplasticity in recrystallized Ni_3Al polycrystals doped with boron is found by careful control of grain size, temperature and initial strain rate [3, 4]. Superplastic deformation is of great interest because of potential cost reduction and considerable freedom in designing complex components. The recrystallization method also has the advantages of cutting down the expense and giving freedom from microporosity, antiphase domains [5–7] and other defects introduced during casting, rapid solidification or powder sintering procedures.

Using the recrystallization method, we have succeeded in controlling the grain size of a boron-doped Ni_3Al over a wide range of grain size, and in particular refining to the order of 1.6 μm minimum [8], which is the critical value indicating superplasticity. Using these recrystallized Ni_3Al polycrystals doped with boron, various mechanical properties at room temperature have recently been reported as a function of grain size. One of the most interesting results from the previous study [8] is that the fracture mode and ductility (strain to fracture) are closely related to the grain size (d); that is, {1 1 1} cracking ($d \geqq 50\,\mu$m), slant-type fracture ($50\,\mu m > d \geqq 8.5\,\mu$m) and cup–cone fracture ($d < 8.5\,\mu$m). Large elongation is developed when the intersection of slip bands becomes insufficient to produce {1 1 1} cracking, and the void formation which results in cup–cone fracture is retarded.

The main purpose of the present series of studies is to investigate the mechanical properties of a boron-doped Ni_3Al polycrystal as a function of temperature. This paper presents the temperature and grain size dependence of the yield stress, ductility and fracture behaviour of this compound.

2. Experimental procedure

Ingots of Ni_3Al (Ni–24 at % Al–0.24 at % B) were prepared by arc-melting, followed by homogenization

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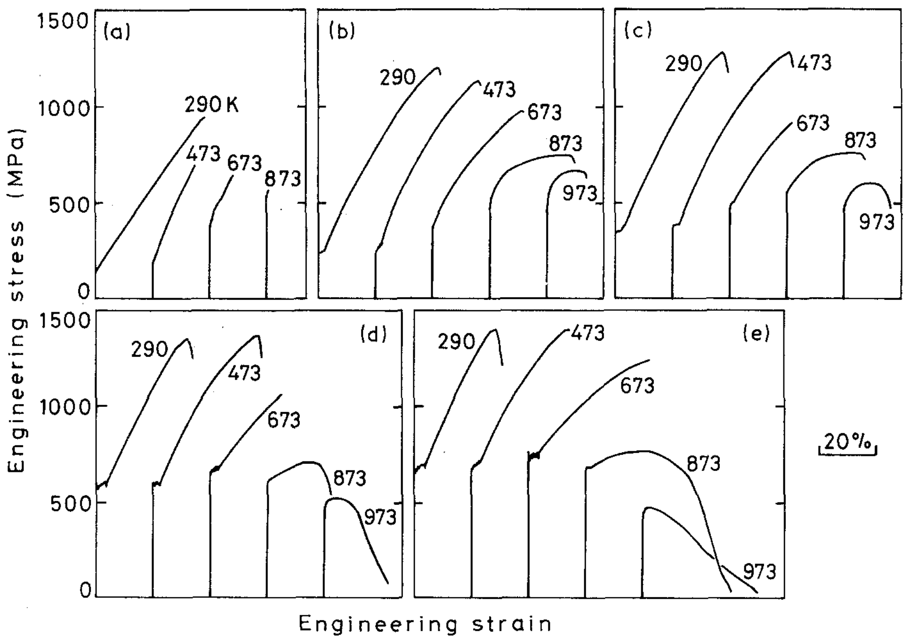

Figure 1 Temperature dependence of the engineering stress–strain curves at each grain size, *d*: (a) 105, (b) 28, (c) 8.5, (d) 2.9 and (e) 1.6 μm.

annealing as described previously [8]. The ingots were cold-rolled with appropriate intermediate anneals. Rectangular specimens with dimensions of 1 mm × 2 mm × 16 mm were taken from the cold-rolled plates using a spark cutting machine. The specimens were subjected to recrystallization annealing at various temperatures to obtain the required grain sizes of 1.6, 2.9, 8.5, 28 and 105 μm. Each grain size was confirmed by the linear intercept method using an optical microscope (OM) and a transmission electron microscope (TEM). It was also checked that equiaxed grains were uniformly distributed for all specimens by OM and TEM observations. Thin foils for TEM studies were prepared by jet electropolishing.

