# THE EFFECT OF PROCESS VARIABLES ON THE STRUCTURE AND PROPERTIES OF ODS, $\gamma'$- HARDENED NICKEL-BASE SUPERALLOYS

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## Abstract

The purpose of this investigation is to obtain a fundamental understanding of the underlying mechanisms of coarse grain recrystallization of ODS, $\gamma'$ - hardened nickel-based superalloys. Experiments have been carried out on a laboratory zone annealer using small bars in which a number of directional recrystallization (DR) speeds and temperatures were evaluated, enabling a 'process window' for satisfactory DR response to be established, on the basis of both structure and stress rupture properties. The influence of the thermal history associated with post extrusion thermomechanical processes on the recrystallization response has also been studied.

Quenching experiments from a calibrated gradient zone annealing furnace have shown that nucleation of secondary recrystallization occurs at higher temperatures than that at which the recrystallization interface grows at a given zoning speed. Microbeam electron diffraction (MED) has enabled the orientation of the submicrometre sized equiaxed grains ahead of the recrystallization front to be studied, and differences in behaviour between INCONEL alloys MA6000 and MA760 have been identified. Selected Area Channelling Patterns (SACPs) in the scanning electron microscope has enabled the texture and average grain misorientations of commercially produced bars of INCONEL alloys MA6000 and MA760 to be established.

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## Introduction

The factors that determine the secondary recrystallization behaviour of Mechanically Alloyed ODS materials are not fully understood. The purpose of the present investigation is to obtain a fundamental understanding of the underlying recrystallization mechanisms of ODS, $\gamma'$ -hardened nickel-base superalloys, and to quantify the effects of the many process variables in their commercial production on the directionally recrystallized (DR) structure and resultant mechanical properties of the final product.

The commercial manufacture of ODS, $\gamma'$-hardened nickel-base superalloys, such as INCONEL** alloys MA6000 and MA760, for gas turbine rotor blade and vane applications can be separated into three essential component processes: mechanical alloying of the powder, consolidation by thermomechanical working to the desired shape, and finally the critical Directional Recrystallization (DR) which is achieved by zone annealing. Individually, each of these processes has a significant effect on the characteristic microstructure and mechanical properties of the final product.

## The Effect of Processing Conditions

There are a number of thermomechanical process steps following extrusion that are necessary for the commercial production of this type of material. Since directional recrystallization is primarily driven by stored energy derived from the combination of all the process steps, the time and temperature exposure of these additional operations can have a significant effect on the subsequent DR response. These aspects formed the basis of studies at the IRC, Birmingham University.

### Materials and Methods

Initial experiments on the effect of zone annealing of INCONEL alloy MA6000 were carried out on rectangular samples cut from 23 x 25 mm bar. Subsequent studies of the effect of extrusion conditions on zone annealing response were carried out on similar samples cut from 35 x 67 mm experimentally extruded bar.

Zone Annealing Treatment. A 3 turn silicon carbide element of approximately 22 mm bore operated inside a thermally insulative cowl, rests on a 25 mm thick board of Fibrefrax with a small hole in the centre to allow the test-piece to enter the hot zone. The element temperature is controlled by a Eurotherm 822 16 programme process controller fed from a thermocouple attached to the outside of the element at the hottest point 19 mm from the bottom. The speed range of this rig is from 1 to 1000 mm/hour.

Method of Experimentation. First it is necessary to determine the temperature range over which it is possible to obtain DR. For this a suitable speed is chosen and a 15 mm length run at the 1st temperature selected. The temperature is then reduced to a temperature at which DR cannot take place e.g. 900°C and a 2 mm length run at this temperature. The temperature is next increased to the 2nd value and a 15 mm long segment run at this. This procedure is continued until 4 segments with "stoppers" between them has been achieved on one bar. Further runs are performed until the temperature range over which DR will take place is mapped out.

Runs made at a fixed temperature with varying rig speed in each of the segments, dividing them with "stoppers" as before allows the complete temperature/speed map to be drawn.

** Trade mark of Inco group of companies.

Zone Annealing Response

The Effect of Zone Annealing Conditions on Structure and Properties. A study has been made of the effect of zone annealing speeds and temperatures on the Directional Recrystallization (DR) structure and stress rupture properties of INCONEL alloy MA6000. The maximum range of zone anneal speed for satisfactory DR is from 15 mm/h up to 400 mm/h. However, as shown in Fig. 1, this is effected by the maximum set temperature in the zone annealer. These parameters critically define the process window of zone annealing for DR, outside of which, recrystallized structures can be obtained but generally with grains of low aspect ratio.

