Chapter 10

Barkhausen Noise in PM2000 Oxide Dispersion Strengthened Alloy

10.1 Oxide dispersion strengthened alloys

PM2000 is an oxide dispersion strengthened (ODS) alloy of composition 20 Cr, 0.5 Ti, 0.5 Y_2O_3, 5.5 Al, balance Fe (wt. %), manufactured by mechanically alloying the metallic components with fine particles of Y_2O_3 (Krautwasser *et al.*, 1994). It has been proposed as a suitable material for biomass-based power plant with an operating temperature of around 1100°C and a pressure of 15–30 bar (Capdevila *et al.*, 2001). A material for this application must have good oxidation resistance, which in PM2000 is provided by the aluminium, and good creep performance. At such a high temperature, this is only achieved using a very coarse grain size in combination with an oxide dispersion.

Powders of the components are severely deformed by ball milling to produce a mixture with a uniform distribution of oxide particles. This is consolidated by hot isostatic pressing, then extruded into tubular form. The microstructure of the material at this stage consists of very fine equiaxed grains (Sporer *et al.*, 1993). These are less than 1 μm wide and heavily strained because of the cold deformation.

Recrystallisation, to give a coarse microstructure for creep resistance, re-

quires a temperature approximately 0.9 of the absolute melting temperature T_M, as compared to around $0.6T_M$ in non-ODS iron alloys. The resulting microstructure consists of columnar grains with their long axes parallel to the extrusion direction (Elliot *et al.*, 1990; Timmins *et al.*, 1990).

A heat treatment of 90 minutes at 1380°C is sufficient to produce a fully recrystallised microstructure (Capdevila Montes, personal communication).

A TEM micrograph of oxide particles in the microstructure is shown in Figure 10.1, and Figure 10.2 gives data on the particle size distributions. It can be seen that the modal particle size is between 20 and 40 nm.

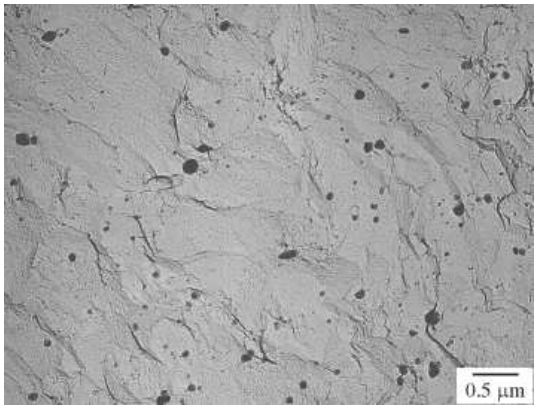


Figure 10.1: Oxide particles in PM2000 microstructure. Micrograph by C. Capdevila Montes; used with permission.

10.2 Relevance of PM2000 to magnetic property studies

It is believed that magnetic properties are influenced both by grain boundaries and by second-phase particles. In PM2000, the oxide particle distribution is not changed to any great extent by recrystallisation, so the effects of grain boundaries on BN can be isolated in this material.

In addition, by comparison with an oxide-free sample with otherwise the same composition, the effect of the particles can be studied. The oxide particles are small compared to the typical domain wall width of 80–100 μm

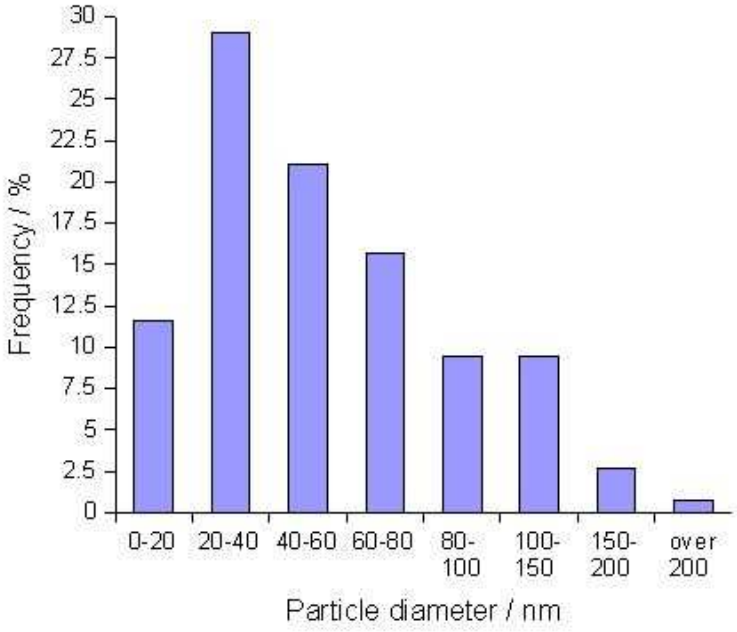


Figure 10.2: Oxide particle size distribution (Capdevila Montes, personal communication).

in ferrous materials, so it was not certain before these experiments were carried out whether there would be a detectable particle effect. However, carbides of ~ 0.2 μm strongly influenced hysteresis and BN in plain carbon steel (Lopez *et al.*, 1985; Gatelier-Rothéa *et al.*, 1992). Some oxide particles of this size exist in the PM2000.

10.3 Experimental Method

10.3.1 Sample preparation

PM2000 is supplied in tubular form, with internal and external diameters of 49 mm and 53.5 mm respectively, by Plansee GmbH. Sections were cut from the tube and heated in a furnace at 1380°C for a range of times between 10 and 90 minutes, then air-cooled. As-received samples were also retained.

Flat surfaces were prepared for BN testing and microscopy by cutting the samples parallel to the tube axis as shown in Figure 10.3, using an 'Accutom'

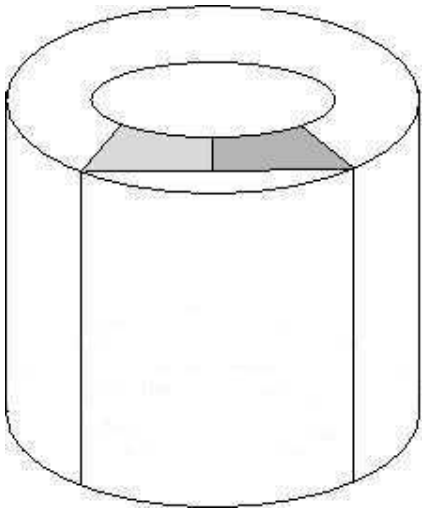


Figure 10.3: Origin of the tube-section samples, marked in grey.

rotary cutter. This geometry allows regions from the inner to the outer surface to be studied; this is important since experiments by Capdevila *et al.* (2001) demonstrated that recrystallisation began at the outer surface and moved as a front towards the inner region.

The surfaces were ground, polished to 1 μm using diamond paste and then etched with Kalling's No. 2 reagent (2 g $CuCl_2$, 40 ml HCl, and 40-80 ml ethanol). A further sample was melted to float away the oxide particles, then prepared in the same way as the others.

10.3.2 BN measurement

BN measurements were made using the same apparatus and conditions as described in Chapter 8, the only difference being the use of a different, and possibly inferior, set of ferrite pieces in the probe since these experiments were carried out earlier than those described in Chapter 8. A magnetising frequency of 4 Hz was used, except in a group of experiments to determine the effect of frequency. It was necessary to adjust the signal amplification because of the wide range of signal amplitudes from this group of samples. As mentioned in Chapter 8, however, signals obtained using different amplifications may not be directly comparable.

10.4 Microstructures

10.4.1 Naked-eye observations

The changes in microstructure on recrystallisation in PM2000 are visible even to the naked eye. Figure 10.4 shows drawings of the microstructures, with the horizontal direction parallel to the tube length. The edge of the sample closest to the outer surface is at the top in all cases. It can be seen that recrystallisation begins near the outer surface and proceeds towards the inner surface.

