Reduction in Defect Content in ODS Alloys

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In order to develop FeCrAl-based ODS alloy tubing with the coarse, high aspect ratio, appropriately oriented grain structures likely to deliver enhanced high temperature (1100^0C) hoop creep strength compared to conventionally formed ODS alloy tubing, flow forming techniques were explored in a European funded programme. The evolution of microstructure in PM2000 alloy tubing formed by warm flow forming techniques has been the subject of continuing investigation and more detailed study in the current work.

The warm flow formed tubes investigated were produced by reverse flow forming using three, 120^0 opposed rollers described around a tube preform supported on a driven mandrel. This produced a complex pattern of shape changing deformation, driven from the outer surface of the tube preforms. The grain size and shape together with the pattern of nucleation and growth of secondary recrystallisation that developed through the thickness of the tube wall during the subsequent high temperature annealing (1380^0C) of these warm flow formed samples is described, as are the textures that formed. The unusual pattern and shape of secondary recrystallised grain structures that formed on the outer surfaces of the flow formed tubes closely follows the pattern and pitch of the flow forming rollers. The local texture, grain shape and pattern of misorientation in the surface of warm flow formed tubes that was associated with the development of these outer surface microstructures are described.

Parallel studies have continued on the influence of microstructural inhomogeneities on the development of secondary recrystallised grain structures in ODS alloys. As part of this work, a separate variant of PM2000 alloy with additions of 1 wt.% ODS-free Fe powder have been manufactured as extruded bar by Plansee GmbH. The initial recrystallisation behaviour of the variant has been studied and cross-compared with the recrystallisation behaviour found in a prototype ODS-Fe_3Al alloy, notably where the latter exhibits fine-grained stringer defects. The implications of these results for understanding and controlling grain structures in these classes of alloys are discussed.

Introduction

Commercial Fe-based mechanically alloyed (MA) ODS alloys such as PM 2000, have a composition and microstructure designed to impart creep and oxidation resistance in components operating at temperatures from ~1050ºC to 1200ºC and above. These alloys achieve their creep resistance from a combination of factors including: the dispersion of fine scale (20-50nm diameter) Y_2O_3 particles introduced during MA which, despite formation of complex oxides involving Al from solid solution, is highly stable to Ostwald ripening; and the presence of a very coarse, highly textured, high grain aspect ratio (GAR) structure which results from and is sensitive to the alloy thermomechanical processing history.[1-3]

Alloys are, typically, hot consolidated to full density using techniques such as Hot Isostatic Pressing (HIP), extrusion, upsetting or forging following which further hot and cold working (e.g. rolling, drawing etc) is used to produce the final alloy form.[4] Subsequently Fe-based ODS alloys are given a secondary recrystallisation anneal to produce very coarse grain structures for creep resistance. A wide variety of product forms can be achieved, including bar, sheet, wire, tube and foil etc.

Despite the benefits offered by the currently processed range of Fe-based ODS alloys they suffer from a number of performance shortfalls. In particular, the high GAR structures induced for creep resistance lead to anisotropic creep properties, which exhibit maximum creep resistance when the principal creep stress is aligned with the major axis of the grain structures.[5,6] But the grain structures evolved during secondary recrystallisation of currently available Fe-based ODS alloys strongly align with the principal product forming direction, which means that in a product such as conventionally HIP'd and hot extruded tube the high GAR direction is along the tube axis.[1,2,5,7] Moreover, in the Fe-based ODS alloys this alignment cannot be altered by directional thermal treatments such as zone annealing.[7] So, for Fe-based ODS alloy tubing currently available for high temperature internally pressurised applications, the direction of maximum creep strength is orthogonal to the direction of maximum principal creep stress (the hoop stress). As a result, creep life in the hoop orientation in Fe-based ODS alloy tube may be no better than 20% of that in uniaxially loaded and crept tube.[8] Moreover, pressurised tube burst data for material with current microstructures indicates a creep life (~ 14,500h / 1100^{o}C / 5.9MPa pressure) that is ~ 10% of that likely to be required for tube for application in high temperature heat exchangers (100,000h life) for power generation applications.[9]