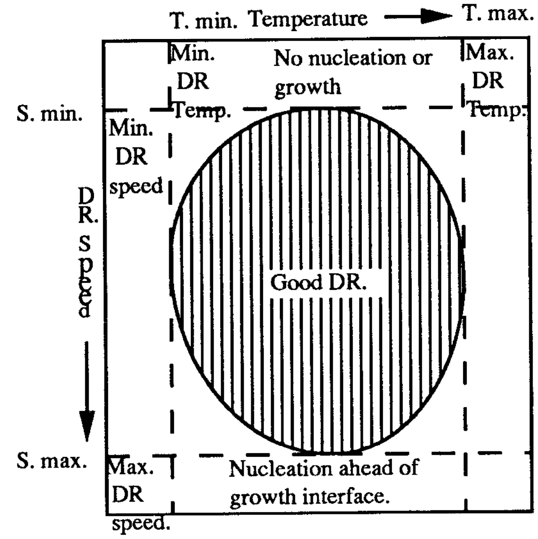



Figure 1- Schematic process capability chart for Directional Recystallisation for a given precursor condition.

The effect of zone annealing speed on the stress-rupture life at 1093°C/130 MPa of INCONEL alloy MA6000 bar produced under a given set of TMP conditions is shown in Fig. 2. This indicates an apparent threshold speed to achieve high stress rupture properties. At higher speeds, a slight steady decrease in life is observed with increasing zone annealing speed. Similarly it can be seen from Fig. 3 that there is both a minimum and maximum temperature threshold to achieve good stress rupture life.

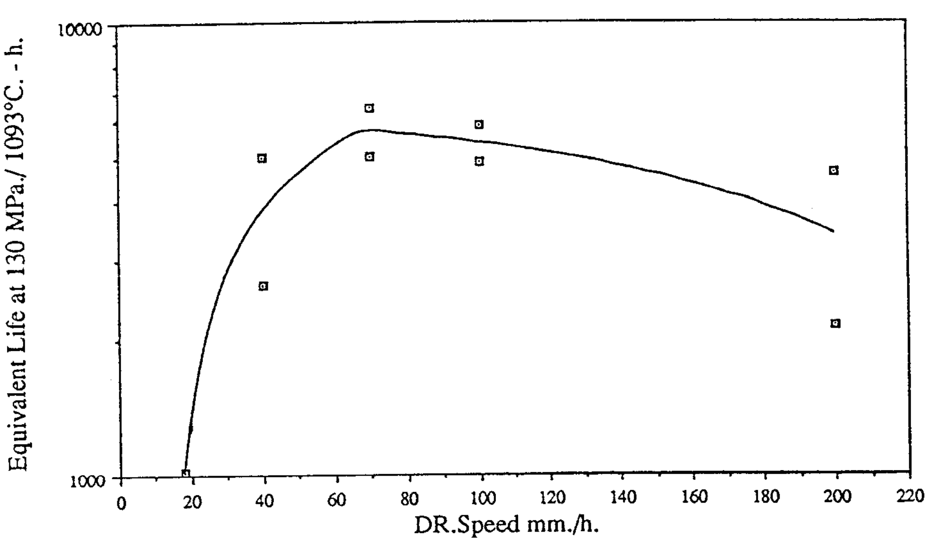



Figure 2- Effect of DR speed at 1220°C on equivalent stress-rupture life at 1093°C/130 MPa of INCONEL alloy MA 6000.