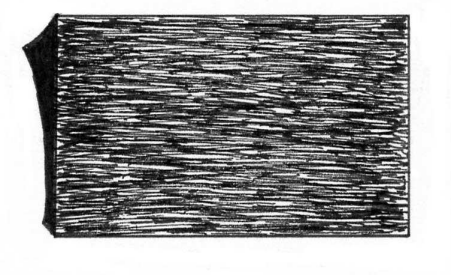
No features are visible on the unrecrystallised sample, which has a dull surface after etching, but after 10 minutes at 1380°C, a clearly visible boundary between recrystallised and unrecrystallised regions appears towards the outer edge of the sample. The recrystallised area, with a more reflective appearance, expands, and after 40 minutes at 1380°C it has occupied the entire visible area. Grain boundaries are few in number, and lie parallel to the extrusion direction. The grain width is variable but of the order of 10 mm.

10.4.2 Optical micrographs

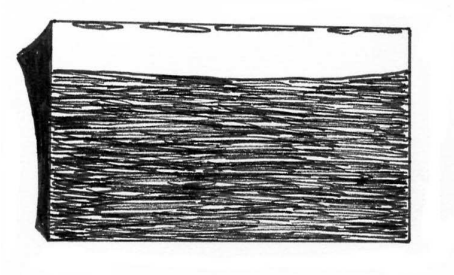
Figure 10.5 shows an unrecrystallised region (top) and a recrystallised region (bottom) in the same sample. The unrecrystallised region has striations parallel to the tube axis but no discernible individual grains. This microstructure occurs uniformly across the unrecrystallised sample. Recrystallised regions are largely featureless and have a more reflective appearance to the naked eye. All observations of partially recrystallised PM2000 showed a similar combination of fully recrystallised and completely unrecrystallised regions, with no intermediate grain growth stages visible.

10.4.3 TEM observation

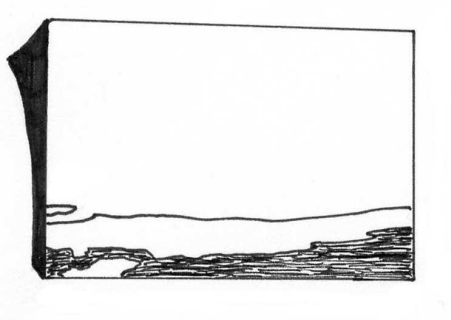
Figure 10.6 shows the microstructure of unrecrystallised PM2000 on a smaller scale. Individual grains can be resolved, but their extremely small size is evident.



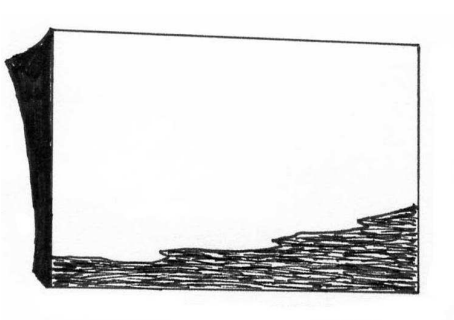
Unrecrystallised



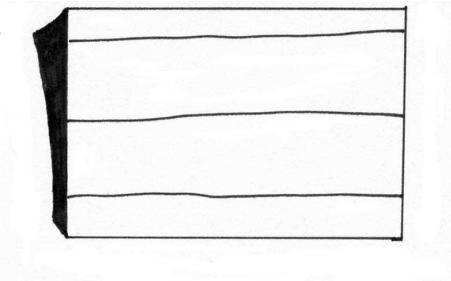
10 minutes, 1380°C



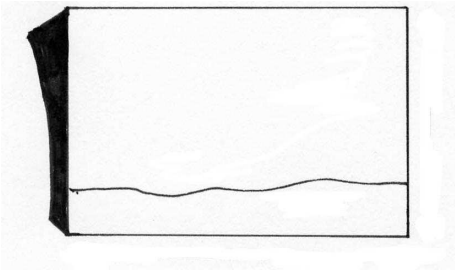
20 minutes, 1380°C



30 minutes, 1380°C



40 minutes, 1380°C



70 minutes, 1380°C

Figure 10.4: Sketches of microstructures of ODS material after different heat treatments. The tube axis is horizontal and the upper edge closer to the outer tube edge.

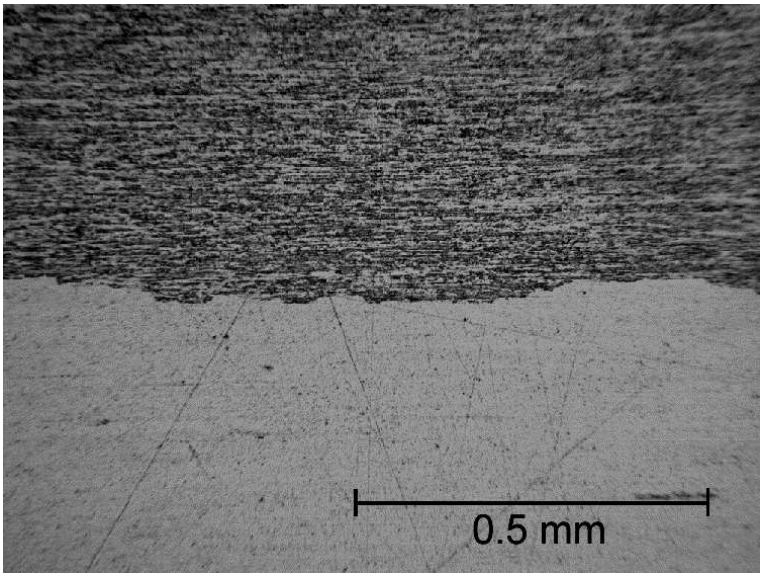


Figure 10.5: Optical micrograph showing recrystallised and unrecrystallised regions.

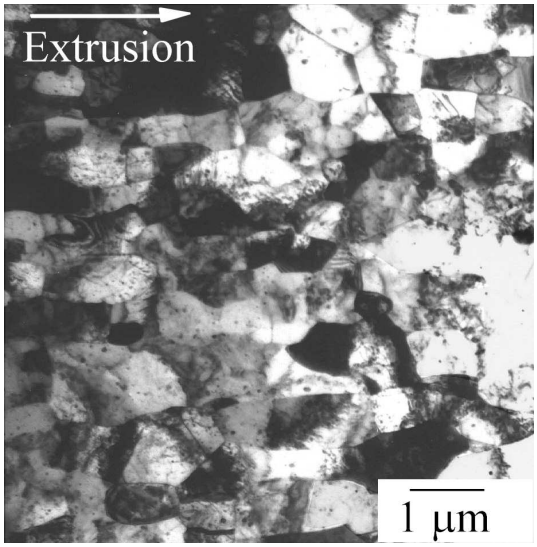


Figure 10.6: Unrecrystallised material observed using TEM. Micrograph by C. Capdevila Montes; used with permission.

10.4.4 Melted (oxide-free) sample

The melted sample shows a more conventional solidification microstructure with a variety of grain shapes and sizes and smooth, rounded grain boundaries (Figure 10.7). Naked-eye observations show that the grains are elongated perpendicular to the sample long axis, with the finest grains near the outside edges where solidification began.

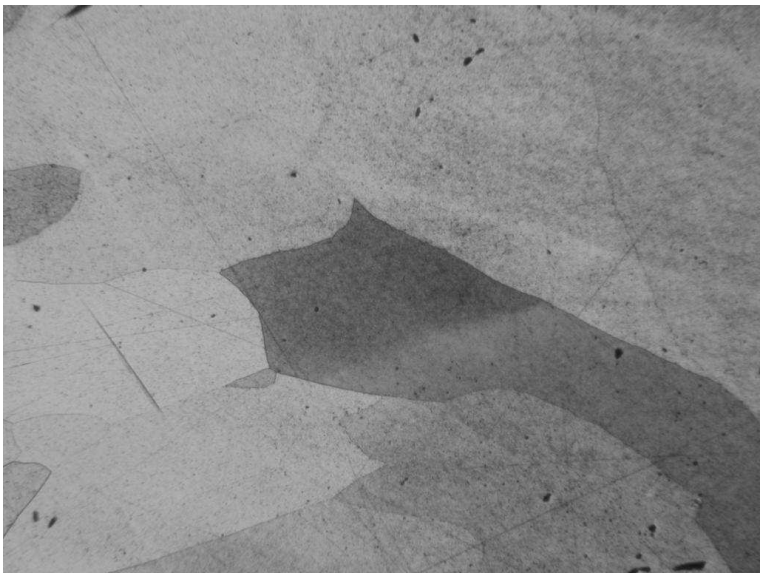


Figure 10.7: Melted, oxide-free sample.