In a recently completed BRITE Euram project the technique of flow forming was used to produce PM2000 alloy tube products with grain structures with improved grain aspect ratio in the hoop orientation.[10] The flow forming techniques applied included water cooled reverse flow forming, where a set of three idle rollers disposed at 120^{0} intervals around a tube preform mounted on a driven mandrel was used in up to 3 passes to produce thin walled tubing products containing total levels of deformation of up to ~90%. The process of flow forming induces a complex pattern of deformation involving both axial and torsional flow in the work piece. Subsequent secondary recrystallisation of these flow formed tubes resulted in evolution of grain structures that were complex and varied in size, aspect ratio and orientation both as a function of position through the tube wall thickness and with the total level of flow forming deformation applied. Achieving such high levels of deformation is not straightforward and results in extreme loading of the flow forming machinery and, in some cases, disintegration of the tube being processed. It was also noted that any hoop-orientated grain structures tended to be confined to the outer surface of the tubes. With these points in mind, it was proposed that flow forming be carried out at higher temperatures (600-700ºC) in order to lower the yield stress of the tube material and, hopefully, to increase the penetration depth of the plastic zone associated with the flow forming rollers. The results obtained from a microstructural investigation of tubes manufactured by flow forming at elevated temperature are discussed.

Also covered are the results of investigations into the defect content of the tube alloy and means by which the grain structure might be modified prior to the final stages of processing such as flow forming.

Flow forming of PM2000 tubes

Extruded PM2000 tubes were flow formed at elevated temperature (600-700ºC) and to various levels of strain then subject to secondary recrystallisation anneals. Optical micrographs of three such tubes are shown in Figure 1. The tube microstructure bears a clear relationship to the paths of the flow forming rollers with grains contained within the helical tracks left by the rollers after processing. Although the grains in all three tubes demonstrated similar degrees of axial constraint, their extent in the hoop direction was less uniform. At lower levels of deformation the grains did not extend far in the hoop direction and tended to be rather "blocky" in appearance as seen in tube W4 (74% deformation). In tube W7 (86% deformation), on the other hand, grains developed to a much greater extent in the tube hoop direction, resulting in grains with a very high GAR orientated parallel to the hoop direction. Due to the promising nature of the grain structure obtained with tube W7, further investigations were performed to determine the influence of flow forming on the development of desirable hoop grain structures in this tube.

