Barkhausen and Magneto-acoustic Emission from
Ferromagnetic Materials.

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Abstract.

Barkhausen emission (B.E.) and Magneto-acoustic emission (M.A.E.) can be detected from specimens in a magnetic field varying at a few millihertz. Comparison of the two signals can indicate the nature of the domain walls responsible for the activity at any particular field. In order to characterize a specimen the strength of the emissions around the hysteresis loop are measured together with the distribution of Barkhausen event sizes.

This technique has been used to measure the effects of:

(A) Microstructure. Both B.E. and M.A.E. are sensitive to dislocations, and the effects of cold-working and its removal by isochronal annealing has been studied in alpha-iron. A simple model of domain wall pinning is presented which enables the dislocation density to be estimated. M.A.E. and B.E. are also sensitive to the growth of precipitates in Incoloy 904 alloy and, for a certain regime of sizes, can potentially be used to monitor the precipitate diameter. B.E. is sensitive to smaller precipitates (~100 nm) than M.A.E. but, unlike M.A.E., its dependence on precipitate size is not monotonic. An understanding of the signal dependence is obtained from Lorentz microscopy.

(B) Radiation damage. The sensitivity of B.E. and M.A.E. to radiation damage is quite small by virtue of the small size of defects present. Nevertheless measurements on neutron irradiated alpha-iron specimens in several microstructural states indicate: (a) an accelerated recovery from the cold-worked condition on isochronal annealing and (b) dissolution of nitrides and carbides which formed in preparatory heat treatments. Measurements on a neutron irradiated iron-copper alloy which was subsequently isochronally annealed indicated effects which were consistent with: (a) removal of dislocation loops formed during irradiation at 550°C and (b) growth of precipitates (probably copper) at 600°C which presumably formed during the irradiation, (i.e. the effect was smaller in unirradiated control specimens). These results suggest that B.E. and M.A.E. might be useful tools for the characterization of radiation effects.

(C) Tensile stress. Both B.E. and M.A.E. are sensitive to applied tensile stress and measurements on a number of different materials indicate that the dependence of M.A.E. is monotonic (except in nickel) whereas that of B.E. is generally quite complex. Since the microstructural and stress dependences are often interrelated it would be difficult to use the technique to measure say residual stress in a practical material unless the exact condition of the microstructure could be determined. Consequently B.E. and M.A.E. were measured from mild steel specimens (4360 steel) which had recieved a number of different heat treatments. The effects of applied tensile stress on the amplitude and shape of the B.E. and M.A.E. profiles were investigated with a view to be able to use the M.A.E. to measure stresses without prior knowledge of the microstructure. It was found that certain parameters in the signal profile were much more strongly dependent upon the stress than on the microstructure for many of the material conditions. Therefore M.A.E. is potentially useful for residual stress measurements.

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Chapter 1. The Barkhausen and Magneto-acoustic Emission Effects.

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1.1 Introduction.

The work presented in this thesis is a summary of the main results obtained from a three year study into the magnetic affects known as Barkhausen and Magneto-acoustic emission (B.E. and M.A.E.) which are known to occur from ferromagnetic materials. The majority of the work is experimental in nature and in general each chapter is a self contained investigation into one particular material system or phenomenon.

The Barkhausen effect has been known for over sixty years and much work has been published, but in many cases an adequate interpretation of results is lacking and there is still the need for systematic investigations using model systems. The magneto-acoustic effect, on the other hand, is a much more recently discovered phenomenon which shows potential for use in many areas, but here there is even less understanding. Both techniques have been proposed for nondestructive evaluation of microstructure and residual stress. However very few workers have used both on the same material and nearly all the recent publications have been simple r.m.s. measurements so that the field dependence of the signals is lost.

In general the successive chapters of this thesis progress to more complex experimental senarios. Besides the experimental techniques of chapter 2 the work can be divided into three main (though uneven) categories. The first is the investigation of single Barkhausen and magneto-acoustic events (chapter 3). Single Barkhausen events were observed many years ago but their acoustic analogue has not been reported to date. The second category consists of various investigations into the effects of microstructure (chapters 4, 5 and 6) and the third considers the combined influence of both tensile stress and microstructure (chapters 7 and 8). These last two categories can be further subdivided. Chapters 4 and 5 look at the influence of dislocations and precipitates respectively. In chapter 4 we consider the effect of cold work (in iron) and its subsequent removal by gradual annealing and in chapter 5 the effect of precipitates of

various sizes (in a nickel superalloy). Chapter 6 investigates the phenomena of radiation damage in alpha-iron and radiation hardening and embrittlement in an iron-copper alloy. In the latter system we have to consider the combined effects of copper precipitation and dislocation loop formation. After these studies of the influence of microstructure we move on to the final category; stress measurement. In chapter 7 the dependence of B.E. and M.A.E. upon tensile stress is measured from various simple materials. Then, in chapter 8, we have the final combination. The effect of tensile stress and microstructure in a mild steel, the aim being to be able to separate the two.

All of the investigations have a possible application (after development) for nondestructive testing. i.e. chapter 4 - plastic deformation, chapter 5 - precipitation hardening, chapter 6 - radiation hardening and embrittlement in pressure vessels, chapters 7 and 8, residual stress measurement.

1.2 The Barkhausen Effect.

On increasing an external field in a multidomain material, domains that have a direction of magnetisation closest to the external field direction will grow at the expense of other domains. This occurs by the movement of the domain walls which separate two domains. In real materials there may be obstacles which tend to pin the domain wall so that movement only occurs when the field exceeds some minimum value at which point the wall breaks free and moves to some new position of minimum local energy. These obstacles could be inclusions, dislocations, dislocation networks or other defects in the crystal structure. There is much evidence for interactions between such defects and domain walls (Degauque et al, 1978; Seeger et al, 1964; Shilling and Soffa, 1977; Astiè and Degauque, 1981). This irreversible motion of domain walls gives rise to step like increments in the magnetisation of the specimen, so that if the hysteresis loop of the specimen could be scrutinised very closely it would be seen that the curve

is not smooth but consists of a large number of very small steps (fig.1.1). At higher applied magnetic fields the domain magnetisation vector may rotate again giving rise to increments in the magnetisation of the specimen. If a sensing coil is wound around a specimen that is slowly magnetised, these individual steps in magnetisation can be detected as sharp transient pulses of voltage across the coil (provided that the rate of change of the magnetisation is large enough). i.e.

$$V(t) = -\frac{d\phi}{dt} \quad (\text{Eq}^n.1.1)$$

ϕ is the magnetic flux linking the coil.

These pulses are called Barkhausen pulses, named after their discoverer (Barkhausen, 1919), who amplified these pulses and detected them as 'clicks' with a pair of headphones.

Since the flux is proportional to the magnetic moment, then as Tyndall (1924) pointed out, the area under a single pulse is a measure of the change in the moment. He found that the change in the magnetic moment for the largest pulses in silicon-steel was 1×10^{-5} T which is equivalent to a complete reversal of magnetization in a volume of $4 \times 10^{-12} \text{ m}^3$. Preisach (1929) measured the sizes of many such pulses and found that on the steepest part of the hysteresis loop the total change in the magnetic moment was equal to that measured in the usual way with a ballistic galvanometer whilst at higher fields it was much smaller. This implied that at the higher fields the changes in magnetization were mainly from reversible events. It has been shown that in many cases the larger Barkhausen pulses are actually a 'cluster' of smaller events, indicating that a single event can induce several others (Bozorth and Dillinger, 1931; Forrer and Martak, 1932). The average size of a Barkhausen discontinuity has been calculated by Bozorth and Dillinger (1930) to be;

$$v = \frac{0.2 \cdot \ell \cdot \mu_r \cdot \bar{i}^2}{c^2 \cdot \rho \cdot M_s \cdot dB/dt} \quad (\text{Eq}^n.1.2)$$

where $\bar{i}^2$ is the mean square output current, ℓ is a length depending on the coil dimensions and the relative permeability μ_r , ρ is the resistivity and

C is a constant. They found that in iron the largest discontinuities occurred at the steep parts of the hysteresis loop where the largest hysteresis loss was. However in 50 Permalloy it was at the knees of the hysteresis loop (fig.1.2). This approach of measurement has been used by several workers to determine the average pulse size as a function of the applied magnetic field (Forster and Wetzel, 1941; Tebble et al., 1950).

1.3 Magneto-acoustic Emission.

In addition to the electromagnetic signal (Barkhausen emission) which occurs through discontinuous changes in the magnetic moment, there may be an acoustic signal. We can understand how this acoustic emission arises as follows: If the distance between magnetic moments in ferromagnetic materials is variable then we can express the interaction energy between two parallel atomic moments as an expanding multipole series (Chikazumi, 1964).

$$W(r, \cos\phi) = g(r) + l(r)(\cos^2\phi - 1/3) + q(r)(\cos^4\phi - 6/7\cos^2\phi + 3/35) + \dots$$

where r, ϕ are defined in fig.1.3.

(Eqⁿ.1.3)

The first term $g(r)$ is the exchange interaction term which is independent of the direction of magnetisation and so does not concern us here. However the second term, known as the dipole-dipole interaction term, does depend upon the direction of magnetisation and will cause deformation of the crystal lattice in the direction of magnetization, giving rise to the effect known as magnetostriction. The third term (quadrupole term) and higher order poles are usually neglected.

Thus each domain in a multidomain specimen will be strained in its direction of magnetization, the strain being either positive or negative depending on the sign of the interaction force (fig.1.4). Consequently when a domain wall moves so that the material changes its direction of magnetisation, then the magnetostrictive strain in that region will change, giving rise to the emission of elastic stress waves in the material. This is known as magneto-acoustic emission (M.A.E.). It was first reported by

Lord (1975), and it has since been observed in a number of materials. For example mild steel and nickel (Kusanagi et al., 1979a), carbon steels (Ono and Shibata, 1980), amorphous metals (Lord and Tyagi, 1982), iron-nickel and iron-silicon alloys (Kwan et al., 1985)). Since the strain is unchanged on reversal of the magnetization direction, 180° domain wall motion will not give rise to any M.A.E.. Thus, for example, very little M.A.E. is observed in hexagonal close packed pure cobalt in which the domain walls are almost entirely of 180° type (Kusanagi et al., 1979a).

1.4 Barkhausen and Magneto-acoustic Emission Signal Sensitivities.

Although both originating from domain wall motion, B.E. and M.A.E. do not give identical information. Their sensitivities depend differently on a number of factors and the two measurements frequently complement each other. The two main areas of difference are:-

- (1) M.A.E. is observed from the whole volume of the material (the attenuation being low at the frequencies of the signal) whereas B.E. is only observed from a thin surface layer because of eddy current screening causing attenuation of the signal path length (typically $\sim 10\text{--}500\ \mu\text{m}$ at 5 kHz: See section 3.1.2). Therefore it will be less prone to the peculiarities of surfaces.
- (2) M.A.E. is most sensitive to 90° wall motion and does not occur at all for 180° wall motion. Table 1.1 shows the domain wall angular dependence of the acoustic and electromagnetic signals.

Table 1.1
The relative B.E. and M.A.E. signal sensitivities
to domain wall type.

	90°	180°	θ°
B.E.	$1/2$	1	$\sin^{1/2}\theta$
M.A.E.	1	0	$\sin^2\theta$

Figure 1.1 The B-H curve demonstrating the discontinuous steps in magnetic induction.

Figure 1.2 Volume of Barkhausen discontinuities in (a) iron and (b) 50 Permalloy, in relation to the hysteresis loop.

Figure 1.3 A magnetic dipole pair.

(A) Positive Magnetostriction

e.g. Iron and Mild Steel

(B) Negative Magnetostriction

e.g. Nickel

Figure 1.4 A schematic diagram of material in a magnetic field with (a) positive magnetostriction and (b) negative magnetostriction. The dotted line represents the same material in zero magnetic field.

Chapter 2. Experimental Techniques.

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2 Experimental Techniques.

This chapter is divided into two parts. The first part describes the main pieces of equipment used for experimental measurements whilst the second outlines the experimental techniques themselves.

2.1 Apparatus.

2.1.1 The Table Mounted Solenoid.

This magnetising solenoid was in operation before starting the project, and was made by the Clarendon Physics Laboratory, University of Oxford. It consists of three coils constructed so as to give a large axial volume of uniform field. The three coils are axially aligned and electrically connected in series, the central coil being 10 cm long and the outer coils 5 cm long. The outer two coils consist of 870 turns of 16 SWG heavily enamelled copper wire in 29 layers and the inner coil of 852 turns of the same wire in 14 layers.

The solenoid has a resistance of ~ 9 ohm, with a self-inductance of ~ 0.1 Henry. The magnetic field is $\sim 8990 \text{ Am}^{-1}$ per amp, the current being monitored by a small resistor in series. The solenoid has a water cooling facility but even so the maximum safe d.c. current is ~ 5 amps.

The field homogeneity was reported to be 0.1% over the central volume of 40mm length by 30mm diameter and 1% over the central volume of 125mm length by at least 30mm diameter (Taylor 1981).