10.5 Hardness measurements

The hardness of the samples was measured using a Vickers indenter with a mass of 30 kg and a 2/3" objective. Ten indents were made on each sample, five close to each edge parallel to the tube long axis. Care was taken to place the indent with its axes parallel and perpendicular to the tube axis so that the hardness in these directions could be measured. For each edge, the overall mean hardness and the means in the parallel (longitudinal) and perpendicular (transverse) directions were calculated (Table 10.1).

Figure 10.8 shows the difference between the mean hardnesses of the inner

Time at	Outer edge			Inner edge		
1380°C/ min	L	T	Overall	L	T	Overall
0	324	332	328	325	321	323
10	292	299	296	292	292	292
20	250	270	260	283	284	284
30	255	275	265	261	276	269
40	248	264	256	264	272	268
50	253	270	262	254	272	263
60	261	271	266	263	278	270
70	250	273	262	150	150	150
80	250	265	258	254	266	260
90	251	271	261	251	269	260

Table 10.1: Hardness (HV30) of recrystallised PM2000 samples. L=longitudinal, T=transverse.

and outer edges of the sample. Softening begins even before a large recrystallised area has formed. The outer edge hardness decreases more rapidly as the recrystallisation front moves from the outer side inwards. When both sides have recrystallised, the difference between hardness is less prominent but the outer edge is usually softer. The exception to this is the 70 minute sample, whose inner edge is anomalously soft (Table 10.1). This sample tapered more towards the inner edge than the others, perhaps giving an insufficient depth of material at the edge for correct hardness determination.

The hardness, as determined from the width of the indent, is similar when measured in transverse and longitudinal directions at short heat treatment times, but is consistently larger in the transverse direction at longer times (Figure 10.9, Figure 10.10). This can be attributed to the alignment of the strengthening oxide particles along the extrusion direction. Before recrystallisation, the fine grains and high dislocation density dominate the behaviour, but as coarsening occurs, strengthening by oxides becomes significant.

The melted sample has a mean hardness value of 211 HV30, which is softer than that of any of the oxide-containing samples, apart from the anomalous measurement discussed above.

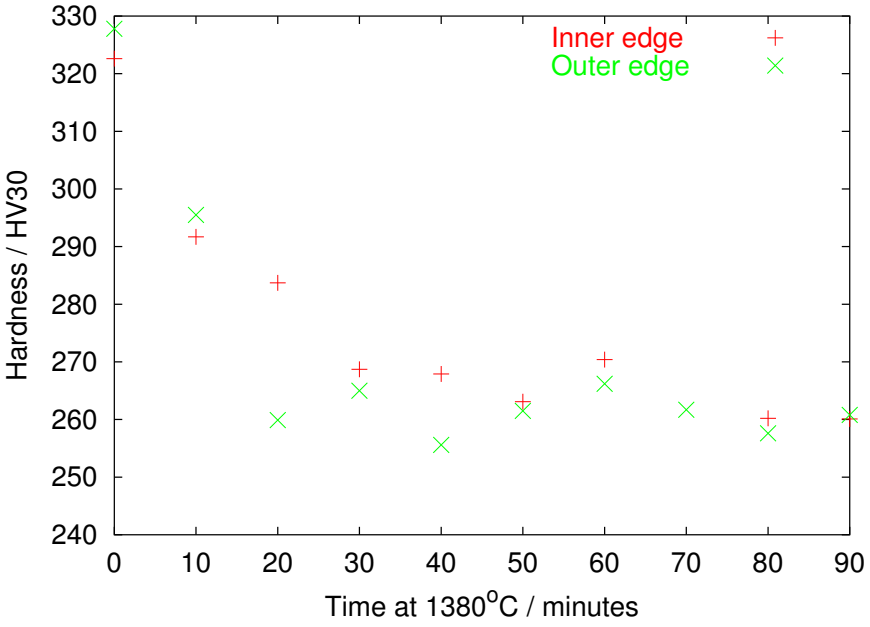


Figure 10.8: Hardness changes on heating PM2000 at 1380°C.

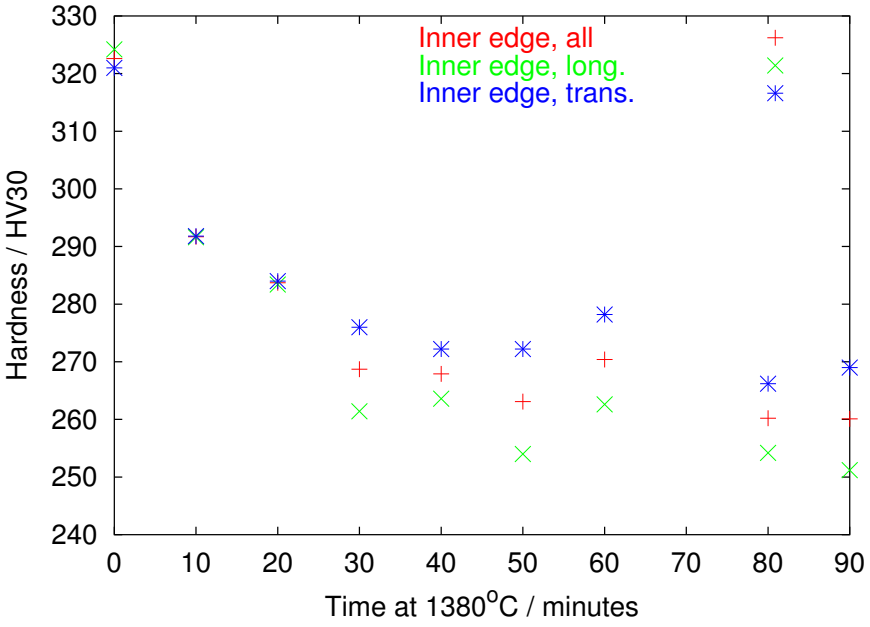


Figure 10.9: Hardness in longitudinal and transverse directions for inner edge.

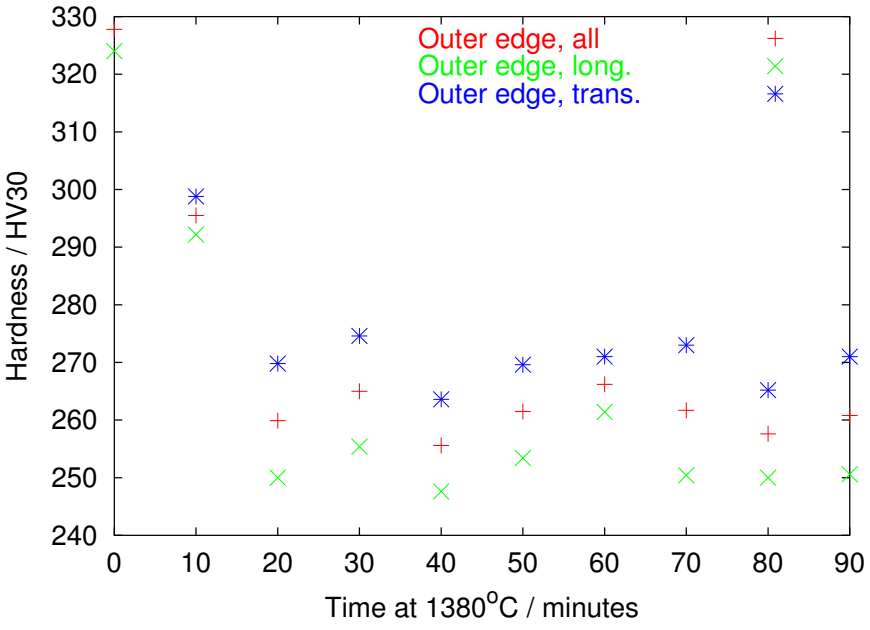


Figure 10.10: Hardness in longitudinal and transverse directions for outer edge.

10.6 Comparison between unrecrystallised, melted and recrystallised PM2000

Initially, the magnetic properties of the unrecrystallised, melted and fully recrystallised (90 minute) sample were compared.

