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Crystal Growth and Cold-Rolling of Two Phase Ni-Base Superalloys

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Abstract

Single crystal growth of the γ' (Ni_3Al) + γ(Ni-base solid solution) two-phase Ni-base superalloys with 16 – 22 at%Al was tried using floating zone method. It was found to be possible to grow single crystals of the low Al contents alloys from the melt. The influence of the Al-content on the solidification behavior was studied. The microstructure of the as-grown alloys consisted of γ' precipitates and γ- phase solid solution matrix. The morphology of the γ' particles ranged from cuboidal shape in alloy Ni-16at%Al to irregular faceted shape in Ni-22at%Al. The possibility of cold-rolling of these two phase Ni-base superalloys into thin foils without intermediate annealing have been studied.

Keywords: superalloys; crystal growth, microstructure, cold-rolling

1. INTRODUCTION

Ni_3Al (hereafter γ') is attractive in applications requiring strength at high temperatures.[1-20] The main obstacle of γ' as potential materials for use at elevated temperatures is their low room temperature ductility in polycrystalline form. At high temperatures, the grain boundaries that are normal to the principal stress axis constitute sources of weakness and preferential nucleation sites for cracks, which may eventually lead to the failure. It was established that grain boundaries of γ' are intrinsically brittle.[5,6] The brittleness of polycrystalline γ' is argued to be due to poor grain boundary cohesive strength.[7,8]

This metallurgical problem was solved by Aoki *et al.*[2], Liu *et al.*[5], who discovered that small additions of boron to Ni-rich γ' greatly improved the ductility by changing the fracture mode from intergranular to transgranular.

Recently Hirano at *al* found that boron-free $\gamma'$was able to be significantly ductilized by the unidirectional solidification using a floating zone method (FZ-UDS) without any alloying elements.[10,11] Stoichiometric γ', in columnar-grained crystal form, was quite ductile and were able to be strained as high as 60% of elongation in tensile test,[12-14] which was much more ductile than that of the boron-doped alloy.

It is known that it is impossible to fabricate boron-doped γ' foils by cold-rolling. The ductility is not enough high to cold-roll to thickness below 800μm. However, taking advantage of the high ductility of the FZ-UDS γ' the fabrication of ductile thin foils of stoichiometric γ' by cold-rolling without intermediate annealing was possible.[15] This technique successfully fabricated thin foils and these foils showed 3.0-14.6% tensile elongation at room temperature after heat treatment.[15]

Using the same technique we investigated possibility of growing two-phase (γ-γ') Ni-rich superalloys with compositions of 16 - 22 at%Al. The first purpose is to study the differences in solidification behavior of the non-stochiomentric Ni-rich composition in comparison with stoichiometric γ'. The second purpose is to fabricate thin foils by cold-rolling. We expected that the thin foils could be promising for high-temperature aerospace and automobile applications as light-weight high-temperature structural materials, for example, honeycomb structure.

2. EXPERIMENTAL PROCEDURE

The raw materials of Ni-rich superalloys were prepared as the feed rods for FZ-UDS in the same way as in the previous report [17]. All Ni-rich alloys were unidirectionally solidified at the growth rate of 25mm/h by the FZ-UDS. The Al-content of the as-grown ingots was determined by inductively coupled plasma spectroscopy. The results agreed fairly well with nominal compositions within 0.4 at%, as listed in Table I.

The as-grown ingots were 9 – 13 mm in diameter and 120 mm in length. The longitudinal sections of the ingots were subjected to metallographic observation. X-ray Laue diffraction was used to check the crystal orientation.

Cold-rolling specimens (1.7 × 8-10 mm in cross section and 80-100 mm in length) were cut from the ingots with the longest dimension parallel to the growth direction by electrical discharge machining. Cold-rolling was carried out along the longest dimension at room temperature by using four-high mills with a back-up roll diameter of 360 mm and a working roll diameter of 120mm. The rolling speed was 3.5m/min. The amount of reduction in thickness was about 0.1mm per pass. The cold-rolling operations were performed without intermediate annealing.