2.1.2 The Straining Machine Mounted Solenoid and Specimen Grips.

This magnetising solenoid was constructed at A.E.R.E Harwell for this project. It consists of a single coil 10 cm long mounted on an aluminium plate which is supported by two large steel pins on the Instron straining machine. The coil is made up of 2040 turns of 1.25mm enamelled copper wire in 28 layers and was designed to give a field of 0.1 Tesla with a current

of 5 amps according to the formula;

$$B = \sum_{n=1}^N \frac{\mu_0 \cdot m \cdot l \cdot I}{(l^2 + 4 \cdot R_n)^{1/2}} \quad (\text{Eq}^n.2.1)$$

where B is magnetic field

m is no. turns per meter

l is the length

I is the current

R_n is radius of each layer

N is the no. of layers

d is the wire diameter

R_i is inner radius of coil

where the R_n may be approximated by;

$$R_n = R_i + \sqrt{\frac{3}{2}} \cdot (n-1) \cdot d \quad (\text{Eq}^n.2.2)$$

The d.c. resistance needed to be < 8 ohm in order to draw sufficient current from the power amplifier. This is given by;

$$R = \sum_n 2 \cdot \pi \cdot R_n \cdot m \cdot l \cdot r \quad (\text{Eq}^n.2.3) \quad \begin{array}{l} r \text{ is the resistance per} \\ \text{meter} \end{array}$$

The solenoid had a resistance of 6.5 ohms and a self-inductance of ~0.2 Henry. The magnetic field was 15400 Am⁻¹ per amp (0.0193 Tesla per amp).

Specimen grips were designed and built by G.T.P. Engineering Ltd. (Swindon). These gripped the specimen with thread tightening collets with are fully tightened before an experiment. The lower grip was designed so that a transducer could be spring mounted on the end of the specimen. Also a bobbin was attached to the upper grip so that a detection coil could be wound around the centre of the specimen.

2.1.3 The Binary Counter and Window Device.

A 24 bit binary counter was built using analogue and TTL logic microchips in order to count the Barkhausen events. This had a capability of counting from 0 to 16,777,215 and output the reading on 24 L.E.D.s in

binary form. The pulse amplitude required to give a count was controlled by a 10 turn potentiometer (threshold level). Early measurements were made by successively adjusting the threshold control and counting all pulses larger than this value. However in evaluating an event size distribution it was necessary to subtract the successive data readings, which gave rise to large errors, and so a window device was constructed.

The window device will give a 10 μ s TTL pulse at the output for every signal pulse which has an amplitude falling between two threshold levels (fig.2.3 inset). The voltage separation between these two levels (the window), and the voltage of the lower level were independently controlled by 10 turn potentiometers. The binary counter is then used to count the number of pulses from the window counter. In this arrangement the event size distribution can be determined directly. In normal operation this was done by setting the window to a particular value throughout the experiment and then continuously incrementing the voltage of the lower threshold level after each measurement.

2.1.4 Amplifiers and Filters etc.

A number of amplifiers and filters were used throughout the project. Most of these were constructed in the laboratory and the gain and bandwidth of these are given in table 2.1. In addition to these several commercial systems were used.

In order to supply the high currents for the magnetising solenoids a 1.2kW d.c. coupled amplifier (Stereo!) was purchased from HH Electric by A.E.R.E. Harwell. This could supply 6 amps into 9 ohms (Stereo) or 10 amps into 9 ohms (mono-bridged) which was more than adequate. Also Harwell supplied the Ithaco bandpass filter (1MHz range, variable), and the amplifier (Gain 26-68dB, 3dB point 4.0MHz), which was used to filter the transducer signal.

A sweep device was constructed for use in the measurement of M.A.E. activity profiles and coercivities. This device could generate 1/4 or 1/2 a

wavelength of a triangle waveform, the output voltage varying at a linear rate from 0 to $\pm V$ or $\pm V$ to 0 (1/4 wavelength), or from $\pm V$ to $\mp V$ (1/2 wavelength). The maximum output voltage (V), and the sweep rate were independently variable.

2.1.5 The Acoustic Transducer and Charge Amplifier.

A 2.5MHz damped piezo-electric transducer (PZT) was used for all the acoustic measurements shown in this thesis. This was purchased by A.E.R.E. Harwell from Meccasonics Ltd. In the early stages of the project a point contact capacitance transducer and a G5KB piezo-electric transducer were investigated but these gave a smaller signal to noise value in the frequency range of interest (1MHz) (Measured using a Q switched Nd: YAG laser with an 850V unfocussed beam. The beam impinged onto an aluminium block with the transducer mounted on the opposite face). The Meccasonics transducer had a sensitivity of -0.1 C m^{-1} and a capacitance of 1.34 nF.

The charge amplifier used was supplied by Harwell and its frequency response was measured using a spectrum analyser with 4.9 pF in series on the input. Fig.2.1 shows the output as a function of frequency with -38.2 dBm at the input. The gain is given by;

$$G = \frac{V_{\text{out}}}{V_{\text{in}} \cdot C_s} \quad \text{where } V_{\text{in}} = 6.8 \text{mVpp} \quad (\text{Eq}^{\text{n}}.2.4)$$

$$C_s = 4.9 \text{pF}$$

2.1.6 The Integrator.

The integrator and its preamplifier was in operation before starting the project, and was designed by the Electronics Laboratory, Clarendon Physics Laboratory. The integrator was used to measure the magnetic coercivity and to plot hysteresis loops.

2.1.7 The Transient Recorder.

A Biomation 8100 transient recorder was supplied by A.E.R.E. Harwell for this project. This was a two channel 2048 bit continuous storage

recorder with arm and trigger facilities. The minimum sampling time is 0.01 μ s. The waveform stored is normally displayed on a oscilloscope after triggering but it can also be output to a Y/t plotter. This equipment was used to record single Barkhausen and magneto-acoustic events and also for calibration purposes.

2.1.8 Detection Coils.

There were two existing similar coaxial coils mounted in the table mounted solenoid. These were wound by Taylor (1981) for magnetic measurements. Each coil was 10mm long and had an internal diameter of 9.3mm and consisted of 3450 ± 50 turns of 40 SWG enamelled copper wire wound in opposite senses on a former. The spacing between the centres of coils was 50mm. These were situated in the centre of the solenoid. These coils are electrically connected in series so that the e.m.f. induced across one coil due to external fields would exactly cancel that induced across the other coil. If then the specimen is situated symmetrically within one of the coils the e.m.f. induced across the two coils is proportional to the rate of change of the magnetic moment. The resistance of the two coils was 390 ohms.

A single detection coil of length 10mm and internal diameter 2.8mm was wound for the straining machine mounted solenoid. Two coils were not used as it was not possible to wind a second coil of the same dimensions in the space available. However the magnetisation rates used were ~ 5 Hz and so only a small d.c. signal was detected across the coil from this, and this was easily removed by filtering. The coil consisted of 3200 turns of 40 SWG enamelled copper wire and had a resistance of 153 ohms.

2.2 The Barkhausen Signals.

2.2.1 Single Barkhausen Events.

The experimental arrangement used to observe individual Barkhausen events is shown in fig.2.2(a). The function generator is set to give a very low frequency ($<1\text{mHz}$) triangle waveform output, which is low pass filtered (3dB at 27mHz) in order to reduce possible noise. This is amplified by the 1.2kW power amplifier which drives the water cooled magnetising solenoid. The specimen is placed in a uniform field in the centre of this solenoid and wound around the centre of the specimen is one of the detection coils. The second similar coil is wound in the opposite sense below the specimen so that the voltage induced across the two coils in series is proportional to the rate of change of magnetisation of the specimen. The slowly varying external magnetic field will give rise to irreversible domain wall movements which induce transient voltage spikes across these coils. These Barkhausen pulses are then filtered and amplified using the same equipment as for the Barkhausen event size distribution measurement (section 2.2.2), in order to be able to compare the waveforms with theoretically derived data. (The measured frequency response of this equipment was used for the theoretical calculations). The bandwidth chosen was 360–100,000 Hz, the low frequency cut off removes the large d.c. signal arising from events which occur from deep within the material (the presence of the low frequency signal makes measurements of single events very difficult), and the high frequency cutoff removes radio-frequency noise. It was necessary to attenuate the signal ($\times 0.05$) before storing it on the Biomation recorder set on pre-trigger mode. A hard copy of any Barkhausen event recorded could be output onto a Y/t plotter.

2.2.2 Barkhausen Event Size Distributions.

The experimental arrangement used to measure Barkhausen event size distributions is shown in fig.2.3(a). The Barkhausen events are detected,

filtered and amplified in the same way as in section 2.2.1. The signal is then input into the window device set with a 100mV window with a variable offset. The number of Barkhausen events with peak heights within the window range is then counted on the binary counter. In normal operation the number of Barkhausen events as a function of peak height (the Barkhausen event size distribution) are counted over a particular field range. In order to do this the window offset voltage was successively incremented by 100mV before measuring the count over the field range of interest. i.e. the offset is successively set for each count, the offset settings being given by $100n \text{ mV}$, where $n=1,2,3,\dots,99$. Then for each setting a count is taken over the field range of interest so that a distribution is built up with 99 readings ranging from 100mV to 10V.

A typical Barkhausen event size distribution is shown in fig.2.4(a) for a cold-worked pure nickel specimen (Johnson-Matthey Spec-pure). In this case the Barkhausen events were counted over the whole field range where Barkhausen emission was observed to occur. The horizontal axis is the amplitude of the Barkhausen event (window threshold) and the vertical axis the logarithm (base 10) of the event count. The number of events is observed to be a strong function of their size, the count decreasing from 266,000 to ~ 2 over the 10V range. Fig.2.4(b) shows the same data plotted on a $\log_{10}$ - $\log_{10}$ scale. This produces a fairly good straight line and so a straight line fit is made. The distribution can then be defined by just two parameters, the total Barkhausen count and the value of the threshold intercept for a single count (that is a measure of the spread of the distribution). These two parameters are consistently used throughout the experimental sections. The single count threshold intercept has more significance than the intercept with the other axis; it describes the largest event likely to be detected in the experiment, probably occurring near the surface and therefore unattenuated by the metal. It could be reasoned that the slope of the logarithmic plot is a better parameter to use than the total Barkhausen event count since the latter is very

dependent upon the sensitivity limit of the equipment. However carrying out a straight line fit on a logarithmic plot of the data was a late progression in the work and so the slope parameter was not generally used.

2.2.3 Barkhausen Emission as a Function of Applied Magnetic Field.

(Barkhausen Emission Activity Profiles).

A schematic diagram for the experiment is shown in fig.2.3(b). The experimental arrangement is similar to that in the previous sections, but now the function generator is set to a slightly higher frequency (typically 5mHz). The activity profile or envelope is then obtained by rectifying and passing the signal through a 2.2 Hz or 300mHz low pass filter and plotted as a function of the current through the magnet on an X/Y plotter.

Fig.2.5 shows a typical profile obtained from the same pure nickel specimen as in section 2.2.2. Most of the activity occurs as a single peak at low fields in the coercive region.

2.3 The Magneto-acoustic Signals.

2.3.1 Single Magneto-acoustic Events.

A schematic diagram for the experiment is shown in fig.2.2(b). The function generator was set to a triangle wave output at low frequencies ($<200\mu\text{Hz}$). The frequency required to separate events is lower than for Barkhausen emission because events can be detected from throughout the volume of the material and also the transient response from a single event is generally longer. The acoustic emission which occurs as the external field is slowly varied was detected by the 2.5MHz damped piezo-electric transducer, and then amplified using a charge amplifier (Gain= 0.13V pC^{-1}). The transducer was mechanically attached to the end of the specimen, using Ultragel acoustic coupling fluid. The acoustic signal was further amplified by 50dB and filtered with a bandpass of 6.3 to 63 kHz. The magneto-acoustic

event was then stored on the Biomation recorder set on pre-trigger mode. A hard copy of any magneto-acoustic event stored could then be recorded on a Y/t plotter.

In order to test the acoustic coupling the pencil lead breaking technique was used. The amplifier gain is adjusted to 26dB and a given length of 0.5mm pencil lead (3H) is broken near the specimen (In this case, on the top of the upper specimen support rod). The impulse derived from such a break is repeatable to within ~10% (Hsu et al., 1977). The coupling is proportional to the amplitude of the first peak in the waveform (fig.2.6)

2.3.2 Magneto-acoustic Emission as a Function of Applied Magnetic Field (Magneto-acoustic Emission Activity Profiles). Method 1: Direct Signal.

A schematic diagram for the M.A.E. activity profile measurement (method 1) is shown in fig.2.7(a). The power amplifier drives the magnetising solenoid as for the B.E. measurements but with the function generator set to a 0.15Hz triangle waveform. The M.A.E. signal is detected by the piezo-electric transducer and amplified and filtered as for the single event measurement (section 3.2.1) but with a bandwidth of 32-100 kHz. A slightly higher frequency low frequency cut-off was used because, although there is some M.A.E. energy below this, the audio frequency noise needs to be filtered out as this varies over the time scale of the measurement (for single acoustic event measurement wait for a quiet day!). The upper frequency limit was increased in order to include higher frequency M.A.E. components. A full wave rectifier and 2.2 Hz filter are then used to obtain the signal envelope which is plotted as a function of the magnetising current on an X/Y plotter. The M.A.E. activity profile obtained is then directly comparable to the B.E. activity profile.

