

Grain control in mechanically alloyed oxide dispersion strengthened MA 957 steel

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Mechanically alloyed oxide dispersion strengthened stainless steels tend to recrystallise into columnar grains, a microstructure ideal for certain creep applications. In other circumstances, equiaxed grain structures are desired. In this paper, two methods are described which have been developed to ensure the reproducible development of equiaxed or refined grain microstructures in an alloy, MA957, which has previously not been amenable to control. Grain refinement has been achieved by controlling the stored energy, so that grain boundary velocities are reduced to a level which allows nucleation to develop at many sites, and by inducing a phase transformation from ferrite to austenite.

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Introduction

Oxide dispersion strengthened alloys produced using a mechanical alloying technique have the potential for higher creep resistance and stability at elevated temperatures, compared with conventional alloys.¹⁻⁶ In the mechanical alloying process, metallic powders or intermetallic compounds are induced to form a solid solution by means of intense deformation. There are two major commercial variants of mechanical alloy: the nickel based alloys intended for aerospace applications,^{6,7} and those based on ferritic iron with potential applications at somewhat lower temperatures. The density of the ferritic alloys is about 10% lower than that of the nickel base alloys, providing a significant strength/weight advantage, and a lower thermal expansion coefficient, which is beneficial when thermal fatigue is an important design criterion.⁸ In nuclear reactor applications, the ferritic structure is more resistant to neutron damage.⁹ A mechanically alloyed ferritic stainless steel Incoloy MA 957 has been developed as a nuclear fuel cladding material for fast breeder reactors.¹⁰

After mechanical alloying and processing into bulk form, MA 957 has an ultrafine microstructure containing sub-micrometre sized grains of ferrite. The hardness in this condition is unacceptably high, so the alloy is used in the recrystallised condition. Recrystallisation can be carried out isothermally, in a temperature gradient, or using zone annealing. All these heat treatments result in a coarse columnar grain structure akin to a directionally solidified microstructure. The reason for the persistent recrystallisation into a columnar rather than an equiaxed grain structure is that the alloy contains a dispersion of oxide particles which are aligned along the extrusion direction,¹¹ so the easiest grain growth path is along that direction.

A columnar (or directionally recrystallised) grain structure is ideal for elevated temperature applications where creep resistance is important. However, when the alloy is in tubular form, its resistance to hoop stresses is found to be less than desirable. Different metal working conditions can influence the degree of anisotropy,^{12,13} but it has not yet been possible to produce an equiaxed microstructure in the range 20–40 μm . The major aim of the present work was to develop heat treatments capable of causing recrystallisation into equiaxed grains, even though the initial microstructure contains a highly anisotropic dispersion of oxide particles. The method used involves the control of stored energy before recrystallisation and exploits a recently discovered phase transformation in MA 957.

Experimental procedure

The chemical composition of the alloy used is given in Table 1. The alloy was supplied in an unrecrystallised condition. It was fabricated by charging a water cooled vertical attritor with three primary powders (elemental iron, prealloyed metallic alloys, and yttria) for mechanical alloying. Consolidation of the resultant powder was achieved by extrusion at 1000°C with the alloy packed in a mild steel can. This was followed by rolling at 1000°C, with a reduction in diameter from 54 to 9.5 mm. Although the extrusion and rolling are carried out at relatively high temperatures, they cannot be classified here as 'hot working' processes, since the final microstructure represents a cold deformed condition with submicrometre sized elongated and heavily dislocated ferrite grains.¹¹