After damaged layers of the specimens were removed by electropolishing, tensile tests were performed using an Instron-type testing machine. Tests were carried out at an initial strain rate of $5.2 \times 10^{-4}\,\text{sec}^{-1}$ and at temperatures up to 973 K in a vacuum of 2×10^{-3} Pa. During tensile testing the samples and their grips were loosely wrapped in titanium foil to keep the surfaces clean. A scanning electron microscope (SEM) was employed to examine the fracture surface and the longitudinal section of specimens. If necessary, the internal structure of deformed specimens was observed by TEM.

3. Results

Fig. 1 shows the engineering stress–strain curves for different temperatures at each grain size (d). It can be seen that the temperature dependence of the stress–strain curve changes with the grain size. In the case of specimens having a coarse grain size of 105 μm, the strain to fracture (elongation) decreases rapidly with increasing temperature. Catastrophic fracture without necking occurs after work-hardening at the temperatures investigated here. In contrast, for intermediate ($28\,\mu\text{m} \geqq d \geqq 8.5\,\mu\text{m}$) and fine ($d < 8.5\,\mu\text{m}$) grained specimens, the serious elongation-loss at elevated temperatures is not found. The work-hardening tends to become smaller with increasing temperature. Moreover, a stress-drop which corresponds to the development of necking as well as the overall reduction in specimen cross-section is observed at the final or from the initial stage of deformation, depending on grain size and temperature.

Fig. 2 shows the temperature dependence of the elongation and yield stress (0.2% offset stress) as a function of grain size. When grains are extremely refined to the order of 1.6 μm, a large elongation is developed at elevated temperatures. In contrast, the elongation of coarse-grained specimens ($d = 105\,\mu$m) drops promptly with increasing temperature as mentioned above. For specimens whose grain size is between these two values, the elongation at elevated temperatures holds a somewhat low value compared with that at low temperatures.

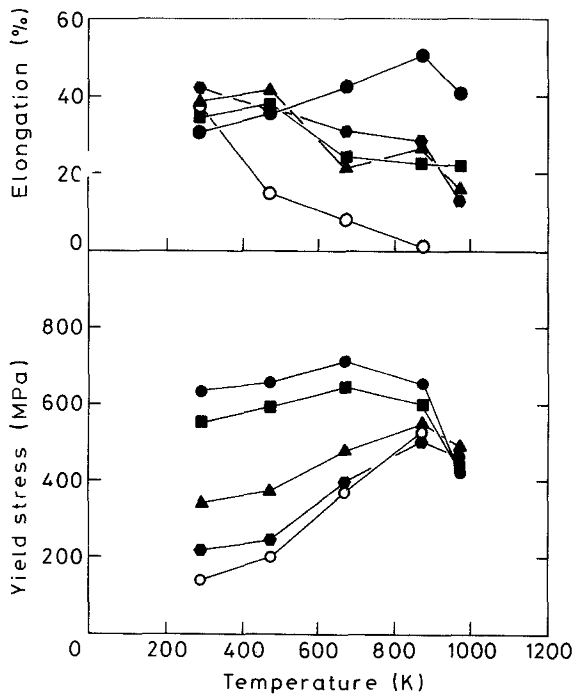

Figure 2 Temperature dependence of the elongation and the yield stress as a function of grain size, at $\dot{\varepsilon} = 5.2 \times 10^{-4}\,\text{sec}^{-1}$: (●) 1.6, (■) 2.9, (▲) 8.5, (⬢) 28 and (○) 105 μm.