The microstructures before and after the cold-rolling were examined by optical and scanning electron microscopy (SEM). Since the difference in composition between the matrix and γ'-phase was relatively small, the contrast in backscattered electron images was only slight, and it was often necessary to etch specimens lightly before examination. The surfaces of the specimens were mechanically polished with emery papers and finished with 0.05 μm alumina powder suspension, then etched in the Marble reagent (5g $CuSO_4$, 20 ml HCl and 20 ml H_2O).

3. RESULTS AND DISCUSSION

3.1 Crystal growth and microstructures of as-grown crystals

Nickel-rich region of the Ni-Al phase diagram (Fig. 1) shows that the γ-phase exists over a wide range of composition, with a maximum melting point of 1455 °C. The γ'-phase, which exists in a narrow range of composition less 24.4 at %Al, is produced via eutectic reaction at 1385 °C[18].

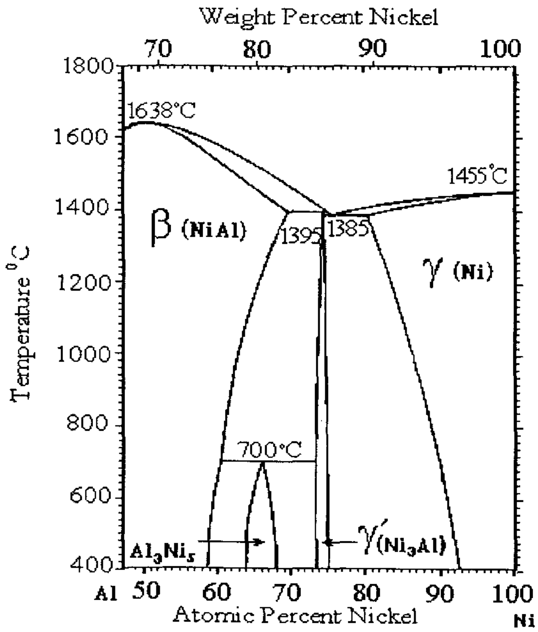


Fig. 1. The nickel-rich region of the Ni-Al system.[18]

Because of the phase diagram both melting and solidifying interfaces were not clear but broadened during crystal growth as schematically shown in Fig. 2. The width of each interface (h) increased with the decreasing Al content. It indicates that γ and γ' grew separately from the liquid. This is different from the case of stoichiometric γ' phase where planar solidification interface is formed.[11] When Ni-rich alloys are cooled from the melt, γ phase primarily solidifies (liquid → liquid + γ). Then at the eutectic temperature, the remaining liquid freezes as a two-phase mixture of γ and γ'. Cooperative growth of these two phases may be possible with the same crystallographic orientation because lattice parameters of both phases differ only by 1%[19-20].

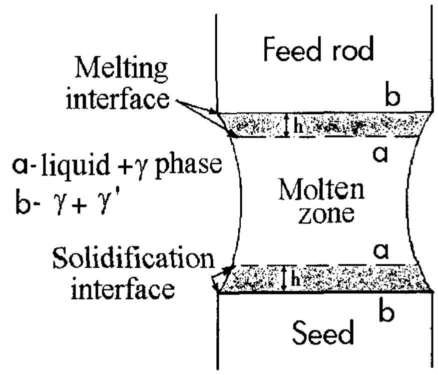


Fig. 2. Schematic of the molten zone

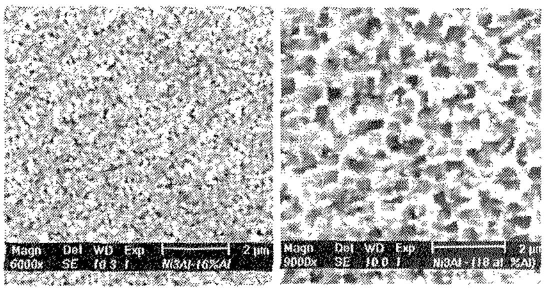
In the actual operation of crystal growth, a little thermal fluctuation in the molten zone and slight compositional fluctuation of the feed rods could disturb a stable solidification process. This often led to difficulty of growing a large single crystal of two-phase Ni-rich composition from liquid.

Nevertheless, we successfully grew a single crystal of 70 mm in length toward near [001] orientation for Ni-16Al. For the other alloys columnar-grained polycrystals were obtained as summarized in Table I. Ni-18Al consisted of a few well-developed columnar grains. In the case of Ni-20Al more columnar misorientation of the grains were included compared to Ni-18Al and the grain boundaries were found to be mainly low-angle less than 12°. In Ni-22Al the columnar structure was formed along the growth direction.