Fig.2.8 shows an activity profile obtained from the same pure nickel specimen as in section 2.2.2. (The activity profiles for both halves of the hysteresis loop are superimposed). The activity profile for a higher

bandwidth (100-1,000kHz), is also shown, indicating that there is some M.A.E. activity in the higher frequency range. The profiles again have a single peak in the activity which occurs at low fields. In order to measure the true peak amplitude in the activity the noise needs to be subtracted, i.e.

$$V_{\text{true}} = [(V_{\text{observed}})^2 - V_{\text{noise}}^2]^{1/2} \quad (\text{Eq}^n.2.5)$$

where V_{observed} is the peak voltage measured from the trace.

The M.A.E. signal obtained before deriving the signal envelope is shown in figs.2.9, 2.10 and 2.11 for cold-worked nickel, iron and solution treated Incoloy 904 alloy respectively, the signal being photographed from an oscilloscope screen. Such traces as these have been observed by a number of workers (Kusanagi et al 1979(a), Shibata and Ono 1981, Edwards et al 1985). Three different bandwidths were used and in this case the magnetising frequency was 5 Hz and the amplification was 46dBs. Unlike the nickel specimen, the iron and Incoloy 904 specimens show two peaks in activity at fields of equal magnitude on either side of the zero field position. Also the nickel specimen has significantly more M.A.E. in the high frequency region.

The effect of varying the current range chosen for the B.E. and M.A.E. activity profile measurements was investigated. It was observed that reducing the current range did not significantly influence the activity profile within the range measured. This is demonstrated for B.E. in fig.2.12 (the sweep frequency has to be altered inversely with the current range in order to keep the magnetisation rate constant). Similarly the effect of using an asymmetrical magnetisation cycle is demonstrated for M.A.E. activity profiles in fig.2.13. Here the current range was constant but the average current progressively offset from zero. Again there were no significant changes in the profile for fields within the range of measurement.

2.3.3 Magneto-acoustic Emission as a Function of Applied Magnetic Field (Magneto-acoustic Emission Activity Profiles) Method 2: Small Amplitude Field Modulation.

A schematic diagram for the M.A.E. activity profile measurement using the second method is shown in fig.2.7(b). A sweep device varies the current through the magnet at a linear rate of $128\text{Am}^{-1}\text{s}^{-1}$ (1.48mHz) over a particular current range (+I to -I). This slower magnetisation rate gives a higher resolution profile than for method 1 but the signal to noise ratio is correspondingly smaller. In order to improve this a function generator superimposes a high frequency small amplitude current on the driving signal. This is typically a 20-25Hz signal of amplitude 50-150mA. The M.A.E. signal is detected by the 2.5MHz piezo-electric transducer and amplified and filtered with a bandwidth of 32-100kHz as in section 3.2.2. The effect of the field modulation is to sample the signal at any given field several times. However, half of the signal obtained in this way is due to the reverse part of the field modulation. It was observed that by making measurements in this way the activity profile obtained was symmetric about a field position close to zero field. Fig.2.14(a) demonstrates this for an Incoloy 904 alloy specimen. Therefore the signal arising from the reverse part of the modulation field was removed by using a gate device. This device is demonstrated in fig.2.7(inset) and is synchronised to the function generator. In fig.2.14(b) the resulting activity profile is shown for the same Incoloy 904 alloy specimen. The averaging at positive and negative fields now no longer occurs. The activity profile or envelope was obtained by rectification and low pass filtering (3dB at 300mHz) and this is plotted on an X/Y plotter as in section 3.2.2. The true voltage values are obtained from the activity profile in the same way as in section 3.2.2.

Fig.2.15 shows the M.A.E. activity profile obtained from the same pure nickel specimen as in section 2.2.2. Again there is a single peak in the activity which occurs at low fields. Also shown in this figure is the effect of varying the modulation frequency at constant amplitude. It was

observed that the progressively higher frequencies produced progressively higher M.A.E. signals. However it was difficult to drive the coil at the higher frequencies due to its large inductance. Also the higher magnetising frequencies would result in only a fraction of the specimen being magnetised due to the skin depth attenuation so the method would no longer be a bulk measurement. If the frequency is below 15 Hz the filtering is inadequate in producing a smooth envelope as is observed in fig.2.15 for a 10 Hz modulation field. Fig.2.16(a) shows the M.A.E. activity profile from a pure cobalt specimen (B.E. activity profile shown for comparison fig.2.16(b)). No M.A.E. activity is observed which is consistent with the fact that in hexagonal close packed pure cobalt the domain walls are almost entirely of the 180° type (section 1.4).

The M.A.E. activity profiles obtained using this technique have a greater resolution than those of method 1. For example the width of the peak in the activity obtained in the nickel specimen profile (fig.2.8) is larger than that obtained by using this method. However in obtaining the signal in this way the result may well be modified because of what has been called 'rectation', i.e. modification of domain wall movements in the minor loops from that in the corresponding part of the hysteresis curve.

2.4 Coercivity Measurements.

Coercivity measurements were made using the table mounted solenoid. A schematic diagram of the experimental arrangement is shown in fig.2.17. The sweep device controls the field of the solenoid. The specimen sits in the upper detection coil and as the external field varies the e.m.f. induced across the detection coils is integrated and displayed on a 3 1/2 digit D.V.M. The integrator has a time constant of 1 second and the preamplifier gain was set to x10 for all measurements. The e.m.f. induced across the detection coils is given by:

$$\text{e.m.f.} = -C \frac{dM}{dt} \quad (\text{Eq}^n.2.6)$$

where dM/dt is the rate of change of magnetic moment, and C is a constant depending on the flux linkage in the coils.

In order to calculate the coercivity a series of integrator readings were taken when the specimen was at different states of magnetisation. The field is swept between these states at a linear rate of $22,500 \text{ Am}^{-1} \text{ s}^{-1}$. It is important to make the cycling frequency as high as possible in order to reduce the time for measurement and hence error due to integrator drift, but not so high as to create too large a signal for the integrator. Firstly, the current is increased to 5.0 A, saturating the specimen, and the reading noted. Then, the current is swept to -5.0 A and the new reading noted. It is then reduced to 0.0 A and a third reading taken. A fourth reading is taken at 5.0 A, a fifth at -5.0 A, and a sixth and seventh at 0.0 and 5.0 A respectively. This procedure is repeated twice giving a total of 14 readings. From each set of 7 readings ((a) to (g)), the remanence can then be calculated (as a function of C) allowing for a small linear drift in the integrator output. i.e.

$$\begin{aligned} (a)-(b) &= 20.C.M_s + D & \text{where } D \text{ is the integrator drift in half} \\ (c)-(b) &= 10.C.(M_s - R) - D/2 & \text{a cycle, typically small } (10^{-3}M_s). \\ (d)-(c) &= 10.C.(M_s + R) - D/2 & R \text{ is the remanance} \\ \text{etc.} & & M_s \text{ is the saturation magnetisation} \end{aligned}$$

The demagnetising field in the specimen causes the magnetic moment to be nearly proportional to the applied field at low fields. Fig.2.18 shows a hysteresis loop obtained from a mild steel specimen demonstrating this linear dependence (the demagnetising field causes the hysteresis loop to be distorted, having a much smaller permeability). Therefore by measuring the gradient of the hysteresis loop ($C.dM/dH$), the coercivity, H_c , can be calculated, i.e.

$$H_c = \frac{-R}{C.dM/dH} \quad (\text{Eq}^n.2.7)$$

The gradient is measured by taking several integrated readings at low fields and then making a least squares straight line fit on a computer.

2.5 Hysteresis Loops and Permeability.

2.5.1 Measurement of Hysteresis Loops.

The schematic diagram for the plotting of hysteresis loops is the same as that for coercivity measurements (fig.2.17). The sweep generator (or function generator) drives the solenoid from +I to -I to +I at a rate of $445 \text{ Am}^{-1} \text{ s}^{-1}$, where I is a current sufficient to saturate the specimen. The output of the integrator is plotted on an X/Y plotter as a function of the magnetising current.

Due to the demagnetising field the hysteresis loop is distorted. I.e. if the cylindrical specimen can be approximated to an oblate spheroid then the demagnetising field can be written as;

$$H_D = -N.M/\mu_0 \quad (\text{Eq}^n.2.8)$$

where N is a constant known as the demagnetising factor.

Therefore in an external field, H, the internal field, H_i , is given by;

$$H_i = H + H_D \quad (\text{Eq}^n.2.9)$$

Using $B = \mu_0.(1+\chi).H$, $H = (B-M)/\mu_0$ and $1+\chi = \mu_r$, we obtain;

$$\mu_{\text{rod}} = \frac{\mu_r.(N+1)-N}{1+N.(\mu_r-1)} \quad \begin{array}{l} \text{where } \mu_r \text{ is the permeability of an infinite wire} \\ \mu_{\text{rod}} \text{ is the observed permeability} \end{array} \quad -(\text{Eq}^n.2.10)$$

If $\chi.N \ll 1$ then $\mu_{\text{rod}} \approx \mu_r$ and the demagnetising effects are negligible. However in the other extreme where $\chi.N \gg 1$ then $\mu_{\text{rod}} \approx 1 + 1/N$, i.e. the permeability is dependent only on the specimen geometry.

In this thesis most of the specimens had a length of 40mm and a diameter of 2.3mm. This gives a demagnetising factor of 1/132 using the ellipsoidal approximation. (Osborne 1945). Consequently if $\chi \gg 10^2$ then the permeability is dominated by the specimen geometry.

2.5.2 Permeability Measurements.

The experimental arrangement used for measurement of the rod permeability is similar to that used for Hysteresis loop measurement (fig.2.17), but the integrator is replaced by a $\times 10$ amplifier. The

frequency of the magnetising field is normally 5mHz (triangle waveform), and the signal from the detection coils is amplified and plotted on an X/Y plotter. This signal is proportional to the rate of change of magnetisation of the specimen and is therefore a measure of the permeability. Fig.2.19 shows a trace obtained from a specimen of Incoloy 904 alloy for half a hysteresis cycle. As the field is reduced from saturation the permeability increases until it is limited by the demagnetising field at low fields (section 2.5.1). Here the B.E. can be directly observed as a 'hairyness' on the trace. Then as the field increases in the reverse direction the permeability is again reduced.

The permeability traces are calibrated by comparison with the hysteresis loop. In principle this measurement gives no more information than the hysteresis loop does, but it is far easier to carry out.

2.6 Hardness Measurements.

Hardness measurements were made using a Vickers hardness machine. All the measurements shown in this thesis were taken using a 10kg weight. The standard procedure was to make at least three indents in each specimen and to average the results.

2.7 Errors in Barkhausen and Magneto-acoustic Emission Activity Profile Measurements.

Usually, in this thesis, errors in the measurements are not directly indicated on the graphs of data and so I should consider them here. There are three categories:

(1) Errors due to changes in the experimental conditions such as small variations in the setting of the magnetisation frequency, changes in the gain of amplifiers with temperature and changes in the impedance of the magnetising and detection coils with temperature. In order to reduce these the equipment was left running for some time before taking measurements so that the temperature would be relatively stable. Also some care was taken

to correctly set the magnetising field rate and amplitude etc. Normally the error arising from these sources would be less than 5%. Another possible source of error in this category is small variations in the specimen dimensions and the exact position of the specimen in the magnetising solenoid. The amplitude of the B.E. signal is monotonically related to the surface area of the specimen and that for the M.A.E. signal related to the volume of the specimen.

(2) An important source of error which was found to be significant for the B.E. activity profile measurement arises from the random nature of the occurrence in time of each Barkhausen event. The exact shape of the profile was found to vary from one measurement to the next. The size of this error (or better described as a scatter) depends on how large and how many single events make up the activity profile. However in iron-based polycrystalline materials it was typically about 5%. This error could always be made as small as one liked, however, by making several measurements (usually two or three). M.A.E. profile measurements did not suffer from this because the emission is more continuous due to transducer ringing.

(3) If sufficient care is not taken a very large error could result from M.A.E. profile measurements due to variations in the coupling of the piezo-electric transducer. This could be described as a weakness of the technique. However much experience in this helps and again repeated measurements reduce the error. Also a technique was used to monitor the coupling before a measurement. Errors can be 30% or more for a single measurement taken without due care.

In addition to errors, when taking B.E. and M.A.E. measurements we need to consider the scatter in readings obtained from different specimens of an identical material which have undergone similar preparation procedures. B.E. being a surface measurement usually gives a larger scatter than M.A.E. does. Since in some cases quite large differences can be obtained it is important to use at least two or three specimens in each condition under investigation.

Table 2.1

The amplifiers and filters constructed for experimental measurements.

Device	Gain	Bandwidth (3dB)
Amplifier	x10,x5	770kHz roll-off
Amplifier/Filter	x1000	200Hz-75kHz
Amplifier/Filter	x1000	360Hz-100kHz
Filter (Chebyshev) 4 pole h.p.	x1	360Hz
Filter (Chebyshev) 2 pole l.p.	x1	100kHz
Filter (Bessel) 6 pole l.p.	x13.2	5.2Hz
Filter (R-C) 1 pole l.p.	x1	2.2Hz
Filter (R-C) 1 pole l.p.	x1	1.15Hz
Filter (R-C) 1 pole l.p.	x1	300mHz
Filter (R-C) 1 pole l.p.	x1	27mHz

Figure 2.1 Frequency response of the amplifier used for acoustic measurements (with a 4.9 pF capacitor on the input).