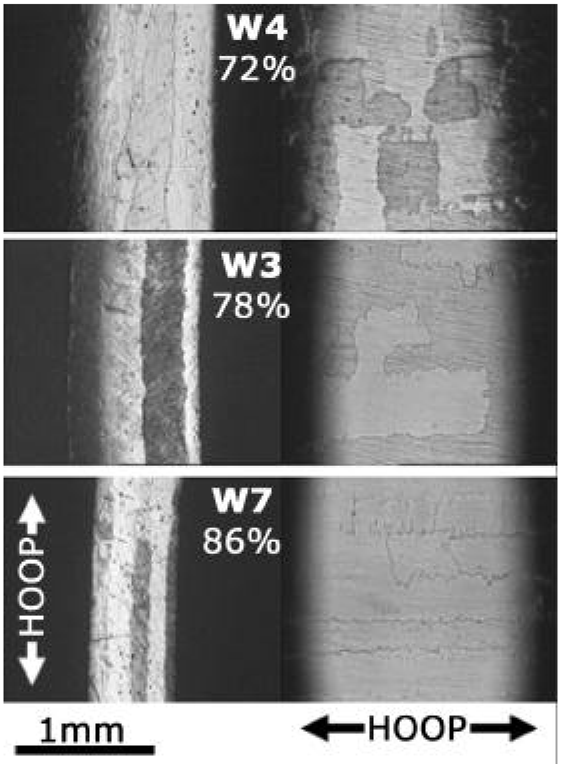
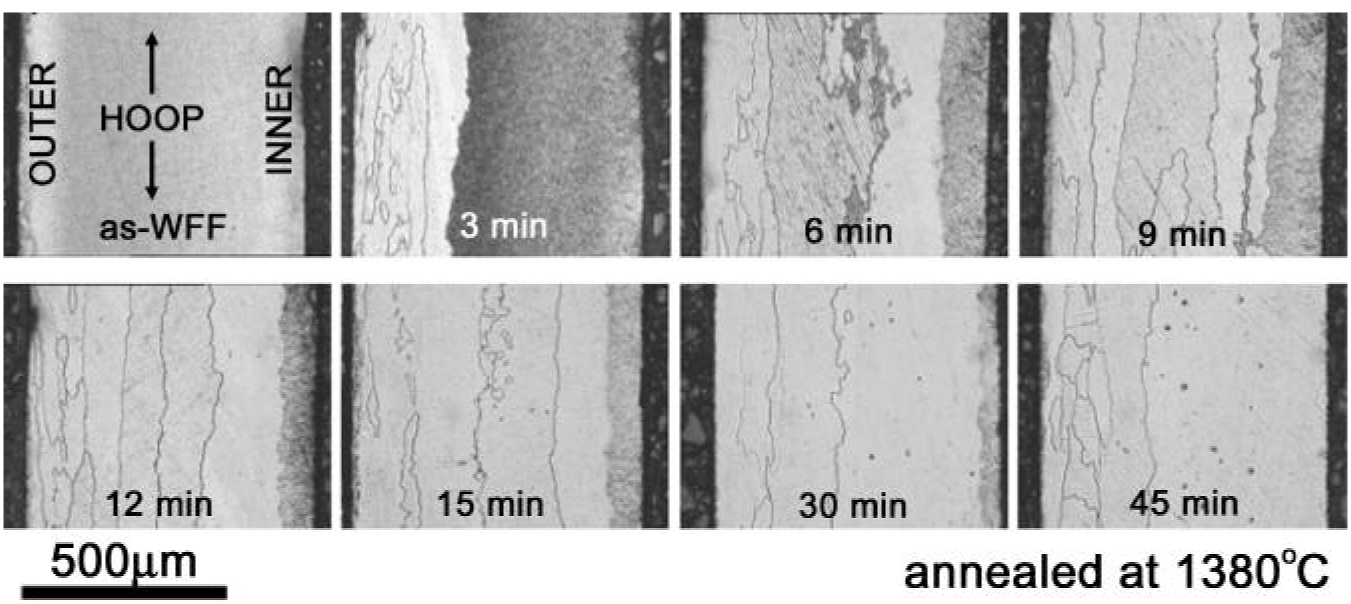


Figure 1
Optical micrographs of transverse sections and plan views of secondary recrystallised, warm flow formed tubes showing the effect of different levels of deformation on grain size and morphology.

Figure 2
Optical micrographs of transverse sections of tube W7 annealed at 1380ºC for different times. The progression of secondary recrystallisation can be seen through the tube wall thickness.



Samples cut from tube W7 were annealed at 1380ºC to effect secondary recrystallisation. A range of annealing times was chosen so that the progression of the secondary recrystallisation process could be observed. Optical micrographs of transverse sections of tube W7 annealed for progressively longer times are shown in Figure 2. It can be seen that the majority (~80%) of the tube had recrystallised after 9 minutes annealing and that recrystallisation commenced towards the tube outer surface and progressed rapidly inwards. However, subsequently, the rate of recrystallisation dropped dramatically and only after 45 minutes did the sample appear to be wholly secondary recrystallised. This is believed to be directly linked to the single-sided nature of the deformation applied during flow forming where plastic strain was highest at the outer surface and decreased towards the inner surface of the tube which was supported on a mandrel. In the earliest stages of recrystallisation, after a nominal 15 second exposure at 1380ºC, it was noted that recrystallisation initiated just below the tube outer surface at a depth of around 100μm, as can be seen in Figure 3. From this position, recrystallisation spread both outwards and inwards, reaching the outer surface within the first minute at temperature. The reasons why recrystallisation does not initiate at the outer surface of the tube are the subject of continuing investigation.