10.6.1 Hysteresis

Hysteresis loops were measured using a VSM on cylindrical samples of length 10 mm and diameter 3 mm. The maximum available magnetising field, 8×10^5 A m^{-1}, was applied to ensure complete saturation. Figure 10.11 shows the central regions of these loops, including the coercive field and remanent magnetisation. Unrecrystallised samples have greater H_C and M_R than either recrystallised or melted samples.

The hysteresis loops in Figure 10.11 are distorted towards the edges of the plot. This effect is believed to be caused by the VSM itself, since it has been observed in other data sets acquired using the apparatus. The loop also appears to be offset on the axes, since the positive and negative H_C and M_R do not have the same magnitudes. In addition, H_C and M_R were found to depend strongly on the rate of change of applied field;[1] this should not occur at the rates used in a VSM, so it was suggested that the experiments be repeated using alternative apparatus (Moorthy, personal communication).

Figure 10.12 shows hysteresis loops measured using a more basic laboratory hysteresis unit, which allows a larger sample size than the VSM. The hysteresis is measured as a voltage per unit length of sample. The samples all had the same diameter so this is equivalent to a voltage per unit volume. The difference between the unrecrystallised and the other two samples is clear, and the recrystallised sample is more hysteretic than the melted sample. This suggests that the oxide particles do affect the ease of passage of domain walls.

[1] These observations were described in the CPGS dissertation of the present author, 2000.

10.6.2 Barkhausen noise

BN data were obtained with the magnetising field in longitudinal (L) and transverse (T) directions on the unrecrystallised and recrystallised samples, and in two arbitrary perpendicular directions on the melted sample. Figure 10.13 shows the RMS noise envelope for these samples from magnetisation at 4 Hz. In the melted and recrystallised samples, the peaks are close to $H = 0$. The unrecrystallised material has a large peak at around 0.3 A in the longitudinal direction, and much smaller peak at around 0.1 A in the transverse direction. The peak positions indicate that domain wall pinning is stronger in the unrecrystallised sample than the others, as would be expected given the high level of strain in the microstructure.

If the magnetising frequency is decreased from 4 to 2 Hz, the comparative heights of the BN peaks are changed (Figure 10.14). The unrecrystallised longitudinal peak shrinks, but the melted and recrystallised sample peaks remain in approximately the same proportion.

The change in frequency causes large differences in the frequency spectra from the unrecrystallised and recrystallised samples, but little difference for the melted material (Figure 10.15, Figure 10.16). As discussed in Chapter 8, the filter allows through frequencies between 3 and 15 kHz, and a part of the signal immediately below and above this. The melted sample noise level is approximately constant across this window, but both the recrystallised and unrecrystallised samples have a higher level of activity towards the higher-frequency end. Noise frequency is considered to be related to domain wall obstacle spacing (Saquet *et al.*, 1999); this would imply here that the spacing is very similar in recrystallised and unrecrystallised material. This, in turn, leads to the conclusion that oxide particles do act as pinning sites, since the difference in grain boundary spacing between these two samples is several orders of magnitude. However, as discussed in Chapter 8, the filtering window is narrow, and it is believed to exclude frequencies of interest, so it may be that this observation does not give a complete characterisation of the BN behaviour of this material.

10.6.3 Conclusion

This comparison demonstrates that both grain boundaries and oxide particles play a role in the magnetic behaviour of PM2000. Oxide particles affect the hysteresis properties. The inter-particle spacing, rather than the grain size, appears to be the factor controlling the noise frequency. The pinning strength is much greater when the sample is unrecrystallised, owing to the presence of high-energy grain boundaries, dislocations or both. In addition, in this sample, there is a large difference in signal amplitude when magnetising perpendicular and parallel to the tube axis. This may be due to the elongation of the grains along the extrusion direction, which gives a larger spacing between obstacles and a larger possible domain wall jump size in this direction.

10.7 BN across a grain boundary

The coarse grain structure of the recrystallised material allows a comparison of noise signals from the grain interior and across a boundary. Figure 10.17 shows the positions at which measurements were taken, and Figure 10.18 the resulting signals. The peak height is lower when the noise is measured across the grain boundary than along it or in the bulk.

10.8 Recrystallisation sequences

The BN behaviour of intermediate stages between unrecrystallised and fully recrystallised material were studied. In these experiments, a different set of ferrite pieces was used from in the experiment described above, giving some differences in the results. Measurements were made at several positions on each sample to test the influence of grain boundaries and recrystallised and unrecrystallised regions on the BN signal.

10.8.1 Unrecrystallised sample

This sample required an amplification of 70 dB to obtain a visible signal; this is in contrast to the previous set of experiments in which peaks could be seen at a much lower amplification. The reason for this may be the differences in ferrite pole piece geometry.

In Figure 10.19, two types of behaviour are observed: a smaller peak closer to zero current, and a larger peak in the higher-current range. All the transverse measurements show the former type of behaviour, but in the longitudinal direction, examples of both types can be seen. It appears that BN behaviour depends very much on position in this sample.

10.8.2 Effect of heat treatment

Figure 10.20 shows a comparison between noise signals in the unrecrystallised region and on the boundary between recrystallised and unrecrystallised material in the transverse direction. The gain used in this measurement is still 70 dB but the boundary region gives larger, more uniform, lower-field signals than the unrecrystallised area. In the longitudinal direction, too, there is a clear difference between these regions but in this case, the signals are much larger, requiring only a gain of 40 dB (Figure 10.21).

After 20 minutes of heat treatment, the gain could again be reduced, to 5 dB. The difference between longitudinal and transverse behaviour decreased (Figure 10.22). Despite the presence of a small unrecrystallised region, the positions of the peaks were much more uniform than in the samples described previously.

The longest heating time for which an unrecrystallised region was present was 30 minutes. When the signal was measured in the longitudinal direction, there was a clear difference in peak position from the recrystallised and unrecrystallised regions and the boundary (Figure 10.23).

In the samples heated for longer times than this, measurements were made within grains and across grain boundaries to test whether the presence of a boundary always affected the signal in the same way as found previously (§ 10.7). However, no consistent relationship could be found between peak

amplitudes or positions and the presence of grain boundaries in the longitudinal direction. Peaks measured along grain boundaries in the transverse direction tended to be slightly larger than those measured elsewhere. This is illustrated in Figure 10.24 for a 70 minute heat treatment, but was not observed in all the samples.

In general, the repeatability between measurements can be very poor, with large differences in peak height and position arising from neighbouring regions on the same grain. It may be that variations in oxide particle content are responsible, but this is unlikely since the volume sampled by the BN probe is very large relative to the particle volume and spacing. Much more likely is that the variations arose from the measurement technique. As noted in Chapter 8, using the equipment requires a certain amount of skill and experience, and the results are affected by the quality of grinding of the ferrite pieces. The measurements discussed here were made earlier than those discussed in Chapter 8, using an older, less well ground set of ferrite pieces, so this may have caused the lack of repeatability.

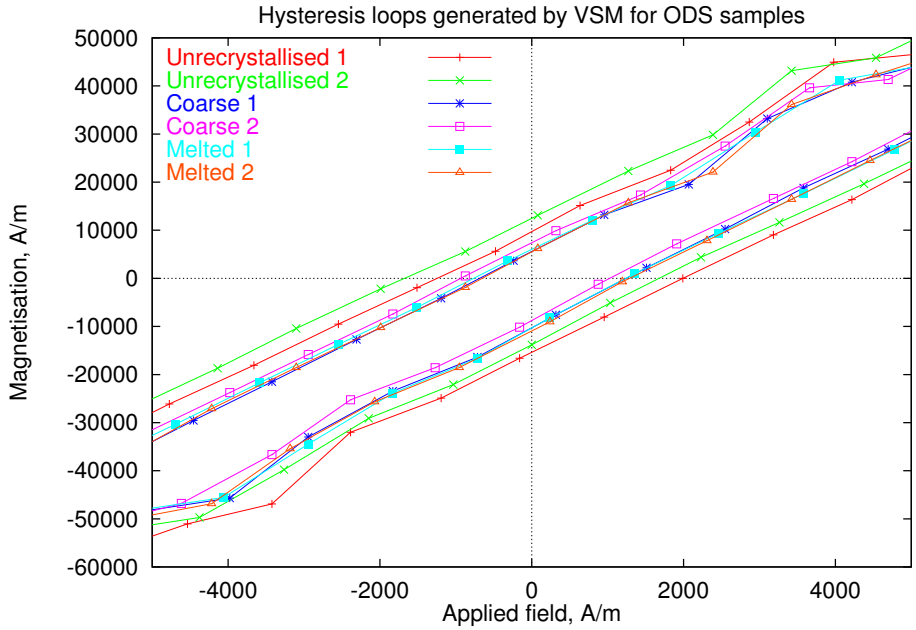


Figure 10.11: Magnetic hysteresis loops as measured by VSM.