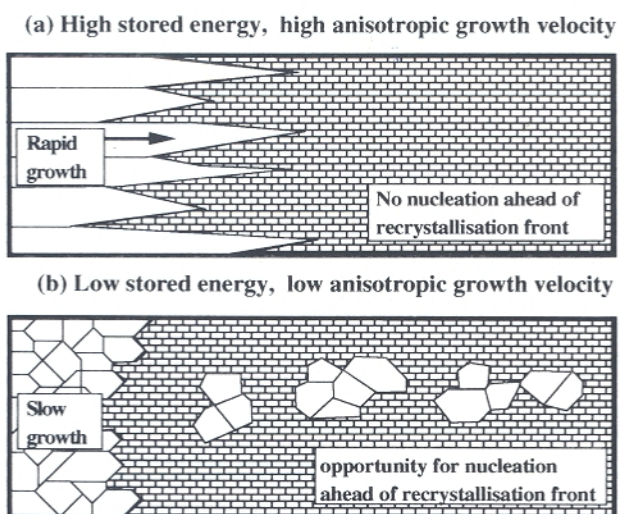
The stored energy before recrystallisation was controlled by 'preannealing' at a temperature high enough to permit recovery but not recrystallisation. Thus, preannealing was carried out at 1150°C before heating to 1350°C to induce recrystallisation.

Square sectioned specimens of dimensions 3 × 3 × 20 mm were cut from the as extruded rod. The heat treatments were carried out in a conventional resistance furnace, the samples being protected by sealing in quartz tubes filled with a partial pressure of pure argon.

Optical microscopy was used to observe the microstructures of both as received and heat treated specimens. The etchant used was 2 g CuCl₂, 40 ml HCl, and 40–80 ml ethanol. Transmission electron microscopy (TEM) was carried out using a Philips EM 400 microscope operated at 120 kV. Thin foils were prepared using a Fischione twin jet electropolisher with 5% perchloric acid, 25% glycerol, and ethanol mixture. The polishing voltage used was about 55 V.

Differential scanning calorimetry (DSC) was carried out using a Netzsch DSC 404/3/413/D, which is a specially designed high temperature, heat flux DSC with computer control and data acquisition. It uses a platinum-rhodium furnace which has very low temperature gradient characteristics. The sample and reference are placed in thermally balanced platinum crucibles. A differential signal is generated when an event causes a difference in heat evolution or heat capacity between the sample and its reference; this signal can be converted into thermodynamic data associated with the event. Experiments can be carried out to a maximum temperature of about 1500°C.

Calorimetric measurements were carried out during continuous heating (10 K min⁻¹); both the sample mass



1 Schematic development of recrystallisation process for *a* high and *b* low stored energy

and reference mass were typically ~ 200 mg. The reference was made of the same alloy as the sample, but it was in the recrystallised state. All the experiments were carried out using an argon atmosphere in the DSC chamber, the argon flowrate being $50 \text{ cm}^3 \text{ min}^{-1}$.

Thermodynamic phase stability calculations were carried out using the Mtdata package from the National Physical Laboratory.¹⁴

Preannealing experiments

It is emphasised above that MA 957 tends to recrystallise into coarse columnar grains parallel to the extrusion/rolling direction, i.e. the alloy directionally recrystallises. This is irrespective of whether the heat treatment is carried out with the sample in a temperature gradient, whether the sample is zone annealed (i.e. a moving hot zone traverses the length of the sample), or whether the sample is isothermally annealed. Indeed, the grains always tend to grow along the extrusion direction, even when the temperature gradient is orientated normal to the extrusion direction.¹¹ For all heat treatments, a jagged recrystallisation front develops and propagates along the length of the sample, presumably because the 'nucleation' occurs at the sample surfaces.