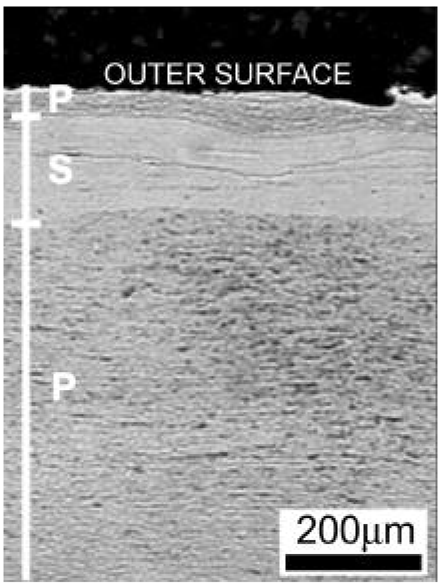
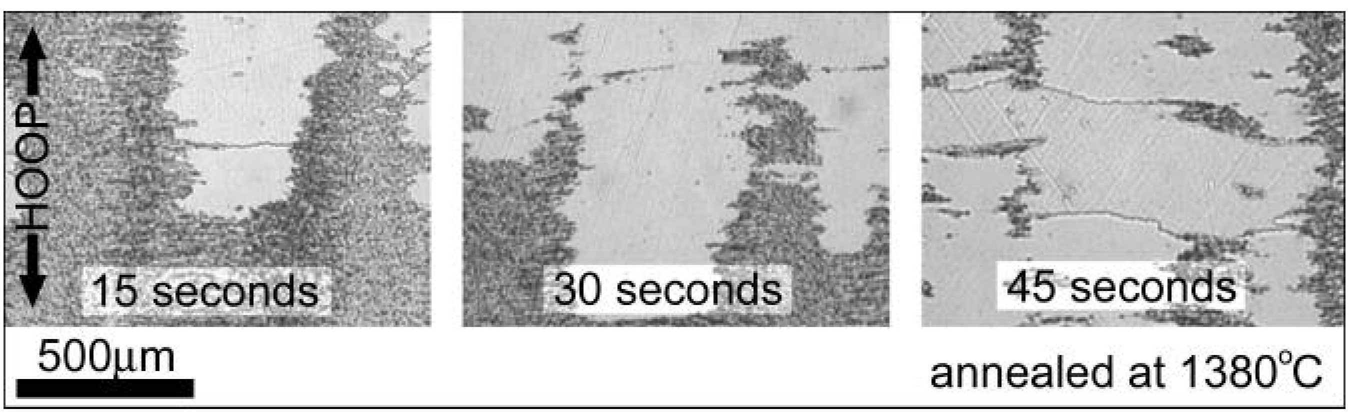


Figure 3
Optical micrograph of a longitudinal section of tube W7 annealed for 15 seconds at 1380ºC, showing that secondary recrystallisation originates from a layer just below the outer surface to spread outwards and inwards with time at temperature.
P denotes primary recrystallised alloy and **S** denotes secondary recrystallised alloy.

Figure 4
Optical micrographs of plan views of shallow taper sections of tube W7. The uneven secondary recrystallising front can be seen to be spreading downwards towards the outer surface of the tube more rapidly in some areas than in others.



Shallow taper sections of the outer surface of tube W7 were prepared so that the recrystallisation behaviour of the outer 100-200μm could be more easily observed. The progression of the secondary recrystallising front can be seen as it approached the outer surface during the first 45 seconds of annealing in Figure 4. The recrystallising front was not planar and did not intersect the tube outer surface evenly. Rather, it advanced more rapidly directly beneath the centres of the tracks left by the flow forming rollers and reached the outer surface first at these track centres. This resulted in the "comb-shaped" distribution of the primary recrystallised alloy seen in the taper sections, as primary recrystallised material extended more deeply into the tube wall below the edges of the roller tracks. The alloy directly below the track edges appeared to be less prone to secondary recrystallisation than the alloy below the track centres and forms weak barriers to the axial spread of secondary recrystallised grains at the tube surface. Secondary recrystallisation appeared subject to no such barriers in the hoop direction and grains were, therefore, free to develop a substantial hoop-orientated GAR.