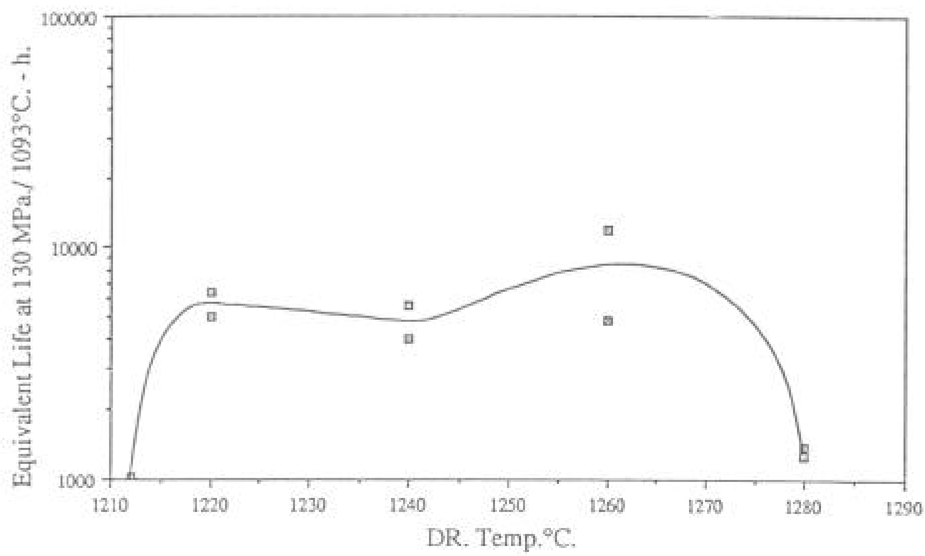



Figure 3- Effect of DR temperature at a fixed speed on equivalent stress rupture life at 1093°C/130MPa of INCONEL alloy MA6000

The Effect of Extrusion Conditions on DR. A study of the effect of extrusion speed over the range 25 to 90 mm/sec on the 35 x 67 mm section bar at extrusion temperatures of 950°, 975° and 1000°C has been made on Mechanically Alloyed INCONEL alloy MA6000 powder. It was previously believed that good quality DR structures could only be produced using unique extrusion conditions, fast as possible at the lowest temperature. However, Fig. 4 shows that satisfactory DR can be obtained at speeds down to 50 mm/sec at temperatures up to 975°C, conditions well within the capabilities of the recently refurbished extrusion press at Inco Alloys Limited, Hereford UK. Interestingly the bar extruded at 1000°C, 50 mm/sec shows low aspect ratio, almost equiaxed, coarse recrystallized grains. This demonstrates the need for highly automated computer controlled equipment where process conditions can be accurately reproduced to exploit this process window, and control the attributes and properties of the final product.

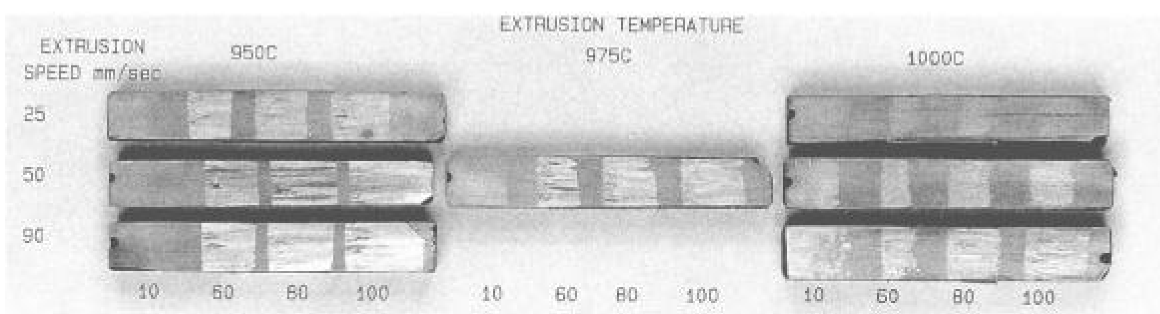



Figure 4- Interaction of zone annealed speed and extrusion parameters on recrystallisation response of INCONEL alloy MA6000

All Mechanically Alloying experience of ODS $\gamma'$- hardened nickel-base superalloys has shown that, either some form of DR structure is developed, or the grains grow from ~0.2 to ~0.6 µm and refuse to grow any larger. This is clearly illustrated (Fig. 4) in all the "stoppers" between the zones DR at various speeds in the DR tests. However, under these conditions of extrusion, coarse equiaxed grains have been obtained in the zones annealed at 60, 80 and 100 mm/h.

The Effect of Preannealing on Zone Annealing. There are a number of additional thermomechanical process steps, following extrusion, that are necessary for the commercial production of this type of material. Since DR is primarily driven by stored energy derived from the combination of all the process steps, the time and temperature exposure of these additional operations can have a significant effect on the subsequent DR structure and resultant mechanical properties. Laboratory zone annealing tests on MA6000 have shown, Fig. 5, that at times and temperatures of up to 1 h/1000°C there is no effect on the DR structure. A marginal detrimental effect is experienced at conditions up to 1 h/1040°C and 4 h/1000°C, especially at the faster speeds of DR, with conditions in excess of these causing a breakdown in the DR response.