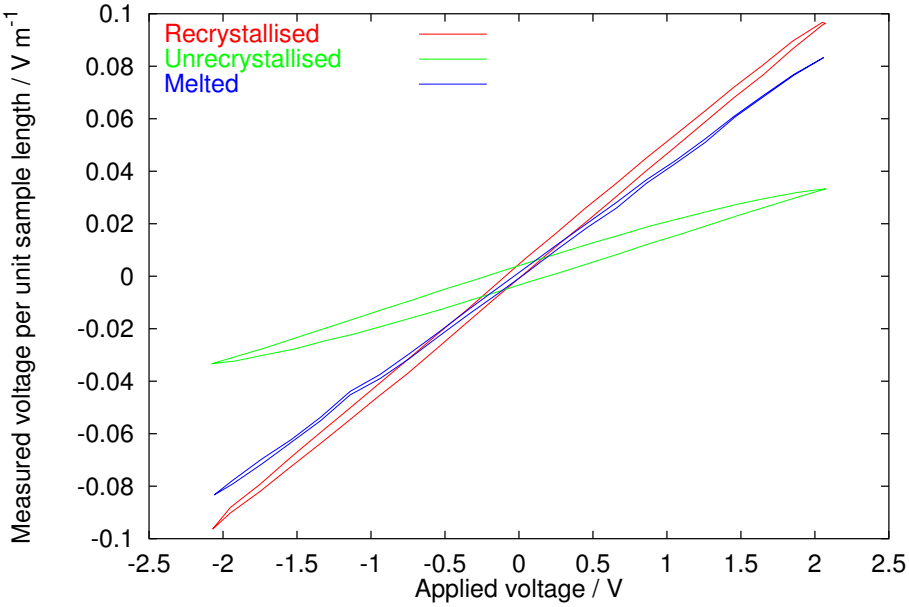


Figure 10.12: Magnetic hysteresis loops as measured by hysteresis unit.

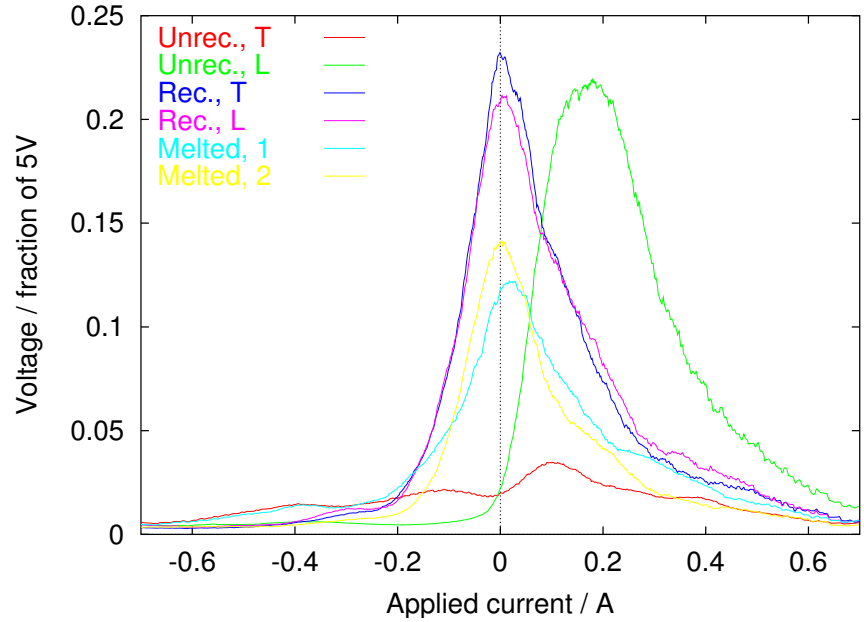


Figure 10.13: BN signals from unrecrystallised, recrystallised and melted PM2000 magnetised at 4 Hz.

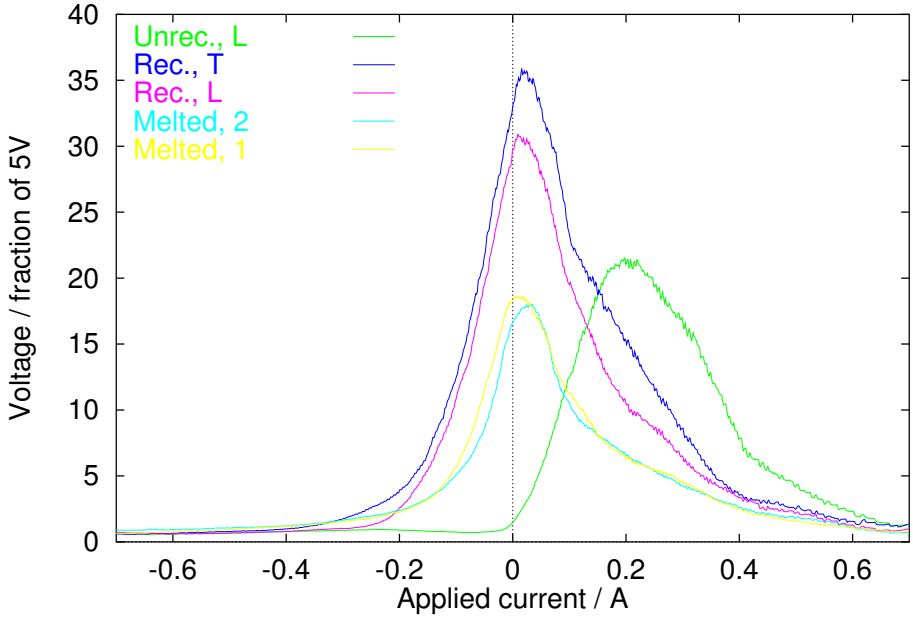


Figure 10.14: BN signals from unrecrystallised, recrystallised and melted PM2000 magnetised at 2 Hz.

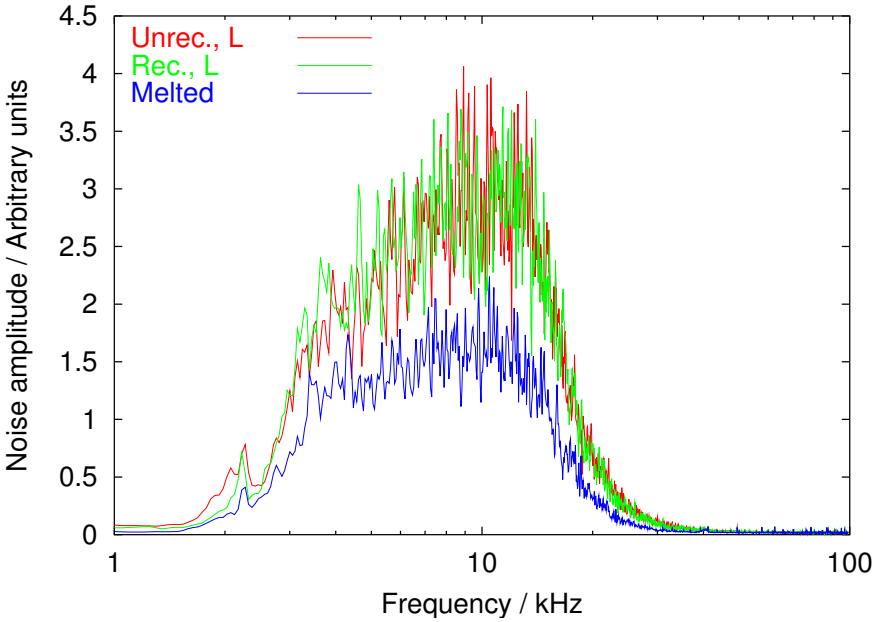


Figure 10.15: Frequency spectra from unrecrystallised, recrystallised and melted PM2000 magnetised at 4 Hz.