Figure 2.2 The experimental arrangement used to observe (a) single Barkhausen pulses and single acoustic pulses.

Figure 2.3 The experimental arrangement used to measure (a) Barkhausen event size distributions and (b) B.E. activity as a function of the field current. The inset demonstrates the operation of the 'window' device.

Figure 2.4 A typical Barkhausen event size distribution obtained for a pure cold-worked nickel specimen for a threshold range of 100 mV. (a) $\log_{10}$ -lin and (b) $\log_{10}$ - $\log_{10}$ ranges.

Figure 2.5 B.E. activity obtained as a function of the magnet current for the cold-worked nickel specimen. The sweep rate is 5 mHz.

Figure 2.6 A typical acoustic signal obtained from breaking a 0.5 mm pencil lead. The pencil break produces a 'step-like' displacement in the metal surface. This subsequently acts as a source of elastic waves which propagate through the material bulk before being detected by the transducer. The amplitude of the first wave arrival, normally a compression wave, will give a reasonable measure of the transducer coupling. If the source- transducer distance is large then the surface wave amplitude can be used as an alternative as this is less rapidly attenuated with distance.

Figure 2.7 The experimental arrangement used to measure the M.A.E. activity as a function of the magnet current, (a) using method 1 and (b) method 2. The inset demonstrates the the operation of the 'gating' device. This accepts only that part of the signal which occurs

Figure 2.8 M.A.E. activity obtained as a function of the magnet current for the cold worked nickel specimen. The upper profile was measured using a bandwidth of 100-1,000 kHz and the lower a bandwidth of 32-100 kHz. The sweep rate is 150 mHz.

Figure 2.9 M.A.E. signals observed from cold-worked nickel before deriving the signal envelope. Three different bandwidths are shown. The superimposed curves indicate the magnet current. Note that there is a single peak in the activity at low fields.

Figure 2.10 Similar M.A.E. signals as fig.2.9 observed from cold-worked iron. Note that there are two peaks in the activity, one at a positive and the other at a negative field.

Figure 2.11 Similar M.A.E. signals as fig.2.9 observed from solution treated Incoloy 904 alloy (iron-nickel-cobalt alloy). Here the activity is a slightly more complex function of the magnet field, although there are two main peaks as was the case for iron.

Figure 2.12 B.E. activity as a function of the magnet current for Incoloy 904 alloy heat treated for 355 hours at 700°C. The profile has a complex shape with three peaks in activity. However as the field range is varied (the sweep frequency being altered in order to maintain a constant rate of change of field), the shape of the profile remains the same. This indicates that the current range used for B.E. measurements is not critical. (The noise background is negligible and the profiles have been placed above one another for clarity).

Figure 2.13 M.A.E. activity (method 1) as a function of the magnet current for the same specimen as in fig.2.12. In this case the current range and sweep frequency were kept constant for each profile at 4.8 A and 150 mHz, but the average current was successively increased for each measurement (therefore the signal actually went off the +2.4 A scale for all but the lower profile). The shape of the profile remains the same within the range measured indicating that the current range chosen is not critical for the M.A.E. measurements. (As in fig.2.12 the profiles have been placed one above the other for clarity, however there is also a noise background of 7.4 mV).

Figure 2.14 M.A.E. activity from solution treated Incoloy 904 alloy, (a) profile without gating and (b) profile with gating. Note that the gate also attenuates the signal by x0.5.

Figure 2.16 There is no observed M.A.E. activity from pure cobalt (a) where only 180° type domain walls exist. However significant B.E. activity (b) can be detected.

Figure 2.17 The experimental arrangement used to make coercivity measurements and to plot hysteresis loops.

Figure 2.18 A hysteresis loop measured from a 0.5%C mild steel which has been homogenised at 1000°C for 1 hour and tempered at 650°C for 10 minutes.

Figure 2.19 The relative permeability as a function of magnet current for solution treated Incoloy 904 alloy. At low fields where the true permeability is high the measured value becomes equal to the reciprocal of the demagnetising factor. The 'hairyness' on the trace is the direct observation of B.E. activity.

Chapter 3. Single Barkhausen and Magneto-acoustic Emission Events.

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3.2.2 Experimentally Observed Waveforms.

3.1 Single Barkhausen Events.

3.1.1 Introduction.

The voltage profile of a single Barkhausen event is largely determined by:

- 1) The permeability and conductivity of the material, and the depth of the event within the material.
- 2) The detection coil impedance.
- 3) The electronic amplification and filtering.

The material permeability and conductivity and the depth of the event affect the degree of eddy current shielding which occurs. Since eddy current shielding is more severe at higher frequencies events detected deeper within the material have proportionally greater low frequency content and so appear broad with a low peak height, the risetime of the 'spike' being determined by the depth of the event. For events very near the surface the voltage profile is sharp with a large peak height, the short risetime being determined by the detection coil characteristics and the bandwidth of the electronics. If there were no high pass filtering, the integrated profile of an event would be independent of its depth, being determined solely by the volume of material swept out by the domain wall, and is simply proportional to the change in magnetisation of the specimen. The eddy current shielding (skin depth attenuation coefficient) as a function of frequency is derived in 3.1.2.

The effects of the detection coil impedance and the electronic amplification and filtering is experimentally determined in 3.1.3, and theoretical event waveforms are determined from the results by computer.

In section 3.1.4 an absolute calibration is made enabling Barkhausen event volumes to be determined and in section 3.1.5 a few examples of experimental waveforms are shown and the event volumes calculated.

3.1.2 Skin Depth Attenuation.

Solving Maxwell's equations in an electrical conductor yields the

following wave equations for the $\mathbf{E}$ and $\mathbf{H}$ fields;

$$\nabla^2 \mathbf{E} = \mu \cdot \epsilon \cdot \frac{\partial^2 \mathbf{E}}{\partial t^2} + \mu \cdot \sigma \cdot \frac{\partial \mathbf{E}}{\partial t} \quad (\text{Eq}^n.3.1) \quad \nabla^2 \mathbf{H} = \mu \cdot \epsilon \cdot \frac{\partial^2 \mathbf{H}}{\partial t^2} + \mu \cdot \sigma \cdot \frac{\partial \mathbf{H}}{\partial t} \quad (\text{Eq}^n.3.2)$$

If we solve these equations for the case of a plane wave with an $\exp(j\omega t)$ time dependence and introduce a complex refractive index, $\mathbf{n}$, such that $\mathbf{E}$ and $\mathbf{H}$ vary as $\exp\{j\omega(t - \mathbf{n}z/c)\}$; then by writing $\mathbf{n} = n - jk$ we find the following;

$$n^2 - k^2 = \mu_r \cdot \epsilon_r \quad (\text{Eq}^n.3.3)$$

and
$$n \cdot k = \frac{\sigma \cdot \mu_r}{2 \cdot \omega \cdot \epsilon_0} \quad (\text{Eq}^n.3.4)$$

for a good metal where $\sigma \gg \omega \cdot \epsilon$ we obtain;

$$n = k = \left[\frac{\sigma \cdot \mu_r}{2 \cdot \omega \cdot \epsilon_0} \right]^{1/2} \quad (\text{Eq}^n.3.5)$$

consequently in a metal the field components are propagated as;

$$\exp(-z/\delta) \cdot \exp j(\omega \cdot t - z/\delta) \quad (\text{Eq}^n.3.6)$$

where $\delta = (\pi \cdot \sigma \cdot f \cdot \mu)^{-1/2}$, $\omega = 2 \cdot \pi \cdot f$

δ is known as the skin depth, and corresponds to the distance over which the amplitude of the wave falls to $1/e$ of its initial value. The magnitude of the skin depth decreases with the inverse half power of the frequency, and of the permeability and conductivity of the metal (Bleaney and Bleaney 1978).

3.1.3 Theoretically Derived Waveforms.

A VAX computer was used in order to calculate the expected detected waveform of a Barkhausen event occurring at some depth within a conductor. This program calculated the waveform using the following stages;

- 1) The values of σ , μ_r and the depth were chosen.
- 2) The domain wall was assumed to move quickly compared to the period of the highest magnetic field frequency component detected at the surface of the material. The velocity of domain walls is typically $\sim 10^4 \text{ms}^{-1}$ (DeBlois, 1958). Therefore if a wall moved as far as 1mm without impediment it would

take only $\sim 0.1\mu\text{s}$. Consequently the frequency is $\sim 10\text{MHz}$, whereas the electronic bandwidth of the equipment is 100kHz . This enabled the domain wall jump to be approximated to a discontinuous step in the magnetisation.

3) The Fourier transform of the step was then multiplied with the skin depth attenuation function.

4) The resulting frequency spectrum was then multiplied with the experimental transfer function (that is the frequency response of the detection coils and electronics).

5) Finally the inverse Fourier transform was calculated and plotted.

The experimental transfer function (which is dependent on the detection coil impedance and the electronic amplification and filtering) was measured in two ways;

(a) The first method measures the function directly by observing the delta-function response. Fig.3.1 shows the experimental arrangement for this measurement. A pulse generator produces a 100ns 8V pulse which is attenuated (21dBs) before driving a small signal coil of 30 turns (diameter 2.55mm , length 6mm , resistance 3.5 ohms), which was centrally located in the upper detection coil. The self-inductance of the signal coil is small ($\sim 1\mu\text{H}$) compared to that of the detection coils, and since the resistance is small compared to the resistance of the supply, (3.5 and 50 ohms respectively), an effective delta-function in magnetisation is 'seen' by the detection coil. The detected signal is then filtered and amplified using the same equipment as that used for Barkhausen event measurements. The signal pulse also acts as a trigger for a further pulse generator which generates a similar delayed pulse. This delayed pulse then defined the gate position on a peak detector. The peak detector could then be used to measure the profile of the detected signal by varying the pulse delay.

(b) The second method measures the experimental transfer function in the frequency domain. Fig.3.2 shows the experimental arrangement for this measurement. A function generator produces a 310 mVpp sine wave which

drives the signal coil via the 21dB attenuator. The detected signal was filtered and amplified as in (a) and this together with the driving signal was observed on an oscilloscope. The frequency was incremented from 200-260,000 Hz and the attenuation and phase shift measured for each value.

The transfer function obtained using the second method is plotted in the time domain in fig.3.3 (the inset shows the delta-function response obtained in (a)). The first peak width is $18.8\mu\text{s}$, corresponding to a roll-off frequency of 27kHz, and this defines the minimum width of a Barkhausen pulse.

Fig.3.4 shows some theoretically derived profiles (events occur at $t=0.2\text{ ms}$). As the depth increases the peak amplitude decreases and the width and peak delay increase. Figs.3.5, 3.6 and 3.7 show the first peak height and full width of the first peak as a function of depth, and the first peak height as a function of permeability respectively (in fig.3.7 the data points are the theoretically derived values).

With a knowledge of the permeability and electrical conductivity of a material it is possible to use this theoretical data to determine the depth of any Barkhausen event. More importantly, with an on-line computer, it should now be possible to deconvolve the depth dependence from the experimental event size distributions in order to obtain the Barkhausen event volume size distributions and the domain wall pinning density directly.

3.1.4 The Coil Constant (C).

The equipment was calibrated in order to determine the volume of material swept out during a single event by domain wall movement. It is necessary to find an expression for the e.m.f. induced across the detection coils as a function of the material saturation magnetisation and event volume (The coil constant, C). The experimental arrangement for detection is the same as that used measuring Barkhausen event waveforms (section 2.2.1). A single layer 78 turn signal coil was wound on a brass

former and inserted in place of a specimen in the upper detection coil. In order to give the same flux linkage this coil was made to be the same physical dimensions as a specimen (40mm long and 2.3mm in diameter). A function generator produces a voltage step, (risetime 5ns), which simulates a Barkhausen event at the material surface (as before, the inductance of the signal coil is small compared to the detection coil). The peak height of the detected signal was then measured as a function of the voltage and current of the step. The coil constant can then be determined. i.e.

The flux linkage, ϕ_1 , of the two coils is given by;

$$\phi_1 = k \cdot \mu_0 \cdot \frac{N \cdot I \cdot l}{(l^2 + 4 \cdot R^2)} \cdot \pi \cdot R^2 \quad (\text{Webers}) \quad \text{where } N=78 \text{ turns} \quad (\text{Eq}^n.3.7)$$

I is current

$l=40\text{mm}$

$R=2.3\text{mm}$

k is a constant depending on the two coil geometries.

On substitution of values and putting I in terms of the observed signal amplitude we have;

$$\phi_1 = 1.34 \times 10^{-12} \cdot k \cdot (\text{peak amplitude}) \quad (\text{Eq}^n.3.8)$$

But for a single Barkhausen event occurring at the surface of a material with saturation magnetisation M_s (in Tesla) the flux linkage, ϕ_2 , is given by;

$$\phi_2 = 2 \cdot k \cdot M_s \cdot \pi \cdot R^2 \cdot \frac{v}{V} \quad \text{where } v \text{ is event volume} \quad (\text{Eq}^n.3.9)$$

V is specimen volume

where k will be the same constant as before.

On setting $\phi_1 = \phi_2$ we obtain;

$$\underline{C = (\text{peak height})/v = 3.73 \cdot 10^{13} \cdot M_s} \quad (\text{volts per cubic meter}) \quad (\text{Eq}^n.3.10)$$

The smallest Barkhausen pulse consistently detectable above the noise using the experimental arrangement in fig.2.3 is ~100mV. This corresponds

to a volume resolution of $\frac{2.7 \cdot 10^{-15}}{M_s} \text{ m}^{-3}$ for a surface event. The largest event observed (10V) are therefore 100x this volume.