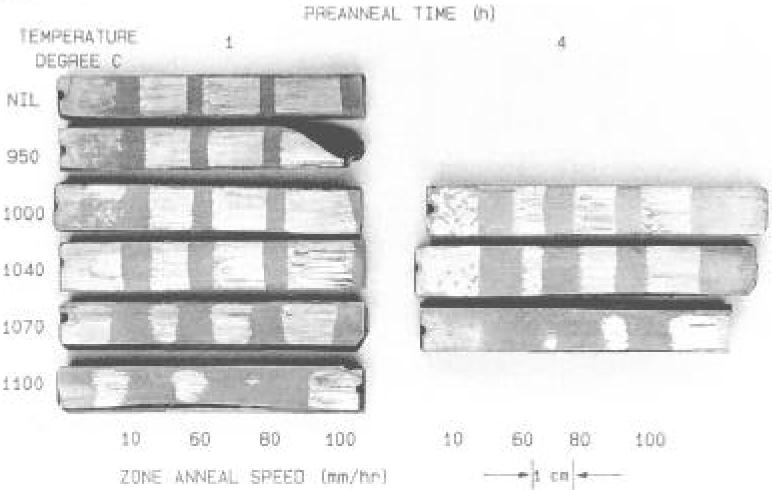



Figure 5- Effect of preannealing condition on response to zone annealing.

Summary. It is clear that both the thermal history prior to zone annealing and the actual zone annealing conditions used have a major effect on the final structure. This suggests that with a better understanding of the basic mechanisms involved in the recrystallization process, improved control of the final grain structure could, perhaps, be achieved in these alloys.

## The Micromechanisms of Zone Annealing

Detailed studies of the micromechanisms of recrystallization in these alloys have been carried out at the Department of Materials, Oxford University.

### Materials and Methods

Initial studies were carried out on specimens of INCONEL alloys MA6000 and MA760 in the form of experimentally extruded cylindrical bars of 10 mm diameter. The microstructures of three commercially extruded rectangular sections, INCONEL alloys MA6000 and MA760 of small cross-section (SCS) and INCONEL alloy MA760 of large cross-section (LCS) were then analysed.

Zone Annealing Treatment. Zone annealing was carried out in a vertical platinum resistance furnace, the details of which are described elsewhere (1). Two series of experiments have been conducted:

(a) Specimens were partially zone annealed at a traverse rate of 38 mm/h through a furnace of maximum temperature 1350°C to produce a recrystallization interface in the sample.

(b) The nucleation process was studied by zone annealing a series of unrecrystallized bars and quenching them from the furnace when the top of the bar reached the required temperature, which was chosen to be progressively higher than that of the interface temperature measured in (a).

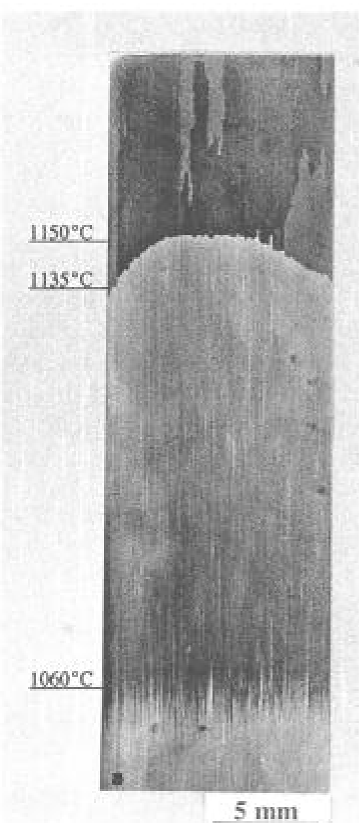
Electron Channelling Patterns. Selected area channelling patterns (SACPs) were obtained from specimens in the scanning electron microscope. By moving the electron beam across a specimen in a direction perpendicular to the direction of growth of the grains it was possible to obtain a series of SACPs from a number of neighbouring grains. Poles were then readily identified with the aid of a channelling map (2).