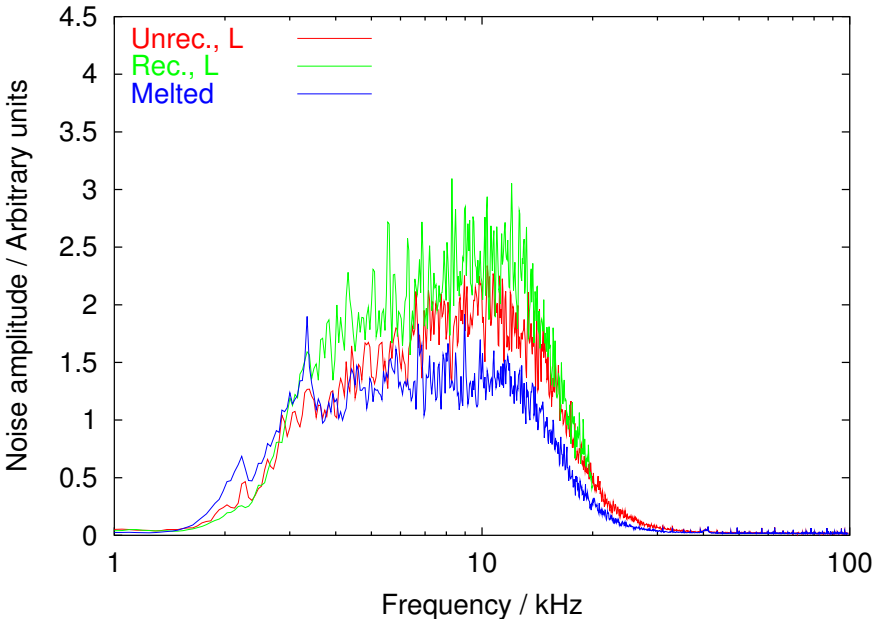


Figure 10.16: Frequency spectra from unrecrystallised, recrystallised and melted PM2000 magnetised at 2 Hz.

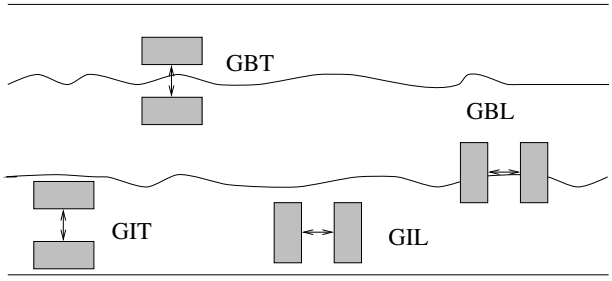


Figure 10.17: Positions in which Barkhausen signal was measured. GB, GI are grain boundary and grain interior; L and T are longitudinal and transverse.

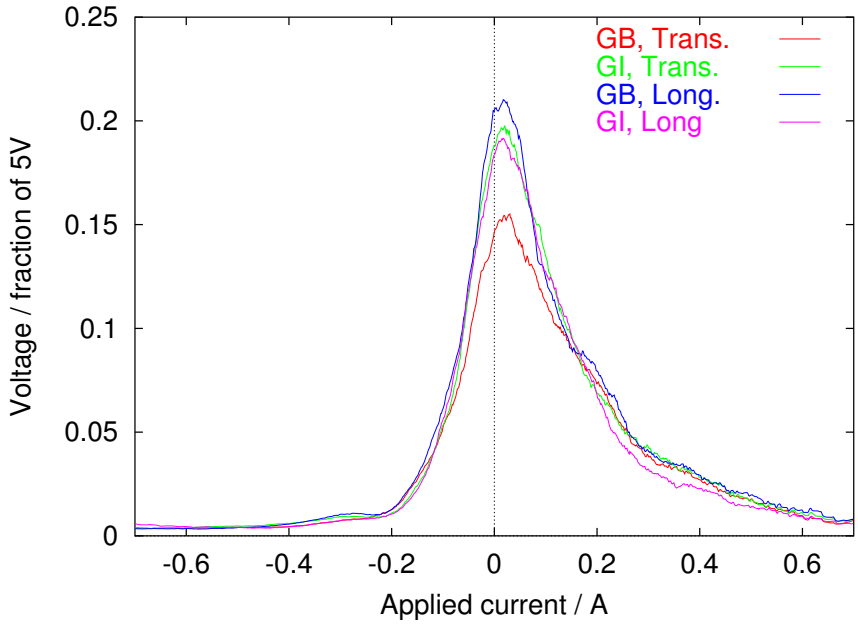


Figure 10.18: Effect on Barkhausen signal of magnetising across a grain boundary (GB) or grain interior (GI) in recrystallised PM2000.

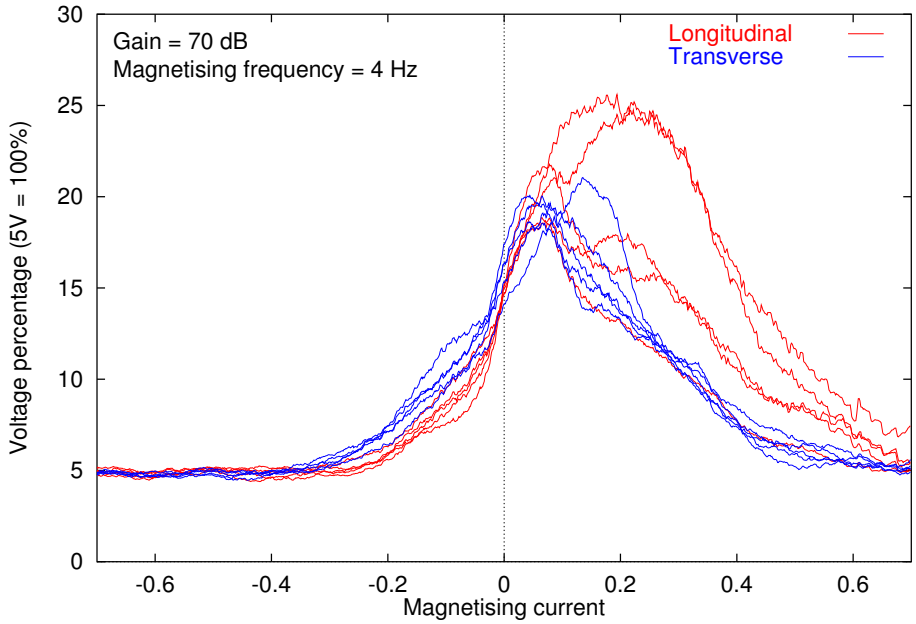


Figure 10.19: BN signal from unrecrystallised sample.

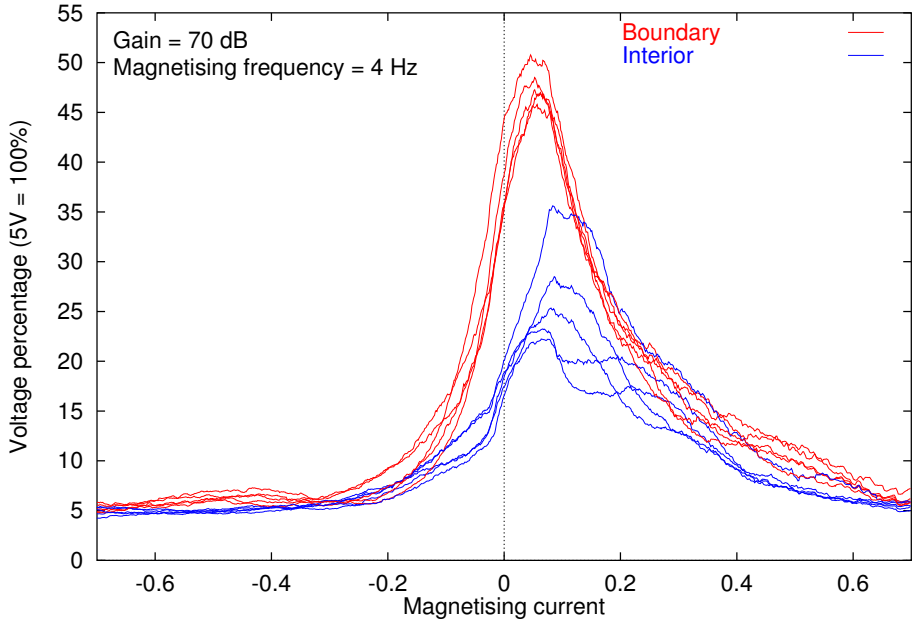


Figure 10.20: BN signals from samples heated at 1380°C for 10 minutes, measured perpendicular to tube length.