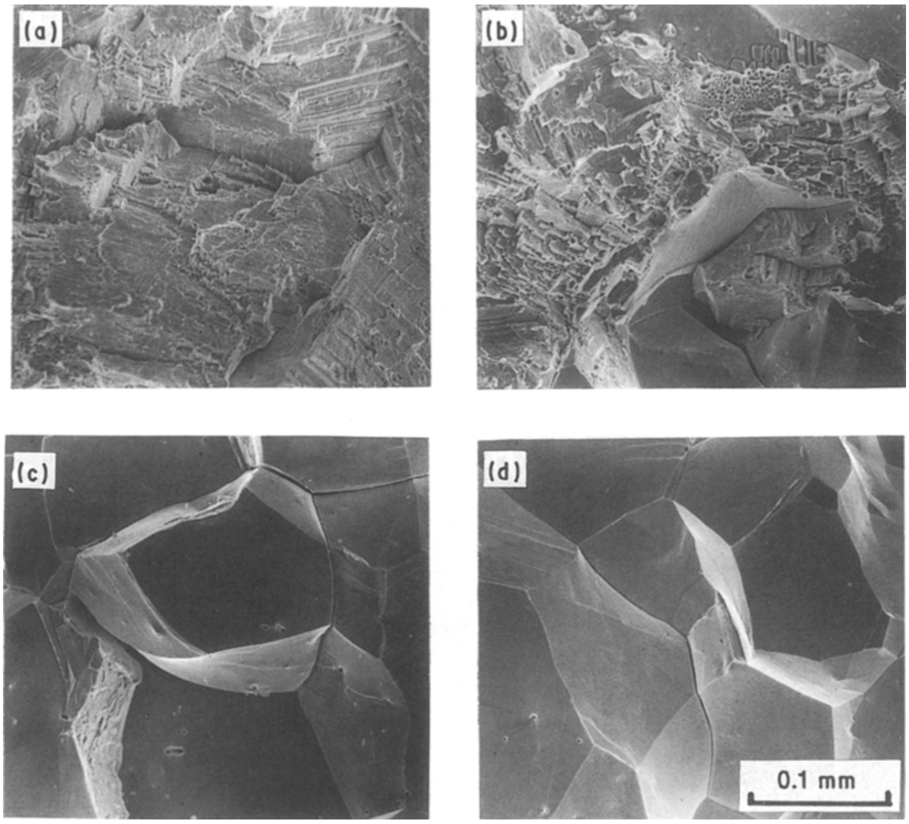

Figure 3 SEM fractographs of specimens having a grain size of 105 μm fractured at (a) 290, (b) 473, (c) 673 and (d) 873 K.

As shown in Fig. 2, the yield stress shows a positive temperature dependence to a peak temperature at each grain size. The peak in yield stress appears at about 873 K for the specimens with $d \geqq 8.5\,\mu$m, and about 673 K with $d < 8.5\,\mu$m. Oya *et al.* [9] reported that the peak temperature is at about 873 K in the grain size range of 4 to 275 μm for the same composition of compounds tested by compression. From both the present results and those of Oya *et al.*, it seems likely that a distinct decrease in peak temperature occurs when $d < 4\,\mu$m in the boron-doped Ni_3Al polycrystals. A similar grain size dependence on the peak temperature is also observed in stoichiometric Ni_3Al without boron [2, 10] and Ni–23Al–0.5Hf–0.2B (at %) polycrystals [11]. According to the results on stoichiometric Ni_3Al without boron investigated by Hanada *et al.* [10] and Weihs *et al.* [2], using the grain size ranges of 12 to 75 μm and 2.9 to 85 μm, respectively, the peak temperature appears at about 973 K when $d \geqq 12\,\mu$m, and is shifted to a low temperature when $d < 12\,\mu$m. In the case of a boron- and hafnium-doped Ni_3Al, Takeyama and Liu [11] reported that the peak temperature changes from around 1073 K for $d \geqq 85\,\mu$m to around 873 K for $d = 15.3\,\mu$m. Therefore, a decrease in peak temperature below a critical grain size which depends on the composition seems to be a general feature of Ni_3Al-based intermetallic compounds.

Figs 3a to d show SEM fractographs for coarse-grained specimens ($d = 105\,\mu$m) tested at 290, 473, 673 and 873 K, respectively. Note the change in fracture mode with increasing temperature, from transgranular fracture at 290 K to intergranular fracture at 673 K and above, through the mixture modes at 473 K. As shown in Figs 3a and b, a transgranularly fractured surface consists predominantly of flat, smooth facets formed with a stepped, blocky structure, which is characteristic of the familiar {1 1 1} slip plane fracture. {1 1 1} cracking is also observed in single-crystalline Ni_3(Al, Ti) with [12] and without boron [13–15], and polycrystalline Ni_3(Al, Ti) [16] and Ni_3Al with [8, 16] and without boron [16].