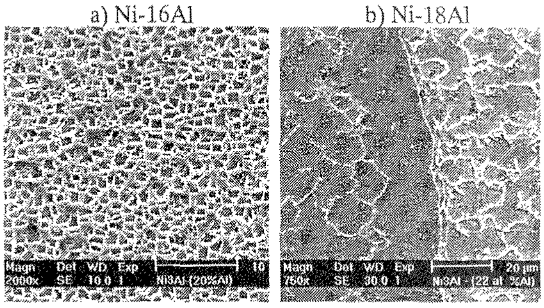
Table I. Chemical composition and as-grown structure of FZ-UDS two-phase Ni-rich crystals.

Sample	Al content, at %	As-grown structure
Ni-16Al	15.9	single crystal
Ni-18Al	18.1	well-developed columnar structure
Ni-20Al	19.7	columnar structure
Ni-22Al	21.6	columnar structure

SEM microstructures of the as-grown crystals are shown in Fig.3. The characteristics of the microstructure are morphology of γ' phase. The γ' particles ranged from cuboidal shape in Ni-16Al to irregular faceted in Ni-22Al. The Fig. 3(a, b) shows, that Ni-16Al and Ni-18Al possess discontinuous networks of γ-phase (gray) containing γ'-phase (dark). The average γ'- particle size in Ni-16Al and Ni-18Al alloys were 150 nm and 300-500nm, respectively.



a) Ni-16Al b) Ni-18Al



c) Ni-20Al d) Ni-22Al

Fig. 3. SEM microstructures of the as-grown crystals.

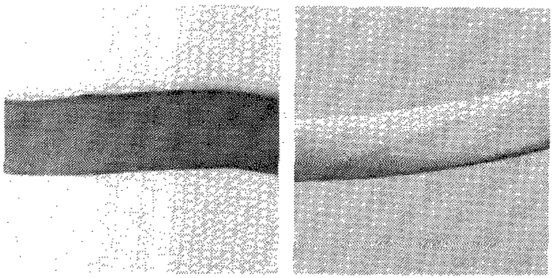
In Ni-20Al and Ni-22Al the irregular γ' precipitates (dark gray) were formed, showing a crystallographic faceting nature of the γ'/γ interface. The particles of γ'-phase (Fig. 3 (c)) were arranged in a square array.

The as-growth Ni-22Al microstructure consisted of cored dendritic arms in the direction of crystal growth axis. The cores of the dendrites were composed of two-phase mixture of residual γ in γ'. The average γ' size was found to be ~ 25 μm.

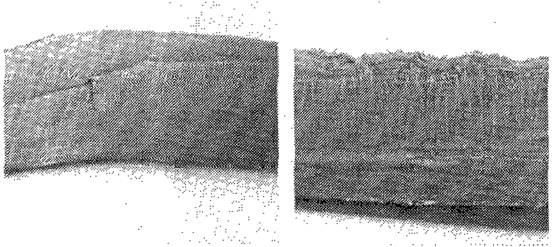
The growth conditions for all the alloys, i.e. growth and cooling rate were the same. However, γ'-particle size increased with increasing Al content. In addition, only slight variations in γ' precipitate size existed between the top and bottom of the grown ingots.

3.2 Cold-rolling, texture and microstructure of as-rolled crystals

Here we present successful fabrication of thin foils of two-phase Ni-rich superalloys by cold-rolling of the directionally solidified ingots. Thin foil of 200 μm in thickness was successfully produced with 80% reduction at room temperature without intermediate annealing. Figure 4 shows the appearance of the foils after 80% reduction for all alloys. The Ni-16Al, Ni-18Al and Ni-20Al foils have a good surface quality. The surface is smooth, flat and crack free. During cold-rolling one foil of Ni-16Al was eventually bent double after 50% reduction. However, no cracks appeared. Single cracks were observed only along the high-angle grain boundaries in polycrystalline Ni-18Al, Ni-20Al and Ni-22Al. It is well known that this type of grain boundaries is very brittle in single phase γ', which is not exception in two-phase Ni-rich superalloys (Fig. 4(c)). In contrast, low-angle grain boundaries are crack-resistant (Fig. 4(b)). Crack initiated along high-angle grain boundaries after 60 % reduction in Ni-18Al and Ni-20Al composition (Table II). The Ni-22Al did not break up to about 80% reduction but many cracks were observed at the edge of the specimen after 28 % reduction (Fig. 4(d)).