3.1.5 Experimentally Observed Waveforms.

Barkhausen pulses were recorded using the experimental arrangement described in section 2.2.1. Fig.3.8 shows two events in pure nickel recorded using a recorder sampling interval of 1µs. The waveforms are very similar to those theoretically derived. Event (b) appears to be larger and occuring from deeper within the material. The permeability for this specimen was measured to be ≈1,700 (section 2.5), consequently the depth and volume of these two events are calculated to be;

event	average depth	volume
	(µm)	(m ³)
(a)	~0	~1.5.10 ⁻¹³
(b)	~20	~3.0.10 ⁻¹³

Fig.3.9 shows two Barkhausen events in Incoloy 904 alloy. In this case event (a) has a larger peak height but event (b) the longer risetime and so it is not obvious which event is the largest in volume. The permeability of the Incoloy specimen was too large to measure using the technique in section 2.5 but was estimated to be ~4,000. Using this figure we obtain the following result;

event	average depth	volume	The absolute accuracy is poor due to the rough estimate of μ_r .
	(µm)	(m ³)	
(a)	~80	~1.6.10 ⁻¹³	
(b)	~225	~1.8.10 ⁻¹³	

So the events are about the same size.

Using the data produced by the theoretical program it is possible to determine the size and depth of any Barkhausen pulse if μ_r is known.

3.2 Single Magneto-acoustic Events.

3.2.1 Rod Resonant Modes.

The profile of an M.A.E. event is largely determined by the specimen geometry and in particular the rod resonance modes. A rod can support both longitudinal and transverse waves. For longitudinal modes in a rod of uniform cross section the frequencies of vibration can be obtained by solving the one dimensional wave equation;

$$\frac{\partial^2 u}{\partial x^2} = \frac{1}{c^2} \frac{\partial^2 u}{\partial t^2} \quad (\text{Eq}^n.3.11)$$

If both ends of the rod are free or clamped we obtain;

$$f_n = \frac{n}{2 \cdot l} \cdot \left(\frac{E}{\rho} \right)^{1/2} \quad \text{where } n=1,2,3... \text{ is the order of the mode}$$

E is the Young's modulus (Eqⁿ.3.12)
l is the rod length
and ρ is the density

But if one end is free and the other clamped,

$$f_n = \frac{n - \frac{1}{2}}{2 \cdot l} \cdot \left(\frac{E}{\rho} \right)^{1/2} \quad (\text{Eq}^n.3.13)$$

The transverse flexural modes can be obtained by solving the three dimensional wave equation, these are;

$$f_n \approx \frac{\pi \cdot r}{4 \cdot l^2} \cdot \left(\frac{E}{\rho} \right) \cdot (n + \frac{1}{2})^2 \quad \text{where } r \text{ is the rod radius} \quad (\text{Eq}^n.3.14)$$

3.2.2 Experimentally Observed Waveforms.

The M.A.E. signals excite the resonant modes of the specimen and so the expected waveform would be that of an exponentially damped sine wave with a frequency close to the resonant mode most easily excited. The velocity of sound in iron and nickel is 5000 and 4900 ms⁻¹ respectively and so all the energy of the M.A.E. signal should excite these modes after times >10μs in specimens of length 40mm.

The M.A.E. events were detected using the experimental arrangement

described in section 2.3.1 with a magnetising field frequency of 100 μ Hz and transient recorder sampling time of 0.5 μ s. Fig.3.10 shows two M.A.E. events obtained from a single crystal specimen of Incoloy 904 alloy. The B.E. was monitored at the same time and this is shown in the upper trace. The M.A.E. events are observed to occur at the same time as the B.E. event. This is the first time that an acoustic signal corresponding to single Barkhausen pulse has been observed (Previous workers detected no M.A.E. below 1Hz using smoothly varying magnetising fields), although only the largest M.A.E. signals are detectable above the noise. Fig.3.10(a) was obtained by using the B.E. signal to trigger the transient recorder, and fig.3.10(b) by using the M.A.E. signal. These coincidences of M.A.E. and B.E. signals were not always observed, as often only one of these signals would be present. The examples shown in fig.3.10 occurred at a field close to the knee region of the hysteresis loop where the M.A.E. activity is at a maximum (fig.2.13). At low fields only B.E. events were observed in this specimen indicating that these events originated from 180 $^{\circ}$ domain wall movements. Single events were also observed from polycrystalline nickel (fig.3.11). In fig.3.11(a) a very narrow bandwidth was used which resulted in a larger signal to noise ratio indicating the resonance in the M.A.E. signal (fig.3.11(b) was taken at the usual bandwidth).

The sensitivity of the transducer / charge amplifier system is $\sim 1.3 \times 10^{10} \text{ Vm}^{-1}$ indicating that the first peak height of the M.A.E. event in Incoloy corresponds to a displacement of only $\sim 30 \text{ fm}$. The dominant frequency component is $\sim 15 \text{ kHz}$ and for this material the Young's modulus is $1.37 \times 10^{11} \text{ Pa}$ and the density is $8,120 \text{ kg m}^{-3}$. If both ends of the specimen are assumed to be free then the first longitudinal mode is 5 kHz , and the first two transverse modes are 5.2 and 14.5 kHz . 5.2 kHz is outside the bandwidth, and so it appears that most of the observed signal is from the second transverse mode. In order to confirm which modes are responsible for most of the detected M.A.E. it would be beneficial to Fourier analyse the signal by using a spectrum analyser, but this was not available.

Although the peak amplitude of a single event will be related to the volume of material swept out by a domain wall, the form of the profile is not very useful since it is almost entirely determined by the specimen geometry. In order to observe M.A.E. signals which were not influenced by this it would be necessary to use a large specimen such that the acoustic path from the event to the transducer involves no reflections. i.e. we could then use a half-space approximation.

Figure 3.1 The experimental arrangement used to measure the frequency response of the B.E. single event measurement system, i.e. the experimental transfer function. Method (a):- Time domain.

Figure 3.2 The experimental arrangement used to measure the frequency response of the B.E. single event measurement system. Method (b):- Frequency domain.

Figure 3.3 The experimental transfer function as measured using method (b). The inset shows the direct measurement using method (a).

Permeability = 1700

Conductivity = $1.64 \times 10^7 \Omega^{-1} \text{ m}^{-1}$

Event	Depth	Risetime	Peak height	Full width (at half peak)
	(μm)	(μs)	(V)	(μs)
(a)	75	28.8	5.0	65.6
(b)	100	35.2	2.9	84.8
(c)	125	43.2	1.8	105.6

Figure 3.4 Theoretically derived B.E. profiles for events at depths of (a) 75 μm , (b) 100 μm and (c) 125 μm .

Figure 3.5 The first peak height of the theoretically derived B.E. signal as a function of the event depth for a material with an electrical conductivity of $2.04 \times 10^6 \text{ ohm}^{-1} \text{m}^{-1}$ and a relative permeability of 1000.

Figure 3.6 The full width of the first peak of the theoretically derived B.E. signal as a function of the event depth for the same material as in fig.3.5.

Figure 3.7 The first peak height of the theoretically derived B.E. signal as a function of the permeability at a depth of (a) 25 and (b) 100 μm , the electrical conductivity being $2.0 \times 10^6 \text{ ohm}^{-1}\text{m}^{-1}$

Figure 3.8 Two experimental Barkhausen events observed from pure nickel. The sweep frequency 100 μ Hz. Although both have a similar peak amplitude (B) corresponds to about twice the volume of material swept out by a domain wall than (A).

Figure 3.9 Two experimental Barkhausen events observed from single crystal Incoloy 904 alloy. The sweep frequency is 100 μ Hz. These have quite different peak amplitudes but correspond to about the same volume of material swept out by a domain wall.

Figure 3.10 Upper trace of both (A) and (B): Barkhausen event in Incoloy 904 observed from the knee region of the hysteresis curve. Lower trace of both (A) and (B): Single acoustic event observed simultaneously using a bandwidth of 6-60 kHz. The acoustic signal peak height corresponds to a displacement at the transducer of ≈ 30 fm.

(a) Bandwidth $6.3 \times 10^3 \rightarrow 1.6 \times 10^4$ Hz

(b) Bandwidth $6.3 \times 10^3 \rightarrow 6.3 \times 10^4$ Hz

Figure 3.11 Upper trace of both (A) and (B): Barkhausen event in polycrystalline nickel. Lower trace of both (A) and (B): Single acoustic event observed simultaneously.

Chapter 4. The Dependence of Barkhausen and Magneto-acoustic Emission upon Plastic Deformation and subsequent Isochronal Annealing in Polycrystalline Alpha-iron.

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4.1 Introduction.

In this chapter the effects of cold working and its removal by isochronal annealing on B.E. and M.A.E. in pure polycrystalline alpha-iron was investigated. The measurement of the effects of dislocations is not only important in its own right, but also provides the basis for the interpretation of measurements in more complex systems. For example the effects of neutron irradiation in iron-copper alloys (chapter 6), and the effects of stress (Chapters 7 and 8). B.E. and M.A.E. are sensitive to both microstructure and stress. There is potential for using these techniques industrially for the measurement of residual stress (particularly M.A.E.), and most of the research to date has been to calibrate the stress dependence. However the strong microstructural dependence needs to be well understood in order to be able to make quantitative stress measurements. Some work has been done in this area. Tiitto carried out a detailed investigation on the effects of microstructure on B.E. in steel (1977), and Ono and Shibata investigated the M.A.E. from iron and four steels. More recently Ranjan et al. (1986) has investigated the effects of cold work and subsequent isochronal annealing on both B.E. and M.A.E. in pure nickel. Their results showed the same general trends as the iron data presented in this chapter. First, however we shall review plastic deformation and recovery in alpha-iron.

4.2 The Deformation and Recovery of Alpha-iron.

4.2.1 Dislocation Structures.

There have been several reviews on the room temperature deformation of alpha-iron, and its dislocation structure (Keh and Weissmann 1961, Keh 1962, Leslie et al 1963). For small strains the dislocation density is proportional to the strain, but at strains greater than 3.5% the dislocations form cell structures. These cell structures tend to decrease in size for higher strains until for strains of 10% and larger they have a constant diameter of $\sim 1\mu\text{m}$. The presence of precipitates results in many

dislocations remaining within the cells whereas for pure iron the cells become more regular and clear of dislocations and the cell walls are thinner (Messenger 1961). For a given strain a smaller grain size results in a higher dislocation density (Keh and Weissmann 1961, Brandon and Nutting 1960, Michalak and Paxton 1961) but the cell size does not appear to depend upon the grain size (Keh and Weissmann 1961).

4.2.2 Recovery.

The recovery of materials on annealing below the recrystallization temperature has been reviewed by Beck (1954), Norwick (1955), Perryman (1957) and Bever (1957) where the general aspects of annealing are considered. The recovery of alpha-iron after plastic deformation has been investigated by Michalak and Paxton (1961) and Keh (1962). There is no observable change in the dislocation structure and density at or below 400°C (Keh 1962). Short anneals at 550°C result in the dislocations straightening accompanied by a sharp decrease in the flow stress although the dislocation density is only slightly reduced (Keh 1962). After several hours the dislocation density decreases continuously whilst the flow stress remains constant but at a larger value than its prestrained value. The dislocation tangles rearrange themselves into regular sub-boundaries, the interior of the subgrains becoming relatively dislocation free (Hale et al 1960). On annealing for a few hours at 600°C partial recrystallization takes place and after longer times the recrystallization is complete.

4.2.3 Point Defect Recovery.

The recovery of point defects in alpha-iron can be divided into three stages (Corbett 1966). The first two of these stages are below 0°C whilst the third stage is from 0.15-0.2 T_m , where T_m is the melting point.

Stages I and II do not concern us here since they occur below room temperature. They are believed to be due to close pair recombination of vacancies and interstitials (Bilger et al 1968), long range migration of

self interstitials (Bilger et al 1968, Minier-Cassayre 1966) and, at higher temperatures, to dissociation of interstitial impurity self interstitial complexes (Fujita and Damask 1964). There are at least nine substages present but few of these have been identified with much certainty.

Stage III has been associated with interstitial elements. Cuddy measured the resistivity recovery of zone-refined iron after cold working (Cuddy 1965, Cuddy and Railey 1966) and found that the recovery stage present at 60°C even for carbon impurity levels of ~2.4 ppm, disappeared for impurity levels of ~0.8 ppm (wt). Glaeser and Wever (1969) measured two recovery stages in quenched alpha-iron in the temperature ranges 27°C-127°C and 127°C-247°C. Later work (Wever and Seith 1973) demonstrated that the recoveries were not present when all the residual interstitial impurities were removed. It was concluded that these were due to carbide precipitation.