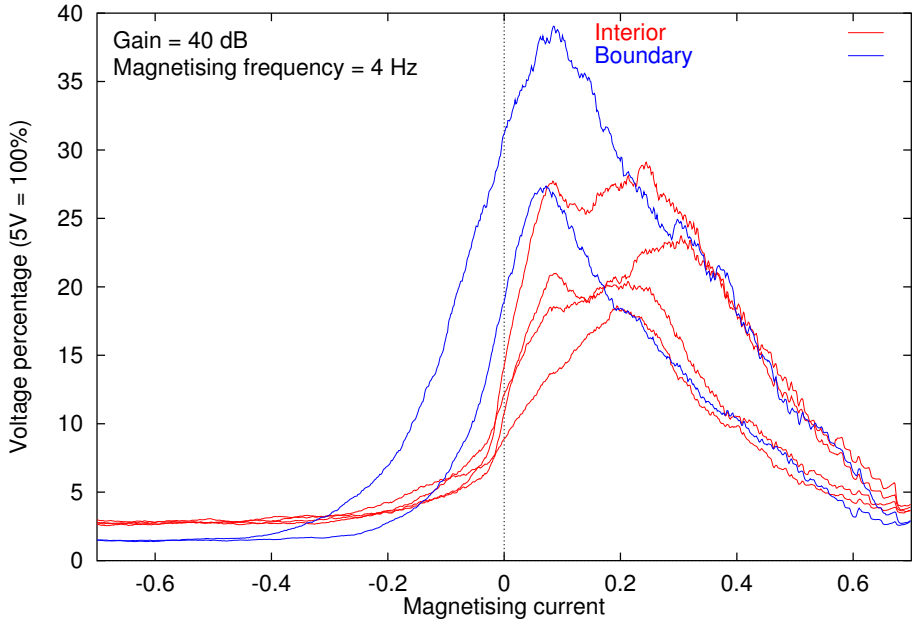


Figure 10.21: BN signals from samples heated at 1380°C for 10 minutes, measured parallel to tube length.

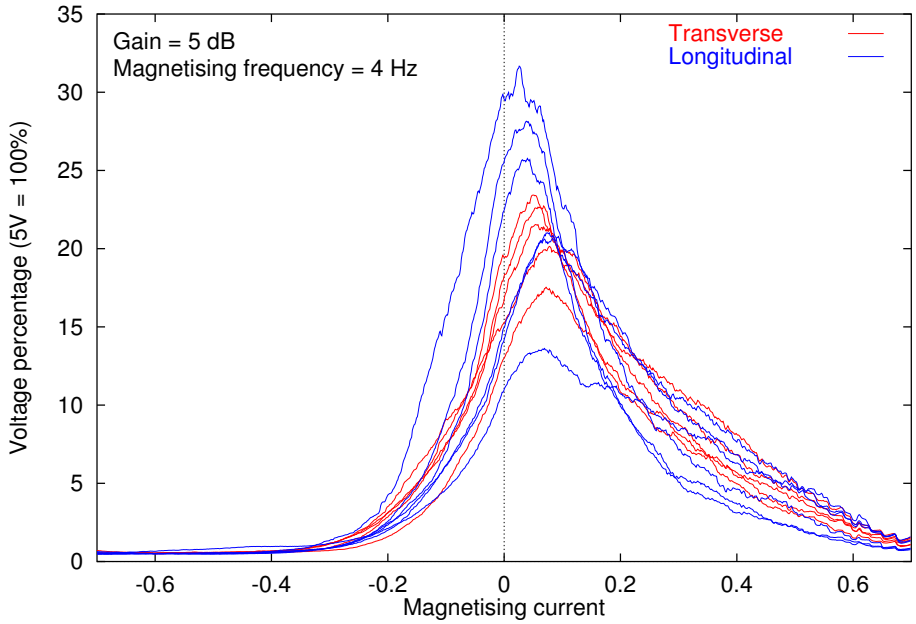


Figure 10.22: BN signals from samples heated at 1380°C for 20 minutes.

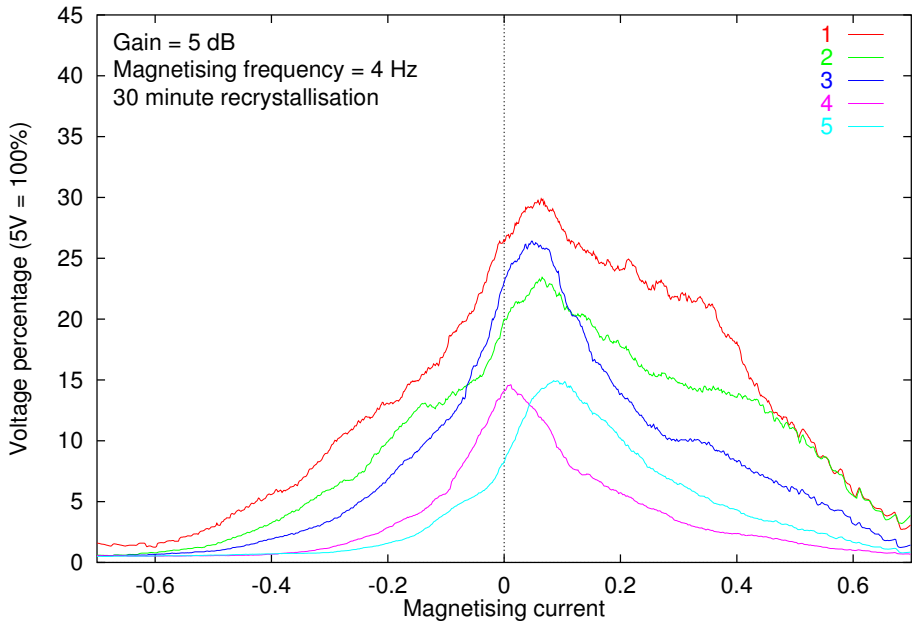


Figure 10.23: BN signals from samples heated at 1380°C for 30 minutes, measured in longitudinal direction. Line 4 was measured on the recrystallised/unrecrystallised boundary, line 5 in the unrecrystallised region, and the others in the recrystallised region.

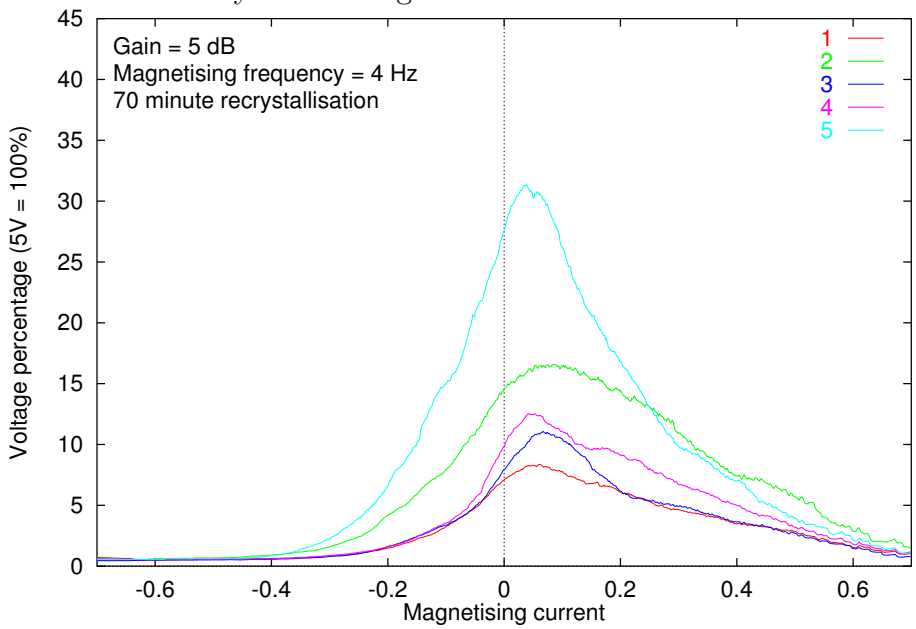


Figure 10.24: BN signals from samples heated at 1380°C for 70 minutes, measured in longitudinal direction. Lines 2 and 4 are signals measured along grain boundaries, and the others come from within the grains.