Fig. 4 shows SEM micrographs of the longitudinal section and fracture surface for specimens having a grain size of 28 μm, fractured at 290 K (Figs 4a and e), 673 K (Figs 4b and f), 873 K (Figs 4c and g) and 973 K (Figs 4d and h). Below 673 K, a grain elongated parallel to the tensile axis was observed, for example, in Fig. 4a. Fracture occurred transgranularly with a slant-type appearance (Fig. 4e). At 673 K and above, an offset at grain boundaries and wedge-shaped cracks at or along the boundaries were visible, as exhibited in Figs 4b to d. It is well established that the grain boundary offset and wedge-cracks result from grain boundary sliding (GBS). The occurrence of GBS in a Ni_3Al-based polycrystal at 673 K and above is also pointed out by Oya *et al.* [9] and Weihs *et al.* [2]. The fact that the grain shape after deformation changes from an elongated grain (Fig. 4b) to an equiaxed one (Fig. 4d) with increasing temperature indicates that the occurrence of GBS is favoured with increasing temperature. When specimens were tested at 873 K and above, the fracture surface was made up of grain boundary facets having a dimple-like pattern (Figs 4g and h). In addition, a nodular appearance was clearly visible in the fracture surface and in the offset region indicated by an arrow in Fig. 4d. The occurrence of such a nodule increased with increasing temperature.

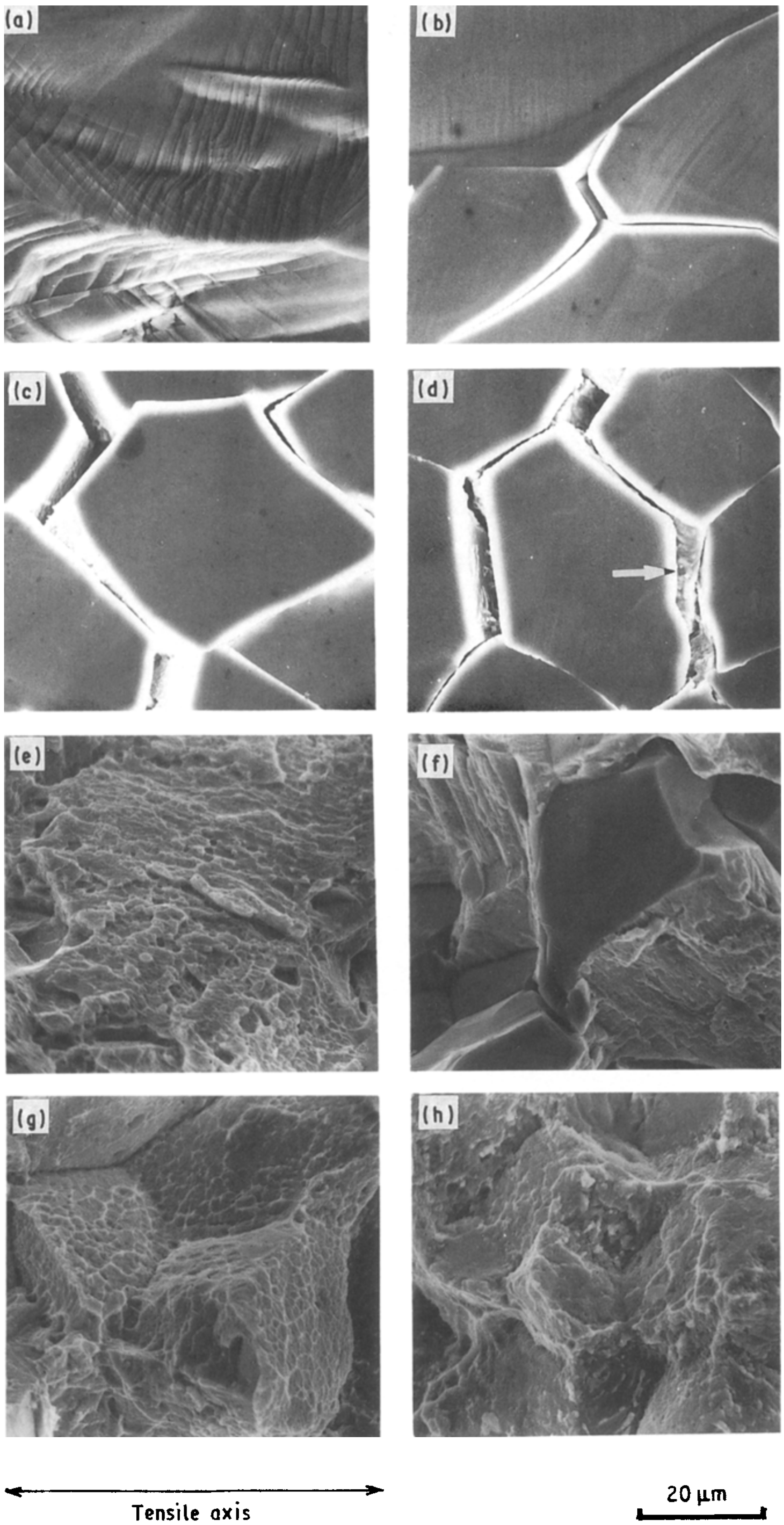

Figure 4 SEM micrographs showing (a–d) longitudinal sections and (e–h) fracture surfaces of specimens having a grain size of 28 μm tested at (a, e) 290 K, (b, f) 673 K, (c,g) 873 K and (d, h) 973 K.