A detailed investigation has been made by Damask and co-workers (Arndt and Damask 1964, Fujita and Damask 1964, Wagenblast and Damask 1962, Wagenblast et al 1964, Damask 1965) using an Fe-0.1%C alloy in the quenched condition and introducing defects by neutron irradiation. They observed the recovery using resistivity (Fujita and Damask 1964), internal friction techniques (Wagenblast and Damask 1962), calorimetry (Arndt and Damask 1964) and electron microscopy (Wagenblast et al 1964). Two recovery stages were found in the unirradiated alloy at 150°C and 230°C and these were thought to be due to the precipitation of $\text{Fe}_{2.4}\text{C}$ and Fe_3C respectively. After the irradiation four recovery stages were observed at -30°, 50°, 265° and 300°C. These were attributed to the break-up of carbon-interstitial complexes at -30°C, the removal of carbon from solution at 50°C, the break-up of carbon-vacancy complexes by migration and the precipitation of Fe_3C at 265°C and the last peak at 300°C was tentatively thought to be due to vacancy migrations.

4.3 Specimen Preparation and Heat Treatments.

The experiment were carried out on two 150 mm iron bars of diameter 5 mm obtained from Johnson Matthey Chemicals Ltd. The impurities are shown in table 4.1. Four cylindrical tensile specimens with dimensions shown in fig.4.1 were machined from these bars.

These specimens were then annealed in vacuum for 16 hours at 950°C in order to remove the cold work. They were then plastically strained by 4.9% using a 5kN Instron testing machine with a strain rate of $1.85 \times 10^{-4} \text{ s}^{-1}$. Fig.4.2 shows the typical stress strain curve obtained. This introduced a fairly large dislocation density into the material without the distribution of the dislocations becoming too inhomogeneous (Leslie et al 1963, Brandon and Nutting 1960). The ends of the specimens were then removed by spark cutting so that their final length was 40 mm and their diameter a little under 2.30 mm.

Then a series of low temperature anneals were carried out at successively higher temperatures on three of the specimens for 1 hour each and B.E. and M.A.E. measurements were made before and after each anneal. The anneal temperatures varied from 250° to 750°C at 100°C intervals.

4.4 Measurements.

Barkhausen event size distribution measurements were made (section 2.2.2) using a magnetisation rate of $43.2 \text{ Am}^{-1}\text{s}^{-1}$ (0.5mHz) over a current range of $\pm 2.4\text{A}$ which was sufficient to take the specimens to saturation. (measurements were not made after the 250°C anneal.)

B.E. activity profile measurements were made (section 2.2.3) using a magnetisation rate of $432 \text{ Am}^{-1}\text{s}^{-1}$ (5.0mHz) over a current range of $\pm 2.4\text{A}$. The B.E. signal gain used was reduced to x1000 and the activity envelope was derived using a 2.2Hz filter.

M.A.E. activity profiles were made (methods 1 and 2, sections 2.3.2 and 2.3.3) using the same current range as for the B.E. measurements, method 2 using a 20 Hz 150 mA r.m.s. modulation field. The plotter X scale

sensitivity was 1 mV cm^{-1} . In method 1 the M.A.E. signal was monitored for three magnetisation cycles, and the results superimposed in order to isolate spurious peaks in the profile. In method 2 the M.A.E. signal was measured for half a magnetisation cycle.

Rod permeability measurements were made (section 2.5.2) using a magnetisation rate of $432 \text{ Am}^{-1}\text{s}^{-1}$ (5.0mHz) over the same current range as for the other measurements.

4.5 Results.

4.5.1 Barkhausen Event Size Distributions.

The Barkhausen event size distribution for one of the specimens measured after straining by 4.9% is shown in fig.4.3. The event count per threshold range is plotted logarithmically against the threshold level. For very small events the count is large, e.g. for the range 200–300 mV the count is 447,700, but this falls away very rapidly to only single counts at 2.6–2.7 volts.

The total Barkhausen event count and the distribution spread for the specimens as a function of the last anneal temperature are shown in figs. 4.4 and 4.5 respectively. Both of these parameters remain fairly constant up to 450°C , the total number of Barkhausen events being $\sim 670,000$ and the distribution spread being 2.7 volts. However after the 550°C anneal the events become a little smaller in size and at higher temperatures there is a dramatic decrease in both parameters until finally after the 750°C anneal the total count has fallen to only $60,900 \pm 1,500$ and the distribution spread to 1.60 ± 0.02 volts.

4.5.2 Barkhausen Emission Activity Profiles.

Fig.4.6 shows a series of B.E. activity profiles from one of the specimens after each anneal. The horizontal scale is the magnetizing current (1A corresponds to $H=8990 \text{ Am}^{-1}$). There appear to be three regions

of activity. The first of these is at low fields where dM/dH is large and there is often a large peak in the activity. The other two regions are at higher fields near the knee of the hysteresis curve where there is often another peak in the activity. The outer peaks can be larger or smaller than the central low field peak, but the first outer peak which occurs when reducing the field from saturation, is never smaller than the final peak when taking the specimen up to saturation in reverse.

Fig.4.7 shows the permeability as a function of the magnetizing field for a specimen (a) after 4.9% plastic strain and (b) the last anneal at 750°C. In (a) the ledges in the permeability plot correspond to the outer peaks in activity in the B.E. activity profile. After annealing (b) the permeability is very much greater so that demagnetising field effects dominate (section 2.5). This results in the permeability rising to the reciprocal of the demagnetising factor for fields lower than the knee regions of the hysteresis loop.

Fig.4.8 shows a schematic representation of the three peaks in the B.E. activity. Regions of activity similar to this have been observed in silicon-iron (Gardener et al 1977). The peak heights of these regions were measured for each specimen after each anneal and the results are plotted in fig.4.9. The central peak height remains fairly constant up to 450°C at ~ 17.2 mV but it then becomes rapidly smaller until at 750°C it is only 0.6 ± 0.1 mV. The initial and final peak heights also diminish at the higher temperatures but in addition they show changes at much lower temperatures. The initial peak height increases from 10.1 mV to 19.5 mV at 550°C and the final peak height shows a similar trend. Generally, as the anneal temperature is increased there is an initial increase in activity but at higher temperatures the activity is sharply reduced. Fig.4.10 demonstrates the redistribution of activity from the low field to the high field regions, where the initial peak / central peak is plotted as a function of the annealing temperature.

Fig. 4.11 shows the initial peak final peak ratio as a function of the

annealing temperature. As discussed above the ratio is never smaller than unity.

4.5.3 Magneto-acoustic Emission Activity Profiles.

Fig.4.12 shows an example of an M.A.E. activity profile measured using method 1 (section 2.3.2). There are just two peaks in the activity, these peaks occurring at the same field as the two outer peaks in the B.E. activity profiles ($\approx 7200 \text{ Am}^{-1}$). Again the initial peak is always larger than the final peak. There is some activity at low fields but it is not sufficient to produce a peak.

Fig.4.13 shows the M.A.E. activity profiles for one of the specimens after each anneal using method 2 (section 2.3.3). The two peak amplitudes and the central low field amplitude were measured for each of the specimens after each anneal and the noise subtracted. They were plotted as a function of the anneal temperature as was done for the B.E. activity profiles, fig.4.14. The initial and final peaks show the same trend, the activity increasing slightly up to 450°C , and then falling away rapidly at temperatures above 550°C . The central dip amplitude shows a small increase at 550°C before falling away at 650°C .

The ratio of the two outer peaks is plotted in fig.4.15. At high temperatures the ratio falls to 1.0.

4.6 Discussion.

4.6.1 General Features of Profiles.

In iron there are three peaks present in the B.E. activity profiles. Two of these are close to a field which corresponds to the 'knee' of the hysteresis loop and the central one occurs in the coercive region. Only two of these peaks are observed in the M.A.E. activity profile, there being little activity at low fields. The activity at low fields where the permeability is high must therefore be due to the pinning of 180° domain walls.

The interactions between domain walls and dislocations are different in iron and in nickel. Theoretical calculations (Scherpereel et al., 1970), indicate that in alpha-iron dislocations interact with 180° domain walls more strongly than with 90° walls. In nickel, on the other hand, the interaction with 180° domain walls is weaker than with other orientations. These predictions are confirmed by a comparison of the B.E. and M.A.E. activity, especially at low fields, in the two materials. We would expect most of the 90° and 180° domain wall movements to occur at low fields. However in alpha-iron we observed a low activity from 90° domain walls (fig.4.13) and a much higher activity from 180° domain walls (fig.4.6). This implies that 90° domain walls interact more weakly with dislocations than 180° domain walls. In cold worked nickel only a single peak in the activity was observed in both the B.E. and M.A.E. profiles (figs.2.5 and 2.11). This activity was found to be due to pinning of domain walls by dislocations. i.e. on annealing at 1200°C for 17 hours both the B.E. and M.A.E. activity were reduced by $\sim 80\%$ (fig.4.16), indicating a reduction in the number of pinning sites. Also the reduction being about the same for both signals implies that domain walls of angles other than 180° are chiefly involved in the pinning. This implies that in nickel the dislocations interact more weakly with 180° domain walls than with other orientations.

In iron the outer peaks in activity from both the acoustic and electromagnetic signals may well be due to domain wall creation and annihilation. If this were so then we would expect the Barkhausen events that occur at the first peak to be larger than those in the final peak, i.e. because of the large energy required to nucleate reverse domains, once these form on defects or grain boundaries (Goodenough, 1954). They grow quickly resulting in large single events, whereas when domain walls are annihilated the volume of material swept out by the domain wall movement is not necessarily large. Experimentally it was observed that the Barkhausen events in the first peak region do tend to be larger than those in the

final peak region, that is the distribution spread is larger (section 4.7.3). But the total number of Barkhausen events counted is also greater than that of the final peak region. However the total domain wall area created is equal to the domain wall area annihilated and the number of events which occur during creation and annihilation are likely to be similar. But because of the skin depth attenuation, (skin depth $\sim 20\mu\text{m}$ in annealed iron at 5kHz using $\mu_r=10,000$) larger events can be detected to a greater depth in the material. Thus a larger distribution spread is consistent with a larger total count for the same number of domain wall displacements in the sample. Similarly a smaller Barkhausen event count would be expected in the final peak when the distribution spread is smaller. This agrees with for instance fig.4.11, where the ratio of the initial to final peak heights is always greater than unity, except after the final two anneals.

Fig.4.15 shows a similar result for the ratio of initial peak to final peak for the M.A.E. activity profiles, although the change is much smaller. Experimentally it was observed that the ratio of the initial and final peak heights measured from the M.A.E. activity profiles were numerically equal to the square root of the same ratio from the B.E. activity profiles, except for after the 450° and 550°C anneals. (The values for the M.A.E. ratio were measured from the profiles taken using method 1, because, although these are less accurate, the method 2 technique introduces a further modification to the peak heights which, although is monotonic, is not linear). Table 4.2 shows the values of these ratios and the square root of the electromagnetic signal ratio. The outer peaks in both the M.A.E. and the B.E. activity profiles originate from the same physical process so that we may expect the peak heights to be related in some simple way. One explanation for the observed relationship is as follows. The acoustic energy that is released by the event, which is assumed to be proportional to the magnetic energy, is converted into the specimen resonances (3.2.1) that are then detected by the transducer. The transducer output is

proportional to the surface displacement which is in turn proportional to the square root of the resonance energy over a limited bandwidth. (i.e. $\text{Energy} = 0.5 \times \text{Stress} \times \text{Strain} = 0.5 \times \text{Elastic Modulus} \times \text{Strain}^2$). In contrast the B.E. signal is proportional to the change in the magnetic energy, not the square root. Therefore the rms M.A.E. signal should be proportional to the square root of the energy of the events whereas the rms B.E. signal proportional to the energy of the events. After the 450° and 550°C anneals this empirical relationship is no longer true (table 4.2). This is just the region where recovery takes place and the dislocation density begins to fall (Keh 1962, Leslie et al. 1963). This point will be discussed in section 4.6.2.

The activity at high fields in the M.A.E. and B.E. activity profiles are believed to originate from domain wall creation and annihilation. The activity at low fields is due to both irreversible 180° domain wall and 90° domain wall movements in the B.E. activity profiles and 90° domain wall movements in the M.A.E. activity profiles, with the 180° domain wall activity predominating in the B.E.

4.6.2 The Effect of Plastic Deformation.

The main trends for all the graphs of the activity and also for the Barkhausen event size distribution and total count is a large reduction in value after the 1 hour anneal at 650°C. After this anneal the dislocation density would be dramatically lower (Keh 1962, Leslie et al 1963) and so the Barkhausen events would be smaller and less numerous. The central peak height from the B.E. activity is reduced to ~5% of its starting value (fig.4.9) and the total count to ~24% (fig.4.4). The effect on the 90° domain wall pinning as indicated by the activity at low field in the M.A.E. profiles, is only reduced to 67% (fig.4.14) again indicating the weaker effect of the dislocations upon 90° domain wall motion. Squaring the ratio shows the M.A.E. activity after the 650°C anneal to be 45% of the original value, which is ~9 times greater than the corresponding value from B.E.

The measurements of dislocation density made by Keh (1962) after anneals at 550°C would indicate that after the 1 hour anneal the dislocation density would be reduced by 10-20% (~14% for 16% prestrain (Keh 1962)) and that the dislocations would be straightened. The B.E. activity central peak height for 550°C is reduced by 26% (fig.4.9) whilst for M.A.E. the peak is actually larger (fig.4.14).

The local stress relief caused when dislocations straighten would tend to increase the M.A.E. signal (section 7.4 and Kusanagi et al 1979b) but not necessarily change the B.E. signal very much (since B.E. does not have the same sensitivity to stress, i.e. it may even decrease; section 7.3.1). Therefore if the dislocation density is reduced by only a relatively small amount the M.A.E. actually may still increase whilst the B.E. activity decreases.