The result of all these effects is that the final secondary recrystallised grain structure varied considerably through the tube wall with four (arbitrary) regions of distinct grain morphology and size. The extent of these regions is shown schematically in Figure 5 and the morphology of the grains is described in Table 1. Except for the outer hoop-orientated structure, the grains in the rest of the tube were found to be elongated, with major axis parallel to the tube axial direction. The grain size was found to increase markedly towards the inner surface of the tube, which implied much more limited nucleation of recrystallisation away from the tube outer surface. Moreover, the GAR changed with increasing grain size becoming more equiaxed in the axial-hoop plane. As mentioned, the flow forming process applies rolling forces from the outside of the tube giving a deformation gradient through the tube wall with high deformation and high nucleation rates at the tube outer surface and lower total deformation and nucleation rates at the tube inner wall.

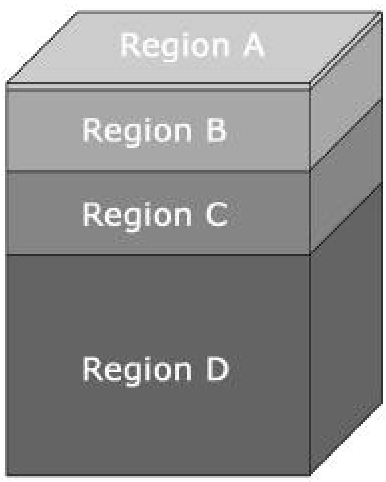


Figure 5
Schematic diagram showing the distribution of the four regions of different grain size and shape through the tube wall.

Table 1

	Region depth [μm]	Grain sizes [μm]		
		Axial	Radial	Hoop
Region A	10	600	10	50000
Region B	140	650	90	100
Region C	150	2500	150	1500
Region D	400	10000	350	7500

The shallowness of the outer layer of hoop-orientated grains is linked to the difference in recrystallisation kinetics seen at roller track centres and edges. It is likely, therefore, that the depth of deformation during forming *per se* is not sufficient to produce such grain structures. The levels or types of deformation at the centres and edges of the tracks left by the flow forming rollers was thought of at least equal significance and worthy of further investigation.

With this in mind, Electron BackScattered Diffraction (EBSD) data was collected from different areas of a taper sectioned sample of tube W7 using a CamScan X500 Crystal Probe. The sample had been annealed for (nominally) 15 seconds at 1380ºC. Data were collected from primary recrystallised material at the outer surface of the tube from positions below both the roller track centres and edges. Data were collected from immediately below the tube outer surface, from points deeper into the tube wall, and also from positions below several separate roller tracks, to check that any effects were periodic and associated with roller tracks and not simply random fluctuations. <100> pole figures and misorientation angle distribution histograms from points at the track centre and track edge and just below the tube outer surface are shown in Figure 6.