A higher-magnification view of the arrowed area of Fig. 4d is illustrated in Fig. 5. Nodules below 1 μm in diameter are developed on grain boundaries. To depict the internal structure of the nodule, TEM observation was made on the same specimen as Fig. 5. The result is shown in Fig. 6. Note the recrystallized grains which contain a high density of dislocations at and in the vicinity of an original grain boundary, indicating that dynamic recrystallization occurs and results in the existence of nodules. The occurrence of dynamic recrystallization at 873 K and above has also been evidenced by Baker *et al.* [17] using a stoichiometric Ni_3Al. Moreover, the presence of nodules has also been reported by Weihs *et al.* [2] for stoichiometric Ni_3Al with 0.35 at % B at 873 K and above, and by Takeyama and Liu [11] for boron- and hafnium-doped nickel-rich Ni_3Al above 1073 K.

For specimens having a grain size of 8.5 μm the fracture mode changed with temperature in a similar way to the case of $d = 28\,\mu$m, as indicated in Fig. 7.

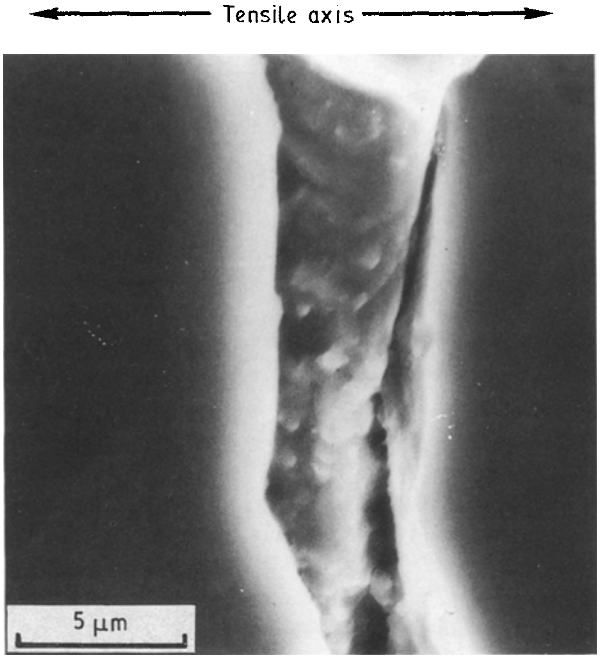

Figure 5 Higher magnification view of the arrowed area of Fig. 4d.

Fig. 8 shows the temperature dependence of the fracture mode for specimens with $d = 1.6\,\mu m$. Below 673 K, the fracture surface exhibited a cup–cone-type appearance. A representative fractograph is given in Fig. 8a, illustrating the central area of the fracture surface tested at room temperature. The jagged appearance in this figure is characteristic of a normal rupture where fracture initiates. At 873 K and above, deep cavities or holes existed in the fracture surface as shown in Figs 8c and d. Mixed modes of cup–cone fracture and "cavitation" fracture are observed in Fig. 8b, where the specimen was deformed at 673 K. A similar relationship between fracture mode and temperature was found for specimens having a grain size of $2.9\,\mu m$.

4. Discussion

The important role of temperature and grain size in deformation and fracture behaviour has been clearly established through the metallographic features shown by recrystallized Ni_3Al polycrystal doped with boron. The temperature dependence of elongation and fracture mode will be discussed as a function of grain size.

4.1. Coarse-grained specimens ($d = 105\,\mu m$)

With increasing temperature the strain to fracture decreased rapidly and the fracture mode changed from transgranular fracture to an intergranular one through mixed modes of these. At 473 K and below, a transgranularly fractured surface consisted of flat, smooth $\{1\,1\,1\}$ facets formed with a stepped, blocky structure, which is characteristic of $\{1\,1\,1\}$ cracking. A detailed mechanism concerning $\{1\,1\,1\}$ cracking has been demonstrated in $Ni_3(Al, Ti)$ single crystals with [12] and without boron [13–15]. According to these single-crystal studies, $\{1\,1\,1\}$ cracking occurs by the joining of microcracks produced by the intersection of operative slip bands in the form of planar arrays of

Figure 6 TEM micrograph illustrating the internal structure of Fig. 5. Note the recrystallized grains containing dislocations at and in the vicinity of an original grain boundary.