The initial and final peak heights for both the electromagnetic and acoustic signals (figs. 4.9 and 4.14) show an upward trend at temperatures below 550°C. The rate of increase in activity for the B.E. outer peaks is about equal to the square of that rate for the M.A.E. activity (using method 1 data) indicating that the same process is responsible in both cases. This trend does not occur for the low field activity and so it must be inherent in the domain wall creation/annihilation process. At these temperatures only impurity complexes and possibly vacancy migration would occur. Cuddy and other workers have found recovery stages in alpha-iron, below 250°C, which they found were due to only a few ppm of impurities (see section 4.1.3). However although the increase in the M.A.E. activity occurs predominantly after the 250°C anneal, this is not the case for the B.E. high field activity. In section 6.5, where the effects of isochronal heat treatments on an iron-copper alloy is discussed, large increases in the B.E. activity were observed after heat treatment at 290°C. This was also observed to occur in pure iron which had been heat treated at 850°C and then at 290°C. The reason was thought to be due to the removal of vacancies quenched in at 850°C which would have the greatest effect on the B.E. since

this is a surface measurement (the surface being subject to the maximum quench rate). Damask and co-workers observed a recovery stage in Fe-0.1%C at 300°C which they tentatively thought to be due to vacancy migration (see section 4.2.3). In this case however, it is not clear how the impurities or vacancies would effect the high field B.E. and M.A.E. activity. It may be that the domain wall creation/annihilation processes are particularly sensitive to the presence of small obstacles.

4.7 A Simple Model for B.E. and M.A.E. Outer Peak Ratio in Terms of the Dislocation Density.

4.7.1 Introduction.

In section 4.6.1 it was argued that the high field regions of activity in the B.E. and M.A.E. activity profiles may be due to the creation and annihilation of domain walls. If this is the case then it is possible to calculate the initial peak / final peak ratio of these profiles in terms of the dislocation density. In doing this several assumptions and approximations are made and so the final results must be considered at best to give only an order of magnitude for the dislocation density. The basis for this model is that the initial peak / final peak ratio is greater than unity due to a net imbalance between the energetics of domain wall creation and annihilation.

Consider first the B.E. When a domain is nucleated there is a change in magnetisation due to the volume of material swept out by the domain walls and also by the volume of the domain walls themselves. On annihilation where the domain walls cease to exist the change in domain wall energy is of the opposite sign and the volume swept out is smaller (section 4.6.1). However in each case the volumes of material swept out by the domain walls is a function of the pinning density, and so the peak height ratio can be calculated in terms of this density. Finally the pinning density is calculated in terms of a dislocation density.

In section 4.6.1 an argument was given for the M.A.E. outer peak ratio

to be numerically equal to the square root of the B.E. outer peak ratio. However the straightening of dislocations at 550°C would slightly reduce the local stress fields which increases the M.A.E. relative to the B.E. and also modifies the outer peak ratio. Consequently the M.A.E. data will give a larger estimate of the dislocation density since the model assumes the dislocations have a constant pinning strength.

The model assumes that the bowing of domain walls at the pinning points does not contribute significantly to the changes in domain volumes and domain wall areas during the creation and annihilation processes. An estimate of the bowing of domain walls is made in section 4.7.6 which supports this assumption. Also a theoretical value for the coercivity is obtained.

4.7.2 The Change in Magnetisation Due to the Creation / annihilation of Domain Walls.

Consider the magnetic moments in the domain wall to be arranged as in fig.4.17. The angle between neighbouring magnetic moments is assumed to be the same throughout the wall thickness. If there are n atoms in the wall separated by a distance a , then the change in magnetisation, M , which occurs on lining up all the moments is given by;

$$M = \frac{A}{a^2} \sum_i (m - m \cdot \cos \theta_i) \quad (\text{Eq}^n.4.1)$$

where A is the wall area, m the atomic moment, θ_i is $i\gamma/n$ and γ is the angle between the magnetisation directions in the two domains.

On doing the summation,

$$M = A \cdot M_S \cdot \left(\delta - \left(\frac{\delta}{n} \right) \cdot \cos \frac{(n+1) \cdot \gamma}{2 \cdot n} \cdot \frac{\sin(\gamma/2)}{\sin(\gamma/2n)} \right) \quad (\text{Eq}^n.4.2)$$

where $\delta = n \cdot a$ is the wall width and $M_S = m \cdot a^3$

But normally $n \gg 1$

$$\text{Therefore } M = \delta \cdot A \cdot M_S \cdot \left(1 - \frac{\sin \gamma}{\gamma} \right) \quad (\text{Eq}^n.4.3)$$

4.7.3 The Volume and Area of Domain Wall Created and Annihilated.

In this section a rough estimate of the volume swept out and the change in the area of domain walls during creation and annihilation is made.

Consider the pinning points to be in the form of a simple lattice of spacing s (obviously a simplification). As two 90° domain walls approach each other (fig.4.18(a) &(b)) and begin to annihilate (fig.4.18(b) &(c)) the volume swept out divided by the area of wall annihilated is of the order of;

$$\frac{V_a}{A_a} = \frac{n \cdot s^3}{2 \cdot n \cdot s^2} = \frac{D^{-1/3}}{2} \quad (\text{Eq}^n.4.4)$$

where D is the pinning density and n an integer.

This value is assumed to be the same for domain wall creation, i.e.

$$\frac{V_c}{A_c} = \frac{n \cdot s^3}{2 \cdot n \cdot s^2} = \frac{D^{-1/3}}{2} \quad (\text{Eq}^n.4.5)$$

The factor of $1/2$ is simply a product of this particular model. In the case of a random distribution of pinning points a different factor would be obtained.

In general there would be some bowing of the domain wall. This would change both the volume of material swept out and the area of domain wall created/annihilated during an irreversible event since the bowing process is reversible. i.e. before a wall movement the area of wall would be slightly larger and a little of the volume would have already been swept out reversibly. This point will be considered in section 4.7.6.

The ratio of values of V_a and V_c were estimated in the following way: First the threshold intercepts from the Barkhausen event size distributions were measured at the fields corresponding to the position of the outer peaks in the B.E. activity profile. Then the volumes were calculated using the calibration in section 3.1.4 and assuming that (a) these events correspond to surface events (which is reasonable) and (b) the volumes so derived are at least proportional to the true average volumes (an error of

the same geometric magnitude of the two readings will cancel out then taking the ratio).

The values for the unannealed prestrained material were found to be;

$$V_c = 1.6 \times 10^{-14} \text{ m}^{-3}$$

$$V_a = 8.5 \times 10^{-15} \text{ m}^{-3}$$

See fig.4.19 (a) and (b).

4.7.4 Calculation of the Ratio and Pinning Density.

The total domain wall area created should be equal to the total area annihilated. Therefore the ratio of the outer peak heights in the Barkhausen activity profiles should be equal to that for single events. This makes the additional assumption that the event size distributions during creation and annihilation are of the same form. For a single event we have;

$$R = \frac{V_c \cdot M_s + A_c \cdot \delta \cdot M_s \cdot C}{V_a \cdot M_s + A_a \cdot \delta \cdot M_s \cdot C} \quad \text{where } C = \frac{1 - \sin \gamma}{\gamma} \quad (\text{Eq}^n.4.6)$$

Where R is the ratio of the changes in magnetisation during creation and annihilation.

Therefore from eq^{ns}.4.4 and 4.5;

$$R = \frac{V_c}{V_a} \cdot \frac{1 + 2 \cdot \delta \cdot C \cdot D^{1/3}}{1 + 2 \cdot \delta \cdot C \cdot D^{1/3}} = \frac{V_c}{V_a} \quad (\text{Eq}^n.4.7)$$

From the Barkhausen event size distribution data (section 4.7.3) we have;

$$\frac{V_c}{V_a} = 1.88$$

The B.E. activity outer peak ratio for the unannealed prestrained material is 1.84 ± 0.03 . This agrees very well with the data from the event size distributions and supports the assumption made above. Therefore we can use the activity profile measurements for the calculation rather than event size distribution data (thus we do not need to carry out the rather lengthy single event distribution measurements).

Consider the magnetic energy available to create and annihilate domain

walls: The external field changes at the same rate during creation and annihilation (set by the magnetisation frequency) and so the same energy is available to initiate these processes. Also both occur over a similar field range (around the knees of the hysteresis loop). Therefore since the domain wall energy is of the opposite sign in the two cases we require;

$$1 = \frac{V_c - A_c \cdot \delta \cdot C}{V_a + A_a \cdot \delta \cdot C} \quad M_s \text{ and } H \text{ cancel} \quad (\text{Eq}^n.4.8)$$

Where, here, we have equated the magnetic energy changes only, since the changes in H and M are the same at the two knees.

Therefore from Eqⁿ.4.7,
$$D^{1/3} = \frac{1}{2 \cdot \delta \cdot C} \cdot \frac{R-1}{R+1} \quad (\text{Eq}^n.4.9)$$

On substitution of the appropriate constants and using the measured values of R we obtain the pinning densities listed in table 4.3. We are assuming 90° wall movements are responsible for the outer peak activity (4.6.1), therefore $\gamma = \pi/2$. δ was taken to be 45nm for such walls. The pinning density becomes much smaller after the 650° anneal as expected.

In the case of the M.A.E. measurements we can use the same expression, even though we have not considered the physical problem since the ratio of the outer peak heights is numerically equal to the square root of those for the B.E. measurements. i.e. for these replace R by R^2 in the expression for the pinning density, D.

4.7.5 Calculation of Dislocation Density.

Let there be an average length L of dislocation associated with each pinning point. Then the dislocation density is simply given by;

$$N = D \cdot L$$

If we consider the pinning points to be in a regular lattice as before then the most effective domain wall pinning will occur when the dislocation is aligned the [↖] with domain wall so that L is equal to the lattice spacing s.

Therefore we have a lower limit given by;

$$N = D^{-2/3}$$

We could do better by considering an arbitrary pair of angles between the dislocation line and the domain wall before summing over all angles. However this calculation will serve as an estimate. The value of N as a function of annealing temperature is shown in fig.4.20. This data shows the large decrease in the dislocation density after the 650° anneal directly. However it is also interesting that the model indicates that the density is initially $\sim 10^{14} \text{ m}^{-2}$. A typical dislocation density for 5% plastically strained pure iron with a similar grain size is $6.2 \times 10^{13} \text{ m}^{-2}$ (Keh, 1962). This indicates that the model is certainly predicting dislocation densities of the right magnitude.

4.7.6 Calculation of Wall Bowing.

In the calculation of the changes in volumes and areas of domain and domain walls during irreversible events it was assumed that the domain wall bowing was negligible. We can calculate an upper limit for this corresponding to the dislocations being aligned with the domain walls so that $L=s$. The pinning energy per unit length of dislocation is then the order of;

where λ is the first magnetostrictive constant.

δ is the Bloch wall width.

$\lambda \cdot \sigma \cdot \delta^2 \approx \lambda \cdot \mu \cdot b \cdot \delta$ (Eqⁿ.4.10) σ is the average stress in a cylinder of
(Friedel, 1967) diameter δ around the dislocation.

b is the interatomic distance.

μ is the shear modulus.

This equation takes no account of the domain wall or dislocation type (again we could consider those types of dislocations which occur in iron). For example, Scherpereel et al (1970) calculated the interaction strength for a number of dislocation and domain wall types with different relative orientations and some of these (for 90° walls) are listed in table 4.4. For iron the value of $\lambda_{100} \cdot \mu \cdot b \cdot \delta$ is 1.9×10^{-11} which compares well with these. The detailed calculations of Scherpereel et al indicated that the

dislocation/ domain wall interaction was only significant for separations smaller than or of the same order as the domain wall width δ . Therefore we shall consider a simplified distance dependence of the form $\exp(-x^2/\delta^2)$ where x is the separation. Other smooth functions with the required properties were found not to greatly influence the final result. Since a length s of dislocation is involved in the pinning the interaction energy is given by;

$$F(x) = -\lambda.\mu.b.\delta.s.\exp(-x^2/\delta^2) \quad (\text{Eq}^n.4.11)$$

Consider the pinning points as a regular three dimensional lattice as in section 4.7.3. (In strained iron the dislocations tend to cluster so that there are dislocation rich regions and relatively dislocation free regions. Therefore a better approximation would be to assume that there were two regions with different pinning densities. This could be incorporated into the model easily provided that the relative volumes of these two regions could be estimated). On applying an external field the domain wall will move and bow slightly before breaking away from a pinning point. The increase in wall energy for each wall segment (fig.4.21) is $w.\Delta A$ where w is the wall energy per unit area and ΔA the increase in area. Therefore on considering the energy as a function of wall displacement, x , and applied field, H , we obtain;

$$E(x,\theta) = \Delta H.M_S.s^2.\frac{x}{2} + \Delta H.M_S.\frac{V(\theta)}{2} - w.\Delta A(\theta) + F(x) \quad (\text{Eq}^n.4.12)$$

where ΔV is the volume of material swept out by bowing alone, ΔH the increment in the field which gives rise to the bowing. Also if the wall makes an angle θ at the pinning point with respect to its original position (fig.4.21), we have;

$$A(\theta) = s^2.\frac{\theta-1}{\sin\theta} \quad (\text{Eq}^n.4.13)$$

$$V(\theta) = s^3.\frac{\theta}{4.\sin^2\theta} - \frac{\cos\theta}{4.\sin\theta} \quad (\text{Eq}^n.4.14)$$