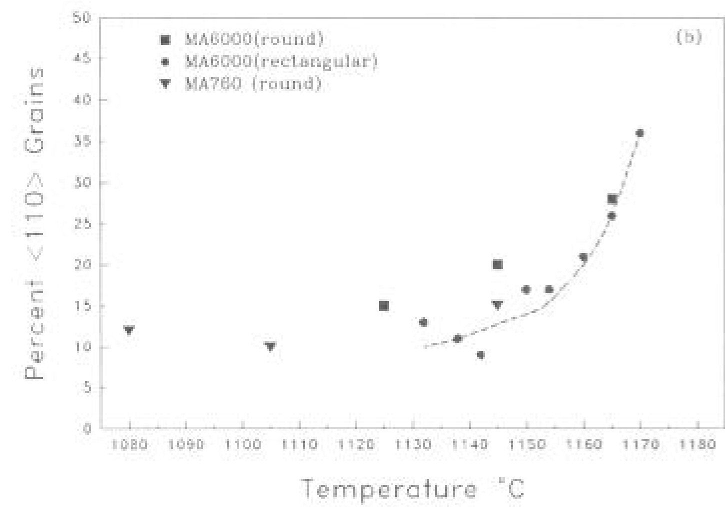
Grain Orientation Determination by TEM. The grains in unrecrystallized samples were too small to be analysed using the SACP technique, so a micro-electron diffraction technique was employed on this foil specimens in the TEM. A pattern was produced from individual submicrometre-sized grains in the unrecrystallized material. The plane of the foils was perpendicular to the extrusion direction of each specimen.

Mechanism of Recrystallization in Experimentally Extruded Bars

As-extruded Material. The material had a fine equiaxed grain structure, of average grain size approximately 0.2 μm.

Zone Annealed Material. A photomacrograph of the interface in partially zone-annealed INCONEL alloy MA760 is shown in fig 6a. The transformation interface is curved, implying that the mobility of the interface is greater at the surface than at the centre of the bar. The $\gamma'$ solvus temperature is 1050 ± 10°C, and a transverse line of changed contrast at this temperature is evident in Fig. 6a.




| Material | Transformation Temperature (°C) | |
|---|---|---|
| | Surface | Centre |
| MA6000(round) | 1155 | 1170 |
| MA6000(rectangular) | 1140 | 1180 |
| MA760(round) | 1135 | 1150 |

Figure 6- (a) Longitudinal section of partially zone-annealed MA760 showing $\gamma'$ solvus at 1060°C. The transformation temperatures are given in the Table.
(b) Showing the change in population of <110> along the temperature gradient in MA760 and MA6000.

SACP analysis of the recrystallized grains showed that their longitudinal axis was close to <110>, i.e. within ± 5°. In an attempt to characterise more fully the structure of the unrecrystallized zone which exists above the solvus temperature, a series of TEM specimens was prepared from points at known positions along the longitudinal axis of the specimens having their plane perpendicular to the extrusion direction. The grain size in this regime was constant and of value close to 0.4 μm. This grain size is consistent with that arising from Zener pinning by the oxide dispersion of mean particle diameter 11 nm and volume fraction 0.025. The majority of grains were oriented in the zone between <100> and <112>. Grains were counted as having a <110> orientation if the {110} pole occurred within the viewing screen of the TEM, i.e. if their orientations were within 10° of {110}. Such grains were usually observed in the plane of section to occur in groups of two or three, rather than in isolation. In INCONEL alloy MA760 the proportion of grains with <110> orientation remains unchanged at 10-14% as the recrystallization interface is approached (Fig. 6b), whereas in INCONEL alloy MA6000, the number of <110> grains was seen to progressively increase as one passed up the temperature gradient. This indicates that a rotation of the grains towards <110> is taking place in INCONEL alloy MA6000. It should be borne in mind that the recrystallization interface in INCONEL alloy MA6000 occurs at a significantly higher temperature (1170°C) than in INCONEL alloy MA760, so that diffusional processes are likely to be more rapid.

<u>The Nucleation of Secondary Recrystallization</u>. The interface in Fig. 6a is the location where the growth rate of the recrystallized grains is equal to the traverse rate of the specimen through the furnace. In order to identify the temperature at which the secondary recrystallization process nucleated in INCONEL alloy MA760, a series of specimens was water quenched after having been passed at the normal traverse rate an increasing distance into the furnace. These were then sectioned and examined metallographically. Fig. 7