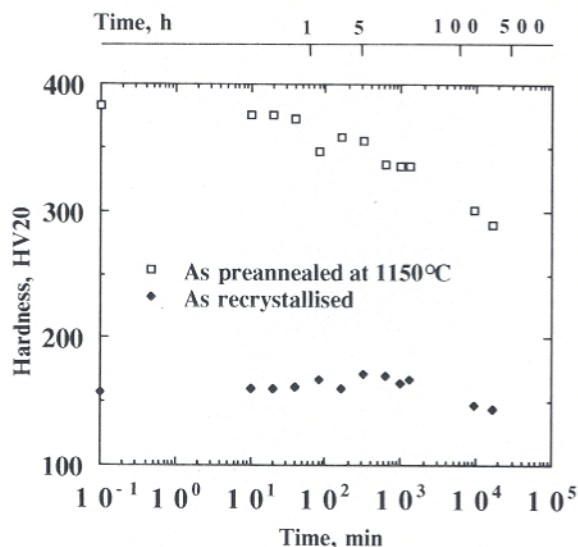
It follows that, unlike for some other alloys, the reason why recrystallisation is directional has little to do with the existence of any temperature gradients during annealing. It has been revealed using TEM that the distribution of yttria particles is not uniform;¹¹ the particles tend to align parallel to the extrusion direction. Consequently, the grain boundary velocity is anisotropic, being highest along the extrusion direction. This explains the development of the directional microstructure.

The particle dispersions are therefore not strong enough to pin grain boundaries; they simply hinder grain boundary motion, to varying degrees along different directions. In

Table 1 Chemical composition of MA 957 alloy, wt-%

C	Cr	Mo	Ti	Y ₂ O ₃	Fe
0.01	14.0	0.3	1.0	0.27	Bal.

Supplied by Inco Alloys (Hereford).



2 Vickers hardness data for preannealed samples and for samples subsequently recrystallised by heating to 1350°C

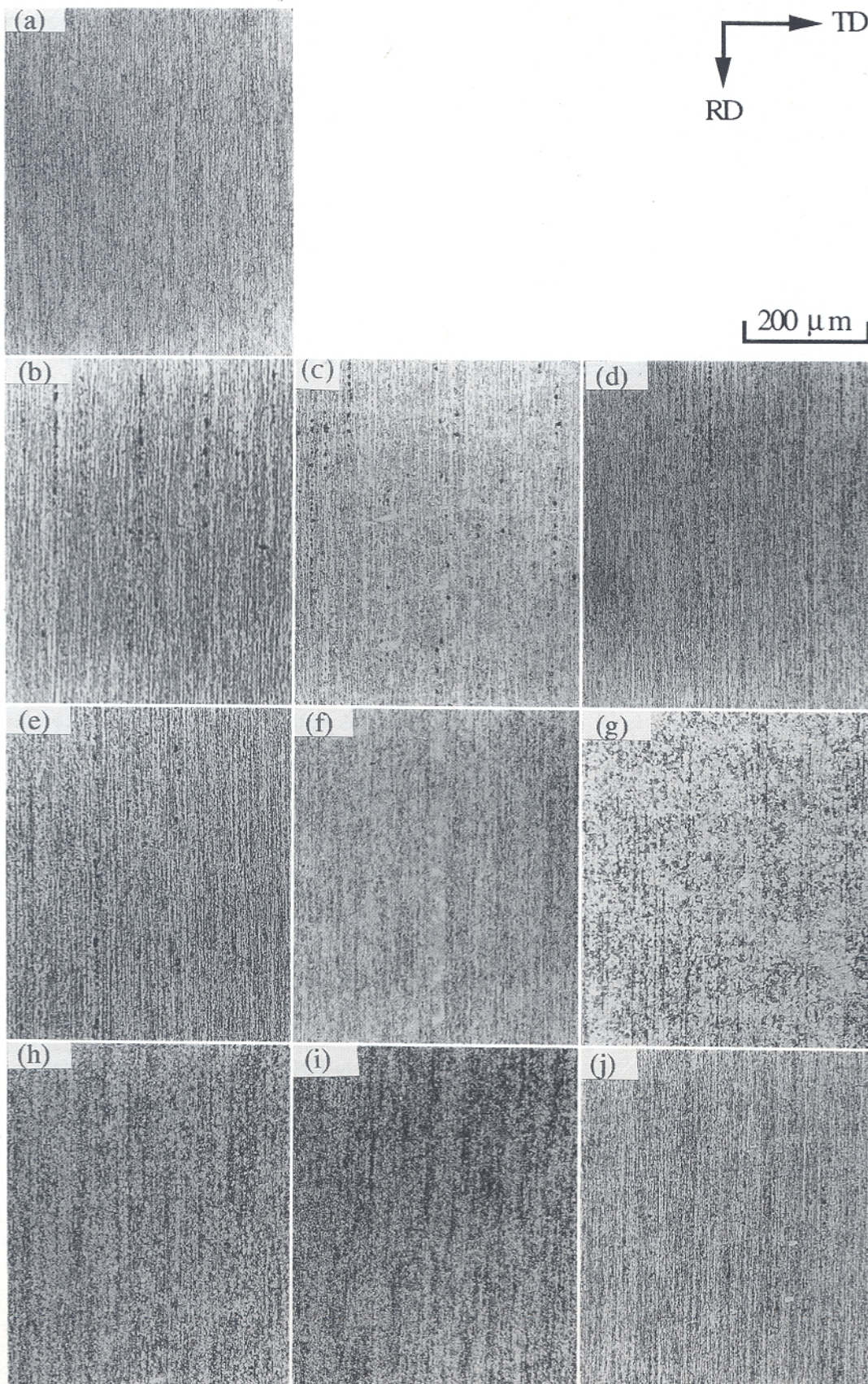
fact, the mechanical alloying and extrusion process results in a cold worked microstructure with an ultrafine, sub-micrometre sized grain structure which is very hard. Most of the stored energy of the alloy is thus in the form of grain boundaries. This stored energy is so large that moving grain boundaries can easily overcome the drag from the particle dispersion.