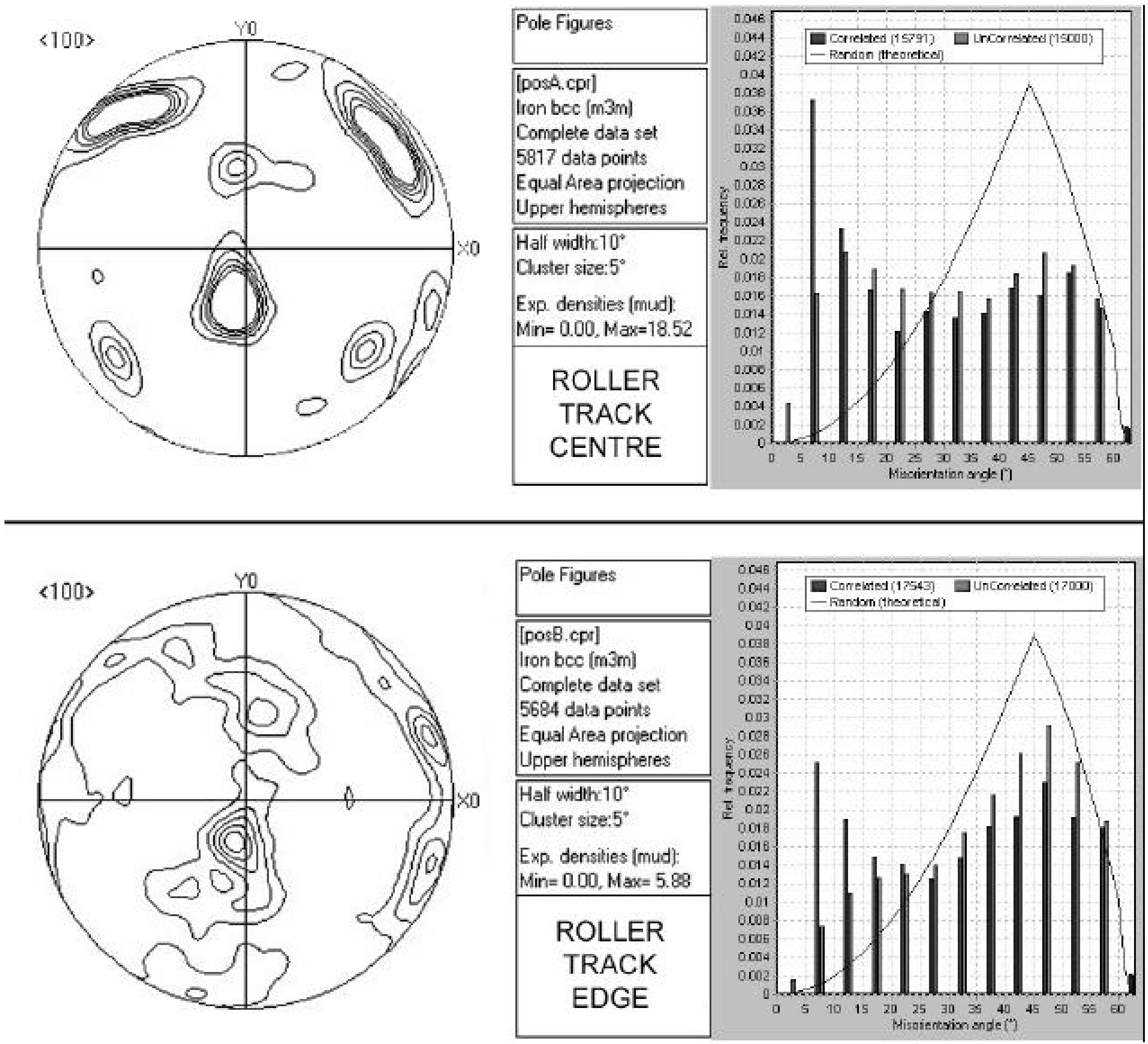


Figure 6
<100> pole figures and misorientation angle distributions from areas at the centre and edge of the flow forming roller tracks just below the outer surface of tube W7.

The plane of the page is parallel to the tapered sample surface and is at an angle of approximately 3º to a plane tangential to the tube surface. The Y direction is parallel to the tube hoop direction and the X direction parallel to the tube axis. The stronger pole figure shows a texture with <100> normal to the tube surface and <110> parallel to the tube axis and hoop directions. The deviation of the main spot from the figure centre is due to the 3º difference in tube surface and sample surface. There is evidence of some fibrous rotation around this central axis and the figure is altogether similar to those found in cold flow formed tubes.[11] There is also a weaker texture with <111> normal to the tube surface and <110> parallel to the tube axis. The pole figure from the roller track edge is much weaker and less well defined. Although there are similarities with the figure from the track centre, the primary texture is barely discernable and the secondary texture seen at the track centre is not apparent at all. The misorientation angle distribution histograms also show differences between the two areas. It can be seen that at the roller track centres there is a slightly higher concentration of low angle grain boundaries, particularly between neighbouring (Correlated) grains, whereas at the track edges the misorientation angle distribution approaches the theoretical random distribution.