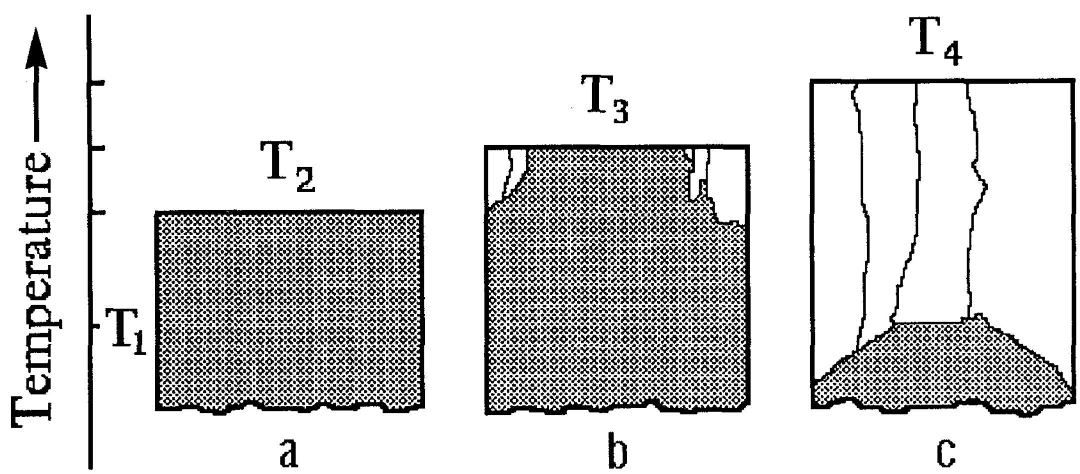



Figure 7- Diamgramatic illustration of nucleation sequence. See test for details.

sketches the structures observed at increasing temperatures above $T_1$, the steady-state interface temperature. No nucleation was observed at $T_2$, ~15° above $T_1$. Quenching from $T_3$ showed that nucleation of recrystallization had commenced at the specimen sides, and quenching from $T_4$ show Fig the recrystallization front to have moved down to $T_1$. A longitudinal se 9 on through the nucleated region is shown in Fig. 8, where it may be seen that isolated internal grains have formed grains of high aspect ratio. Metallographic sections were also made perpendicular to the specimen axis and the TEM foil of fig shows a grain of <110> orientation of about 2 μm diameter in a fine (0.4 μm)-grained matrix.

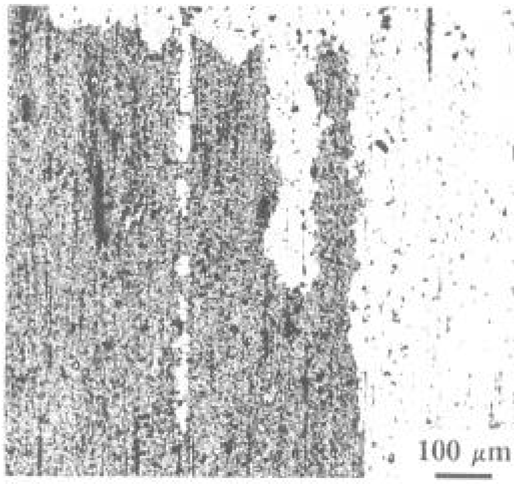



Figure 8 - Optical micrograph of longitudinal section of MA 760 quenced from 1165°C

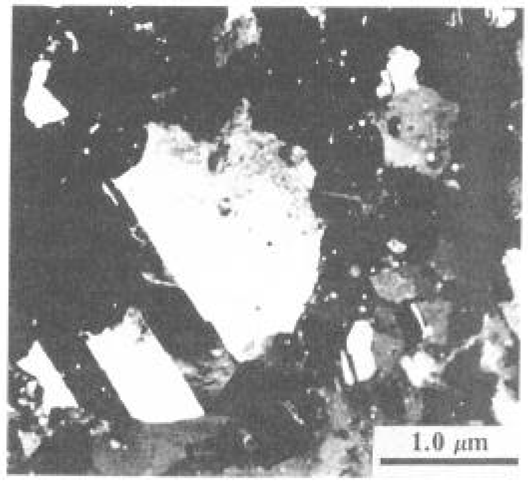



Figure 9 - TEM micrograph of specimen of Fig. 8

Secondary recrystallization appears to be triggered by small groups of grains with axes close to <110> either coalescing (by the rotation process observed in INCONEL alloy MA6000), or by a process of thermally activated unpinning. During the zone annealing of INCONEL alloy MA6000, the high recrystallization temperature permits the embryo groups of grains to approach more closely the <110> axis (Fig. 6b), thus lowering their interfacial energy further. The nucleation rate will thus be enhanced, thus accounting for the higher GAR observed in zone-annealed INCONEL alloy MA6000 than in INCONEL alloy MA760.