a) Ni-16Al b) Ni-18Al



c) Ni-20Al d) Ni-22Al

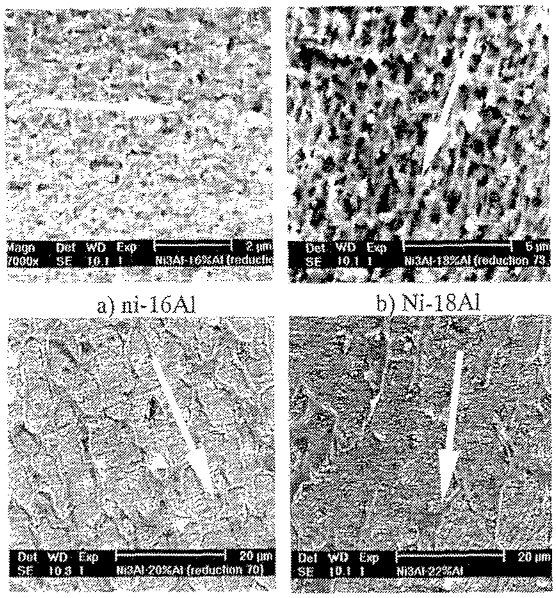
Fig. 4. General view of the as-rolled foils.

Table II. Results of cold rolling: initial rolling orientation, final rolling reduction, reduction when cracking starts and rolling texture.

Sample	Initial rolling orientation	Final reduction (%)	Reduction where cracking starts (%)	Rolling texture
Ni-16Al	<1.0 0.8 3.8>	80	—	{110}<102> +{110}<121>
Ni-18Al	<0.1 0.0 0.9>	76	60	{110}<771>
	<0.3 0.0 0.9>	82		+{110}<121>
Ni-20Al	—	80	62	{110}<116>
Ni-22Al	—	80	28	{110}<241> +{110}<121>

There are some differences in rolling texture among the samples, as shown in Table II. Foil Ni-20Al has a single strong {110}<116> texture. Foils Ni-16Al, Ni-18Al and Ni-22Al have a weak texture {110}<121>, and in addition, strong {110} texture existed around rolling direction <102>, <771> and <241>, respectively. The {110} rolling plane of two-phase Ni-rich superalloys is the same as that of stoichiometric Ni_3Al^{15}. The texture can be related to the initial rolling orientation, but the details are not known.

It should be noted that polycrystalline γ-γ' alloys could be cold-rolled without the addition of ductility enhancing elements.



a) ni-16Al b) Ni-18Al

c) Ni-20Al d) Ni-22Al

Fig. 5. SEM microstructures of the as-rolled foils. Arrows indicate the rolling directions.

Figure 5 shows the SEM micrographs of the polished and subsequently slightly etched foils. The γ' precipitates are elongated along the rolling direction. The shape changes: a) from cuboidal to rectangular (Fig. 3(b), Fig. 5(b)); b) irregular facets to elongated facets or rectangular (Fig. 3 (c, d) and Fig. 5 (c, d)). The grains are uniformly elongated toward the rolling direction and long slip traces are observed in the elongated grains.

4. CONCLUSIONS

Single crystal growth of the $\gamma'+\gamma$ two-phase Ni-base superalloys with 16-22 at%Al was tried using by floating zone method. It was found single crystals were grown for Ni-16at%Al, while columnar-grained polycrystals were grown for other alloys. The microstructure of those in as-grown crystals consists of γ' precipitates and γ-phase solid solution matrix.

It was found to be possible to fabricate foils of the polycrystalline these alloys at room temperature by cold rolling. Total reduction of 80 % was obtained from the initial 1.7mm-thick sheet. No heat treatment was performed during the entire rolling process. It can be noted that such thin foils were produced from two-phase Ni-rich superalloys without alloying elements and intermediate annealing treatment.

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