It would appear that the deformation level at the surface of the tube along the centres of the flow forming roller tracks is significantly higher than that at the track edges. The texture at the track centres is stronger and there is a higher concentration of low-angle or subgrain boundaries, implying higher dislocation concentrations. This is not unreasonable as the geometry of the flow forming rollers is such that the diameter of the rollers at the leading and trailing edges is less than that at the centre-line of the roller face so that the roller bears much more heavily on the tube along the track centres than it does at the edges. It may be that the reason that the material at the track centres recrystallises so much more readily than that at the track edges relates to a greater local driving force for recrystallisation.

PM2000 variant

A variant of PM2000 rod containing up to 1 wt.% of pure Fe powder was produced by Plansee GmbH, Lechbruck, Germany. The Fe powder was chosen to introduce ODS-free regions into the PM2000 in order to investigate any effects that this might have on the recrystallisation behaviour of the alloy.

The presence of fine-grained stringers of recrystallised material has previously been observed in ODS-Fe_3Al.[12, 13] A major contributory factor credited in their formation was the presence of ODS-free regions within the ODS-Fe_3Al due to the entrainment of fragments of mill attritor in the alloy powder during mechanical alloying. A lack of oxide dispersoid might be expected to enable recrystallisation to occur and progress much more readily locally than usually seen in ODS alloys. This was found to be the case and stringers of recrystallised material have been found in consolidate, as-extruded ODS-Fe_3Al which had undergone no secondary recrystallisation annealing. A transverse section of such a stringer is shown in Figure 7 with recrystallised grains arrowed.

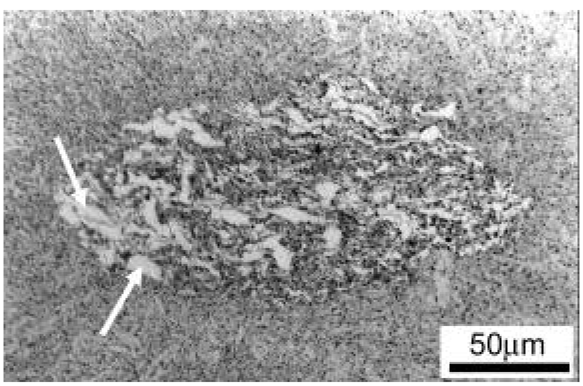


Figure 7
Stringer of recrystallised material in as-extruded ODS-Fe_3Al (PMWY2).

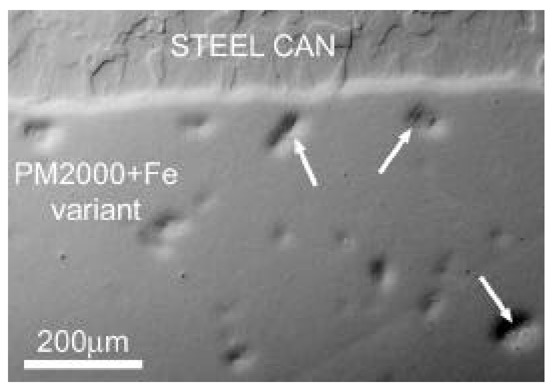


Figure 8
Recrystallised regions in the as-extruded PM2000+Fe variant.

It was suspected that the addition of Fe powder to PM2000 might produce similar recrystallisation effects. Early findings suggest that this is the case. Figure 8 shows a transverse section of the as-extruded PM2000 variant where, already, certain areas have recrystallised (arrowed). The recrystallised regions are much more uniform in size, morphology and distribution than those seen in ODS-Fe_3Al, presumably because they originate from carefully controlled additions of sized powder rather than from random fragments of mill attritor. The similarities between the fine-grained stringers found in the prototype ODS-Fe_3Al and those found in the PM2000 variant are clear in the micrographs in Figure 9. Here a transverse section of the PM2000 variant after annealing at 1380ºC for 1 hour is compared to a transverse section of ODS-Fe_3Al after secondary recrystallisation annealing (inset).