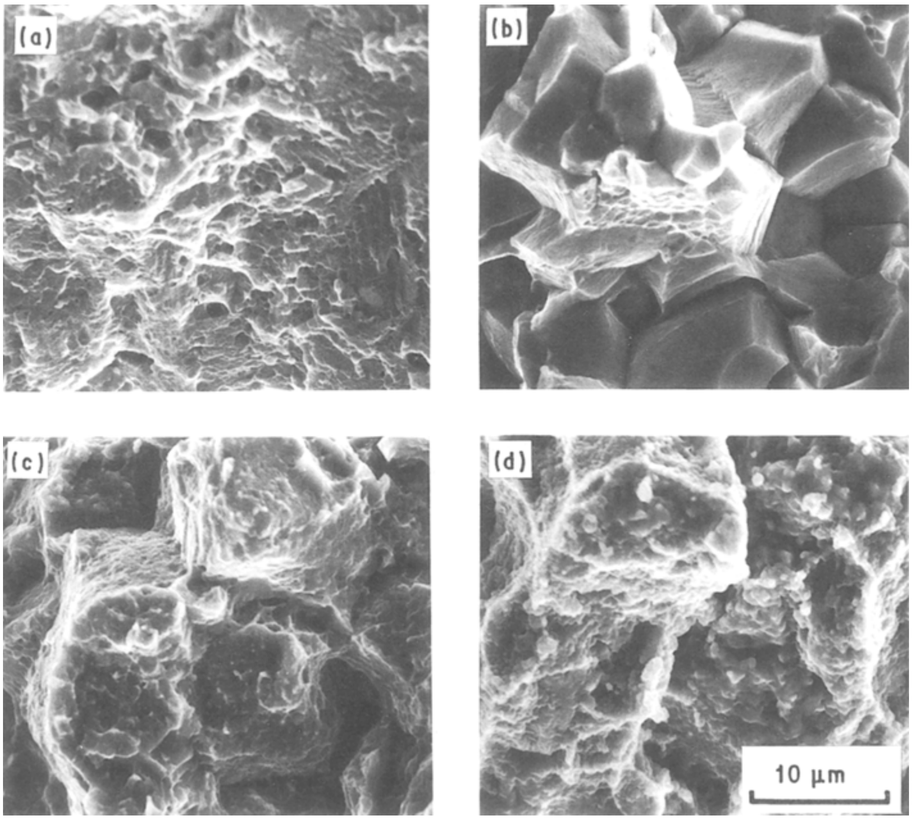

Figure 7 SEM fractographs of specimens having a grain size of 8.5 μm fractured at (a) 290, (b) 673, (c) 873 and (d) 973 K.

dislocations. An increase in stress level will therefore facilitate the formation and joining of the microcracks and reduce the strain to fracture. This view has actually been evidenced in both single crystals [15, 18] by the negative temperature dependence of the elongation at temperatures where positive temperature dependence of the flow stress is exhibited.

In the argument for polycrystals, a stress will allow stress concentration to occur at grain boundaries as well as intersections of slip bands, and intergranular fracture will appear when the concentrated stress at grain boundaries exceeds the cohesive strength of the boundaries. Moreover, at 673 K and above where GBS occurs, the effect of stress concentrations arising from GBS should be taken into account in addition to that of dislocation glide, because it is well recognized that GBS results in a stress concentration at grain boundary junctions or along the boundaries to cause an intergranular fracture [19, 20]. It is therefore expected that for coarse-grained specimens an increase in stress level with increasing temperature accelerates the intergranular fracture and limits the strain to fracture, by further concentrated stress at grain boundaries due to dislocation glide and GBS. This view is verified by the experimental results in Figs 2 and 3.