The abnormally growing grains are of relatively remote orientation (<110>) from those of the matrix (<100>-<112>), and their interfaces will possess an advantage in <u>mobility</u>. Nucleation has been shown to commence at the specimen surface, rather than the interior, and the curvature of the steady-state recrystallization interface (Fig. 6a) indicates that boundary mobilities are a maximum at the surface. The origin of this mobility gradient appears to be related to the strain gradient in the original extrusion, which in turn may influence the strength of the texture.

<u>Recrystallization Behaviour of Commercial Scale Extrusions</u>

The zone annealed bars of rectangular section have been crystallographically analysed. The samples were taken from the centre of the cross-section as illustrated in Fig. 10.

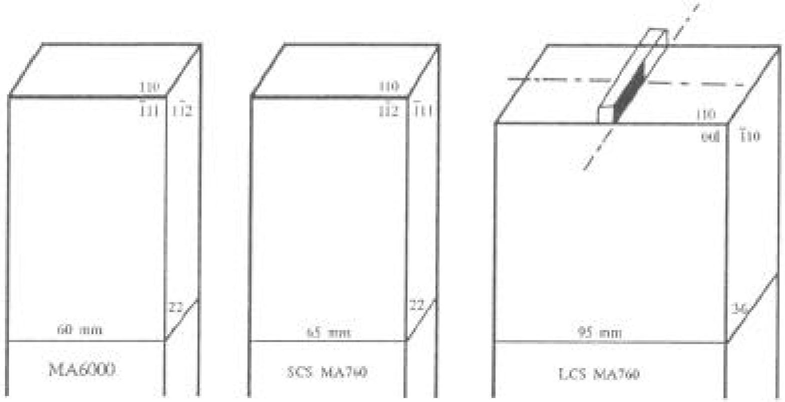



Figure 10 - Showing dimensions of commercial extrusions, and texture after zone annealing.

1. Isothermal Annealing Response of INCONEL alloy MA760. In order to assess the influence of processing variables on the propensity for secondary recrystallization, samples have been isothermally annealed both in the as-extruded condition and also after heat-treatments simulating the hot straightening applied to the extrusions prior to zone-annealing. An example is given in Fig. 11 of two series of samples which were extruded at 12 mm/s and 80 mm/s. They were isothermally annealed at 1230°C for periods between 30 sec and 5 h, and optical micrographs of the section indicated by the shaded area in Fig. 10 are shown. The left hand side of each micrograph corresponds to the external surface of the extrusion and the right hand side corresponds to the centre line. It is found that nucleation of recrystallization invariably occurred first at the specimen surface, and the recrystallization front moved towards the centre.