To summarise, the directionally recrystallised microstructure is anisotropic because the growth velocity is much higher along the extrusion direction. Once initiated at the sample surfaces, the recrystallisation front propagates so rapidly that there is no opportunity for recrystallisation to develop from other locations in the sample (Fig. 1*a*). The grain intercept along the transverse direction tends to be insensitive to the heat treatment, probably being controlled by the number of initial sites at which recrystallisation can commence. Thus, a more isotropic grain structure could in principle be produced by reducing the grain velocity along the longitudinal direction (Fig. 1*b*). This can be achieved by reducing the stored energy, via some recovery process before recrystallisation.

Recrystallisation in MA 957 tends to occur at temperatures in excess of 1300°C , so a set of preannealing experiments was designed to dissipate some of the stored energy. This involved prolonged annealing at 1150°C before heating to 1350°C to induce recrystallisation.

Although the hardness decreases slightly during preannealing at 1150°C for 10 min–80 h (Fig. 2), optical micrographs reveal no obvious structural changes (see Fig. 3). This is consistent with recovery effects occurring during the preannealing process. Subsequent recrystallisation at 1350°C in some cases produced equiaxed grain microstructures (Fig. 4), but the results were erratic. Such behaviour is a reflection of the fact that mechanical alloying is a difficult process, the alloys being somewhat inhomogeneous. Indeed, an attempt to reproduce the equiaxed microstructure shown in Fig. 4 failed, as shown in Fig. 5. However, preannealing for 160 h at 1150°C gave a reproducible equiaxed, fine grained microstructure, irrespective of the position of the sample within the extruded bar, as shown in Figs. 6*a* and 6*b*. This is because in all positions the stored energy (and hence the growth velocity) has been reduced sufficiently to give the transition from a directionally recrystallised to an equiaxed microstructure.

Consistent with this, further preannealing (280 h at 1150°C) resulted in a deterioration of the microstructure,



a as extruded; after isothermal annealing at 1150°C for *b* 10, *c* 20, *d* 40, *e* 80, *f* 160, *g* 320, *h* 640, *i* 960, *j* 1280 min

3 Optical microstructures of as extruded and preannealed MA 957 samples: no obvious structural changes can be seen