4.2. Fine-grained specimens (d < 8.5 μm)

The fracture surface exhibited a cup–cone fracture appearance for fine-grained specimens tested at 473 K and below. It is known [8] that a cup–cone fracture initiates as a normal rupture resulting from the formation of voids by dislocation interactions and their subsequent coalescence by localized plastic strains. The final separation by shear rupture leads to the "cone" part of the fracture. Recent study [8] also revealed that at room temperature the tendency for void formation increases with increasing stress level, namely with decreasing grain size. This finding seems to be valid until 473 K, because a larger elongation is developed for the specimen of d = 2.9 μm compared with that of d = 1.6 μm at a given temperature in this temperature range.

When specimens were deformed at 673 K and above where GBS occurs, the fracture surface had a large number of deep cavities. It is well established that GBS plays a prominent role in promoting the formation and growth of intercrystalline cavities [21]. Deep cavities or holes in Figs 7c and d indicate that cavitation fracture results from the growth and linking of cavities formed at or along grain boundaries by stress concentration due to GBS. There is thus a rough similarity between cup–cone fracture and cavitation fracture in the respect that the entire process of fracture is composed of cavity formation and coalescence. The main difference between the two fracture modes is the source of the cavities, namely, whether cavities are formed mostly by GBS at 673 K and above or by dislocation interaction below that temperature.

It is natural to think that the stress concentration which arises at irregularities on sliding boundaries can be accommodated by some relaxation process, for example, dynamic recrystallization. Such being the case, at 873 K and above where both GBS and DR occur, the elongation to fracture may be controlled by the competition between the promotion of a cavity by GBS and its suppression (accommodation) by DR. Note that in this temperature range the elongation for

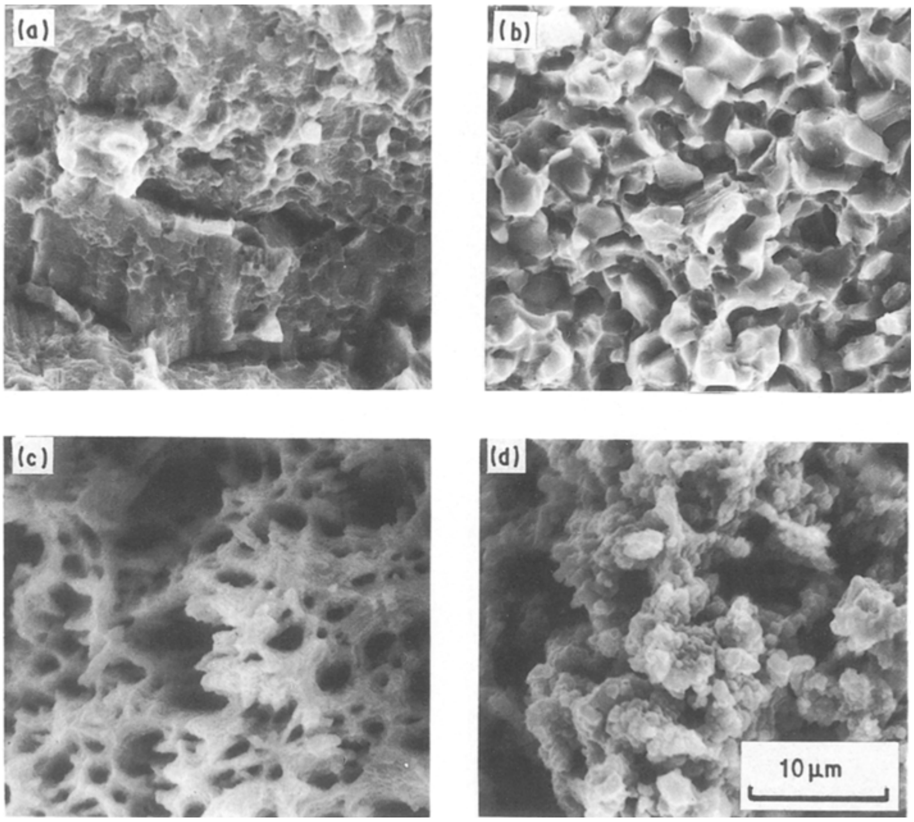

Figure 8 SEM fractographs of specimens having a grain size of 1.6 μm fractured at (a) 290, (b) 673, (c) 873 and (d) 973 K.

the specimen of $d = 1.6\,\mu$m is larger than that of $d = 2.9\,\mu$m at a given temperature (Fig. 2). This fact can be understood as follows: because DR preferentially occurs at or in the vicinity of grain boundaries as verified by metallographic observations in Fig. 6, the total volume fraction of dynamically recrystallized grains is considered to increase with decreasing grain size. Moreover, McLean [22] has pointed out that a spherical cavity will be stable if $r > (2\gamma/\sigma)$ where r is the cavity radius, γ the surface energy and σ the tensile stress at the grain boundary. Assuming that r is proportioned to sliding length, and in turn to grain size, the stress to produce a cavity is increased with decreasing grain size. Therefore, improvement in elongation by refining grains may be resulting from the further occurrence of DR which relieves the stress concentration, and the limited sliding distance which raises the stress for cavity formation.