10.9 Tests on unprepared samples

Pieces of the recrystallised material in their original tube-section shape were also tested to determine whether it was possible to obtain a meaningful signal without sample preparation, since this would be useful for nondestructive testing. Because of the tube and probe geometry, it was only possible to take BN measurements in the longitudinal direction.

The outer surface was smooth, so measurements could be obtained easily, but the inner surface was irregular and posed greater difficulty in measurement. However, two or three measurements could be taken on the inner and outer surfaces of a series of samples: unrecrystallised and 20, 30 and 80 minutes.

A pronounced difference in BN signal amplitude between inner and outer surfaces was observed in the unrecrystallised, 20 and 30 minute samples. The amplification required to obtain a signal was 10 or 20 dB on the outer surface, but 70 dB on the inner surface (Figure 10.25). By contrast, the 80 minute sample requires only 10 dB amplification on both surfaces. The inner edge is more heavily deformed during extrusion, is the last part of the sample to be reached by the recrystallisation front, and is therefore highly stressed until recrystallisation is complete.

These changes are very obvious and could be used as a simple test for full recrystallisation in this material, should one be required. Examination of the actual noise signals shows that the characteristic high-field peak of the unrecrystallised material only occurs on the outer surface, and on the inside, this peak is close to an applied current of zero (Figure 10.26, Figure 10.27). The reason for this is not clear.

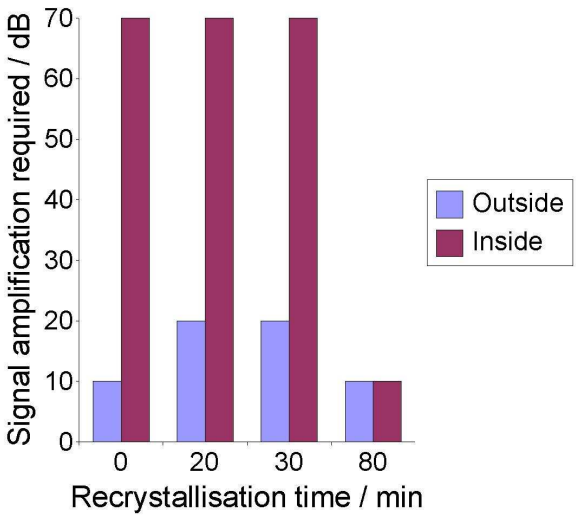


Figure 10.25: The amplifications needed for a visible signal from unprepared samples of PM2000.

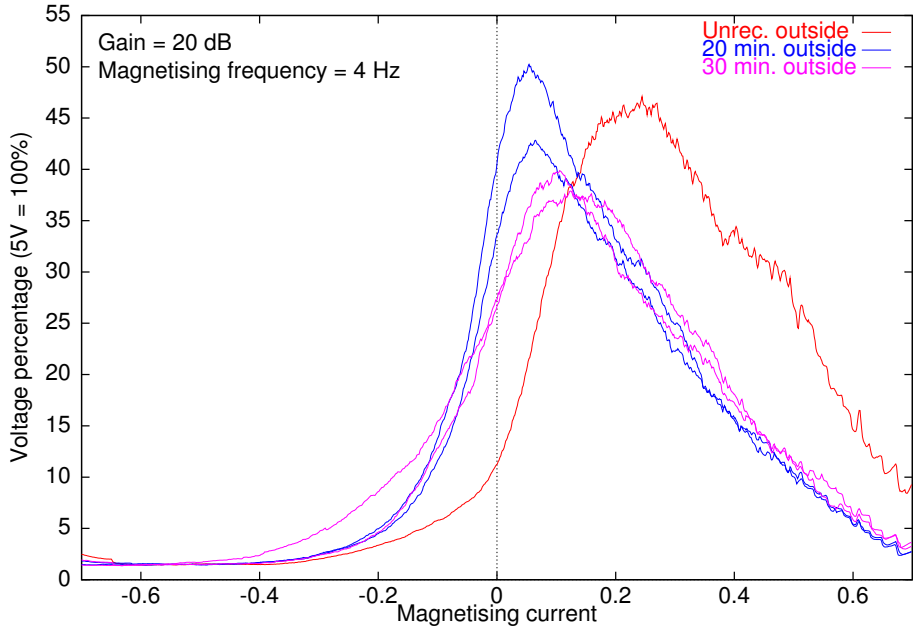


Figure 10.26: Barkhausen signal for outer surfaces of curved samples.

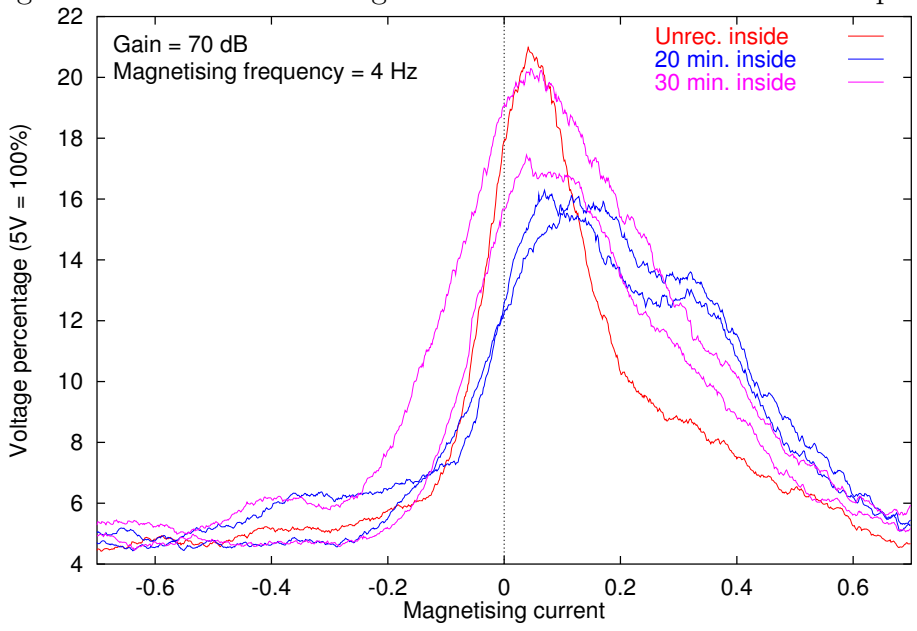


Figure 10.27: Barkhausen signal for inner surfaces of curved samples.

10.10 Conclusions

The transformation from a fine- to a coarse-grained microstructure gives a clear change in BN behaviour, even on unprepared, curved tube sections. The oxide particles in the microstructure also have an effect on magnetic properties, although this is more obviously visible in hysteresis than in BN measurements. Recrystallised and unrecrystallised regions within the same sample could also easily be discerned on the basis of their BN behaviour. A large difference between longitudinal and transverse noise signals was observed in unrecrystallised material, but this decreased on recrystallisation. The changes in BN are not directly related to the hardness, because the oxide particles influence the hardness strongly, but the BN only weakly.

The effect of grain boundaries in recrystallised PM2000 has not been clarified. Initial studies gave a clear difference between noise amplitude when measured across a grain boundary and within the bulk, but attempts to reproduce this result on other samples did not succeed. Overall, the repeatability seemed rather poor.

As mentioned in Chapter 8, the frequency filtering range used in these measurements is narrow and there is the possibility of important information going unrecorded. If possible, these experiments should be repeated using apparatus with a larger frequency range to check the conclusions.