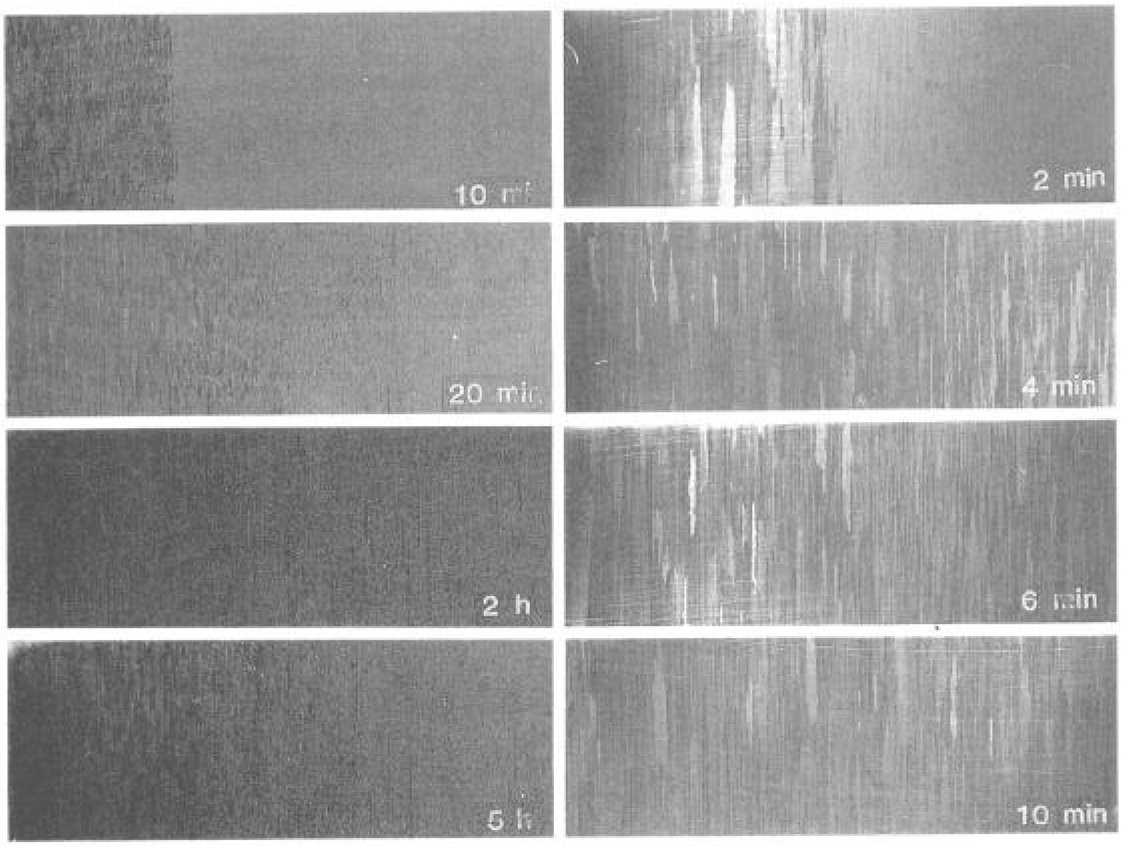



Figure 11 - Samples of LCS MA760, extruded at 12 mm/sec (LHS) and 80 mm/sec (RHS) annealed isothermally at 1230°C for the times indicated. Plane of section marked in Fig. 10.

2. Textures. SACPs revealed the orientation of each individual grain on the shaded planes in Fig. 10, and the results are marked on the sketch. In every case the orientation along the extrusion direction is ⟨110⟩, which is again in agreement with fibre texture in the experimental extrusions. Extrusion of rectangular sections has resulted in preferred orientations along the width and thickness directions, as shown in Fig. 10. The difference in texture between the SCS and LCS INCONEL alloy MA760 arises from a difference in the processing history between the samples - the LCS bar having been extruded at a 100°C higher temperature (close to the $\gamma'$ solvus) than the SCS bar. The presence of $\gamma'$ in the SCS bar is likely to change the slip characteristics of the latter material and hence the extrusion texture.

3. Grain Boundary Misorientation Distributions. The SACP technique enabled differences in grain boundary misorientations to be identified between the three specimens. All grains exhibited ⟨110⟩ vertically, and the orientations

normal to the plane of section can be summarised as follows:

INCONEL alloy MA6000: The main series of grains with <112> normals showed an angular spread of $\pm 8^\circ$. A series of narrow grains were also present across the section whose normals possessed no rational orientation relation with the principal grains. These thus represented a series of longitudinal high-angle grain boundaries running down the specimen. These have been identified previously, and have been shown to have the effect of deflecting high temperature fatigue cracks and lowering their propagation rate (3).

INCONEL alloy MA760 (SCS): The main series of grains with <111> normals showed an angular spread of only $\pm 3^\circ$. Three narrow (~ 100 μm) isolated grains were present with a <110> normal, thus representing an angular rotation of 35.16% w.r.t. the bulk of the grains. Apart from the latter grains, the texture of this bar is thus tending towards that of a single crystal, consisting as it does as an array of low-angle grain boundaries.

INCONEL alloy MA760 (LCS): This consisted almost fully of grains with <110> normals, although their individual misorientations ranged from $\pm 5^\circ$ near the surface to $\pm 10^\circ$ towards the centre. A small surface region showed a band of odd <112> grains, doubtless arising from extrusion strain heterogeneity. The texture in this case thus has a higher angular spread than the SCS bar, and higher-angle boundaries are present.

## Conclusion

1. Improved understanding of the zone annealing process has allowed the construction of a process map for satisfactory commercial production of ODS hardened nickel-base superalloys.

2. For the zoning speed studied, nucleation of secondary recrystallization occurs at a higher temperature than that at which the recrystallization interface grows along the specimen axis.

3. The matrix grains remained Zener pinned by the yttria dispersion up to the recrystallization temperature.

4. Abnormal growth of grains with <110> fibre axis occurs after thermally activated unpinning, or coalescence of groups of these grains.

5. The interfaces of the abnormal grains have a mobility advantage over the matrix grains. This advantage appears to be greater at the specimen surface than within the interior.

6. SACP has enabled the texture and the distribution of high-angle grain boundaries to be characterised in DR ODS materials manufactured to commercial dimensions.

## References

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