In general, the occurrence of DR is also dependent on the strain rate in addition to the grain size and temperature. Very recently, by careful controlling of these values, we have succeeded in finding superplasticity with a maximum elongation of 160% in the same composition of compounds [3, 4]. It was also found that DR is an important process operating during the superplastic deformation [4].

4.3. Intermediate-grained specimens ($28\,\mu\text{m} \geqq d \geqq 8.5\,\mu\text{m}$)

At 473 K and below, a slant-type fracture appeared with relatively large elongation for intermediate-grained specimens. It has been pointed out [8] that a slant-type fracture is the result of the shear rupture formed by separation along the plane of maximum shear strain concentration with the retardation of both {1 1 1} cracking and cup–cone fracture. At 673 K where GBS occurs, intergranular fracture was exhibited in addition to the slant-type fracture. Above that temperature the fracture surface consisted mostly of grain boundary facets possessing a dimple-like pattern. Moreover, the so-called "wedge" type of intergranular crack is clearly visible in the longitudinal section of the specimens fractured at 673 K and above. It has been repeatedly demonstrated [19, 20, 23] that a wedge-shaped crack initiates mostly at grain boundary junctions by GBS and propagates along the grain boundaries to give intergranular fracture.

Assuming that unstable fracture occurs if a critical displacement is produced near the tip of a growing crack, intragranular deformation may also affect the growing rate of the crack. If sufficient plastic relaxation occurs in the grains to blunt a crack, the crack once formed propagates more slowly and thus final fracture occurs at higher strain. The existence of a dimple-like grain boundary facet and an isolated crack on the fracture surface supports this view. Therefore, in the temperature range where GBS and DR occur, the strain to fracture in Fig. 2 for intermediate-grained specimens is presumably related to the GBS, DR and plastic flow. In contrast, as discussed earlier, for coarse-grained specimens ($d = 105\,\mu$m) brittle intergranular fracture has undoubtedly occurred by the rapid growth or nucleation of a small number of long, intercrystalline cracks, which is analogous to the absence of non-propagating cracks.

5. Conclusions

The temperature and grain size dependence of the yield stress, ductility and fracture behaviour in recrystallized Ni_3Al polycrystals doped with boron was investigated by tensile tests at temperatures up to 973 K in the range of grain sizes from 1.6 to 105 μm. The results obtained are summarized as follows:

1. The yield stress showed a positive temperature dependence to a peak temperature in somewhat different manners depending on the grain size.

2. For coarse-grained specimens (grain size $d = 105\,\mu$m), the elongation decreased rapidly with increasing temperature. The predominant fracture mode changed with temperature from the transgranular fracture of $\{1\,1\,1\}$ cracking to brittle intergranular fracture. Such an embrittlement at high temperature was interpreted by a high stress concentration at grain boundaries caused by increased flow stress level and the occurrence of grain boundary sliding.

3. For intermediate-grained specimens ($28\,\mu m \geqq d \geqq 8.5\,\mu m$) the elongation was not so markedly decreased with temperature. The fracture mode changed from a slant-type fracture at low temperatures to a ductile intergranular fracture at high temperatures. The suppression of serious high-temperature embrittlement was explained in terms of the slow propagation of cracks formed by GBS, owing to stress relaxation by dynamic recrystallization and plastic flow.

4. For fine-grained specimens ($d < 8.5\,\mu$m), the elongation at elevated temperatures was slightly decreased (when $d = 2.9\,\mu$m), or rather increased ($d = 1.6\,\mu$m), compared with the respective value at room temperature, while the fracture mode changed from cup–cone fracture at low temperatures to cavitation fracture at elevated temperatures for both specimens. Achievement of the large elongation at elevated temperatures for specimens with an ultra-fine grain size of 1.6 μm was explained as that the further occurrence of DR with increasing volume fraction of grain boundaries sufficiently accommodates the cavitation promoted by GBS, and that the restricted sliding distance due to extremely small grain size raises the stress for cavity formation.

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