The implication is that the fine-grained stringers in ODS-Fe_3Al are introduced by ODS-free regions and this effect can be repeated in a different alloy, PM2000, by controlled introduction of ODS-free regions. The influence of these ODS free regions on the more macroscopic recrystallisation behaviour of these alloys is the subject of continuing study.

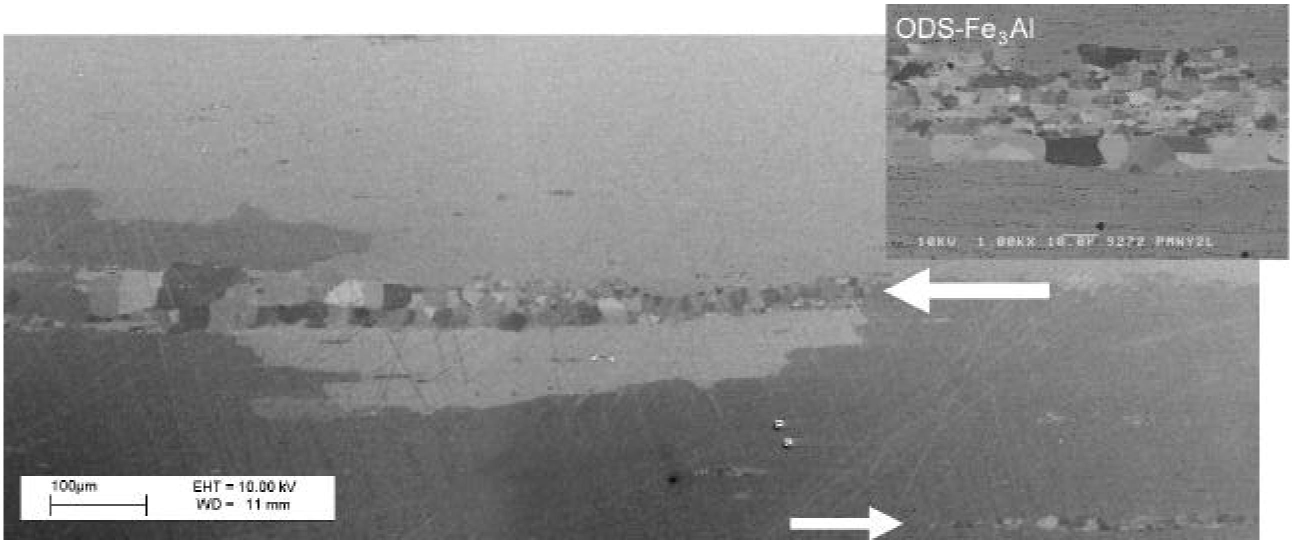


Figure 9
FEG-SEM channelling contrast image of stringers of fine grains (arrowed) in secondary recrystallised PM2000+Fe variant compared to similar fine-grained stringers found in secondary recrystallised ODS-Fe_3Al (inset). Longitudinal sections.

Conclusions

- Flow forming has been used to produce PM2000 alloy tubes with hoop-orientated grains at the outer surface and enhanced hoop creep strength.
- Grain structures formed in flow formed and secondary recrystallised PM2000 tubes are complex and are influenced by macroscopic variations in level of deformation-through-thickness.
- Variations in the level and nature of deformation at the outer surface of the tubes are spatially directly related to the tracks on the surface left by the flow forming rollers.

- A PM2000 alloy variant has been produced containing up to 1 wt.% of ODS-free Fe-powder.
- Early results suggest similarities between the recrystallisation behaviour of the ODS-free Fe variant and ODS Fe_3Al (variant PMWY2) containing fine-grained stringers.

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