Now as the field increases the domain wall will move so as to minimise the energy. Therefore for any particular field;

$$\frac{\partial E}{\partial x} = 0 \quad \text{and} \quad \frac{\partial E}{\partial \theta} = 0 \quad (\text{Eq}^{\text{NS}}.4.15(a),(b))$$

From Eqⁿ.4.12 we obtain;

$$\frac{\partial E}{\partial x} = \Delta H \cdot \frac{M_S}{2} \cdot s^2 - 2 \cdot x \cdot \frac{\lambda}{\delta} \cdot \mu \cdot b \cdot s \cdot \exp(-x^2/\delta^2) = 0 \quad (\text{Eq}^{\text{n}}.4.16)$$

and,

$$\frac{\partial E}{\partial \theta} = \Delta H \cdot \frac{M_S}{2} \cdot \frac{\partial}{\partial \theta} [V(\theta)] - w \cdot \frac{\partial}{\partial \theta} [A(\theta)] = 0 \quad (\text{Eq}^{\text{n}}.4.17)$$

On substitution of $\Delta V(\theta)$ and $\Delta A(\theta)$ from Eq^{NS}.4.13 and 4.14 we obtain;

$$\frac{\partial E}{\partial \theta} = \Delta H \cdot \frac{M_S}{4} \cdot s^3 \cdot \text{cosec}^2 \theta \cdot (1 - \theta \cdot \cot \theta) - w \cdot s^2 \cdot \text{cosec} \theta \cdot (1 - \theta \cdot \cot \theta) = 0$$

-(Eqⁿ.4.18)

Eqⁿ.4.16 indicates that a progressively larger field, H, is required in order to increase the wall displacement x, until a critical field is reached when the wall will break free. This critical field is the maximum value such that there is still a real solution for the displacement and is given by the nontrivial solution of;

$$\frac{\partial H}{\partial x} = 0 \quad (\text{Eq}^{\text{n}}.4.19)$$

Therefore, from Eqⁿ.4.16 we obtain;

$$\frac{\partial H}{\partial x} = \frac{4 \cdot \lambda \cdot \mu \cdot b \cdot \exp(-x^2/\delta^2)}{\delta \cdot M_S \cdot s} - \frac{8 \cdot \lambda \cdot \mu \cdot b \cdot x^2 \cdot \exp(-x^2/\delta^2)}{\delta^3 \cdot M_S \cdot s} = 0 \quad (\text{Eq}^{\text{n}}.4.20)$$

Hence;

$$x = \frac{\delta}{\sqrt{2}} \quad (\text{Eq}^{\text{n}}.4.21)$$

On substitution into Eqⁿ.4.16;

$$H = 2 \cdot \frac{\sqrt{2} \cdot \lambda \cdot \mu \cdot b}{e \cdot M_S \cdot s} \quad (\text{Eq}^{\text{n}}.4.22)$$

It is interesting to note that for a typical pinning density of $1 \times 10^{20} \text{ m}^{-3}$

(see table 4.3) we find H is 1600 Am^{-1} (20 gauss).

On substitution of Eqⁿ.4.22 into Eqⁿ.4.18 we obtain the following transcendental equation for θ ;

$$(1 - \theta \cot \theta) \cdot \frac{\lambda \cdot \mu \cdot b \cdot \operatorname{cosec} \theta - W}{\sqrt{2} \cdot e} = 0 \quad (\text{Eq}^n.4.23)$$

Notice that the bowing angle θ is independent of the lattice spacing s (i.e. the pinning density). This equation was solved iteratively on a small computer to obtain;

$$\theta = 5.9^\circ$$

Remember this was an upper limit for the bowing angle where the dislocation was taken to be parallel to the domain wall. Therefore the bowing by dislocations does appear to be quite small as was assumed in the calculation of the dislocation density.

4.8 Conclusions.

Both B.E. and M.A.E. are sensitive to dislocations and the agreement with the expected temperature dependence of the dislocation density is good. Both signals show a large reduction above the temperature at which the dislocation density is reduced. By using both techniques together it was possible to determine whether 90° or 180° domain walls were responsible for the activity. In iron the 180° domain walls appeared to be more strongly pinned by the dislocations whereas in nickel domain walls at angles other than 180° were much more strongly affected by them. In iron the irreversible motion of domain walls at low fields is almost entirely due to 180° domain walls whereas at higher fields near the 'knee' regions of the hysteresis curve 90° domain walls also give an important contribution. The effect of stress recovery through dislocation straightening in iron is observed as a change in the M.A.E. relative to the B.E. at both high fields (most easily observed in terms of the outer peak height ratios) and at low fields.

For practical testing of industrial materials, the different dependence of B.E. and M.A.E. on stress, makes it potentially possible to use these techniques in order to determine the residual stress and the microstructure of a specimen in an unknown condition. However the stress dependence of M.A.E. and B.E. will be considered in more detail in chapters 7 and 8.

TABLE 4.1
Iron impurity levels in parts per million
(from Johnson Matthey Chemicals Limited)

p.p.m.									
Co	Cu	Mg	Si	Al	Ca	Cr	Ni	Mn	Ag
3	2	2	2	1	1	1	1	<1	<1

TABLE 4.2
Outer peak height ratios for B.e. and m.a.e. activity profiles showing correspondence between
 $\sqrt{\text{B.e. and m.a.e. activity ratios}}$ for low annealing times.
(Figures in brackets are only estimated)

Last Anneal	M.A.E. Activity Signal Initial Peak/Final Peak (Direct Method)	B.E. Activity Signal Initial Peak/Final Peak	B.E. Activity Signal $\sqrt{\text{(Initial Peak/Final Peak)}}$
4.9% Strain Only	1.37 $\pm$ 0.01	1.84 $\pm$ 0.03	1.36 $\pm$ 0.01
250°C	1.4 $\pm$ 0.2	2.04 $\pm$ 0.05	1.43 $\pm$ 0.02
350°C	1.35 $\pm$ 0.05	1.8 $\pm$ 0.1	1.34 $\pm$ 0.04
450°C	1.29 $\pm$ 0.04	2.11 $\pm$ 0.09	1.45 $\pm$ 0.03
550°C	1.10 $\pm$ 0.02	1.86 $\pm$ 0.03	1.36 $\pm$ 0.01
650°C	(1.00)	1.03 $\pm$ 0.08	1.01 $\pm$ 0.04
750°C	(1.00)	1.15 $\pm$ 0.09	1.07 $\pm$ 0.04

Table 4.3
The pinning densities estimated from the B.E.
outer peak ratio for the iron specimens.

Last Anneal (°C)	Pinning Density $\times 10^{20}$ (m ⁻³)
4.9% Strain Only	6.9-8.0
250	10.4-12.6
350	5.0-8.6
450	11.0-15.1
550	7.2-8.4
650	0.00-0.04
750	0.007-0.35

Table 4.4

The maximum dislocation / domain wall interaction energy, $F_{\max}$, for various dislocation and domain wall orientations. (Data taken from Scherpereel et al, 1970)).

90° Type A Bloch Wall

b	t	Wall		$\delta/\text{\AA}$	θ	γ	ω	$F_{\max}/\text{Jm}^{-1}$
		Plane						
a/2 [111]	$[\bar{1}00]$	(010)		408	0°	54°44min	-45°	1.45x10 ⁻¹¹
a/2 [111]	$[\bar{1}00]$	(010)		408	90°	54°44min	-45°	1.93
a [001]	$[\bar{1}00]$	(010)		408	0°	90°	-90°	1.85
a [001]	$[\bar{1}00]$	(010)		408	90°	90°	-90°	2.94
a [010]	$[\bar{1}00]$	(010)		408	0°	90°	0°	1.41
a [010]	$[\bar{1}00]$	(010)		408	90°	90°	0°	1.41

90° Type B Bloch Wall

a/2 [1 $\bar{1}$ 1]	[0 $\bar{1}$ 1]	(111)		516	-35°16min	144°44min	-54°44min	2.00
a/2 [1 $\bar{1}$ 1]	$[\bar{1}10]$	(111)		516	35°16min	90°	0°	1.51
a [011]	[0 $\bar{1}$ 1]	(111)		408	-35°16min	0°	-	1.12
a/2 [1 $\bar{1}$ $\bar{1}$]	$[\bar{1}11]$	(110)		516	35°16min	0°	-	0.75
a/2 [1 $\bar{1}$ $\bar{1}$]	$[\bar{1}11]15^\circ[00\bar{1}]$	(110)		516	20°16min	15°	-90°	1.26
a/2 [1 $\bar{1}$ $\bar{1}$]	$[\bar{1}11]30^\circ[00\bar{1}]$	(110)		516	5°16min	30°	-90°	1.76
a/2 [1 $\bar{1}$ $\bar{1}$]	$[\bar{1}11]45^\circ[00\bar{1}]$	(110)		516	-9°44min	45°	-90°	0.75
a/2 [1 $\bar{1}$ $\bar{1}$]	$[\bar{1}11]60^\circ[00\bar{1}]$	(110)		516	-24°44min	60°	-90°	0.58
a/2 [1 $\bar{1}$ $\bar{1}$]	$[\bar{1}11]75^\circ[00\bar{1}]$	(110)		516	-39°44min	75°	-90°	1.05
a/2 [1 $\bar{1}$ $\bar{1}$]	$[\bar{1}11]90^\circ[00\bar{1}]$	(110)		516	-54°44min	90°	-90°	1.40

where the plane of the domain wall lies on the yz axis and the dislocation is in the second domain. We then have;

a the lattice parameter,

b the dislocation burgers vector,

t a unit vector defining the positive sense of the dislocation line,

δ the domain wall width parameter,

θ the angle between the projection of the magnetisation direction in the first domain on the yz plane and z axis,

γ the angle between b and the z axis,

ω the angle between b_e and x axis.

Figure 4.1 The dimensions of the iron tensile specimens.

Figure 4.2 The stress strain curve for 4.9% plastic deformation.

Figure 4.3 A typical Barkhausen event size distribution obtained for an iron specimen after straining by 4.9% and before annealing.

Figure 4.4 The total Barkhausen count as a function of the final annealing temperature for the iron specimens.

Figure 4.5 The Barkhausen distribution spread as a function of the last annealing temperature for the iron specimens.

Figure 4.6 The B.E. activity profiles for one of the iron specimens obtained after each anneal. (The last two profiles are shown on the following page on an expanded scale).

) Before Anneal

) 750°C / 1 hour

Figure 4.7 The permeability as a function of the magnet current for an iron specimen (a) after 4.9% strain and (b) after the 750°C anneal.

Figure 4.8 A schematic representation of the B.E. activity profiles showing the three 'peaks' in activity.

Figure 4.9 The initial, central and final peak heights from the B.E. activity profiles as a function of the annealing temperature for the iron specimens.

Figure 4.10 The ratio of the initial and central B.E. peak heights as a function of the annealing temperature for the iron specimens.

Figure 4.11 The ratio of the initial and final B.E. peak heights as a function of the annealing temperature for the iron specimens.

Figure 4.12 An example of an M.A.E. activity profile using method 1 for an iron specimen after the 250°C anneal. Note the two peak structure.

Figure 4.13 The M.A.E. activity profiles for one of the iron specimens obtained after each anneal using method 2.

Figure 4.14 The initial peak, low field and final peak heights from the M.A.E. activity profiles (method 2) as a function of the annealing temperature for the iron specimens.

Figure 4.15 The ratio of the initial and final M.A.E. peak heights (method 2) as a function of the annealing temperature for the iron specimens.

Figure 4.16 The unannealed (as received plus machine worked) and annealed (1200°C for 17 hours) activity profiles as a function of the magnet current for pure nickel. (a) B.E. and (b) M.A.E.

Figure 4.17 A schematic representation of the atomic magnetic moments in a Bloch domain wall with a thickness of n atoms.

(B)

(D)

$$\frac{\Delta V}{\Delta A} = \frac{2 \cdot s^2 \cdot x}{4 \cdot s \cdot x} = \frac{1}{2 \cdot D^3}$$

Figure 4.18 A model representation of two 90° domain walls approaching each other ((A) and (B)) under the influence of an external magnetic field, and then beginning to annihilate ((C) and (D)),

Figure 4.19 The Barkhausen event size distributions obtained from (A) the initial peak region and (B) the final peak region for an iron specimen after 4.9% strain. The sweep rate used was 10 mHz and the current ranges 1150 - 950 mA for (A) and -950 - -1150 mA for (B).

Figure 4.20 The calculated dislocation density for the iron specimens as a function of the annealing temperature.

Figure 4.21 A schematic representation of domain wall pinning and subsequent bowing on the application of a magnetic field. Here the domain wall is drawn much thinner than δ for clarity. The circles represent the pinning sites which have an associated pinning energy of $f(x)$. As the field, H , increases the wall moves through the pinning points a distance x and the bowing angle θ increases. Eventually a critical value of H will be reached at which point the domain wall will break free. Note that for this representation it is necessary that $s > \delta$.

Chapter 5. The Dependence of Barkhausen and Magneto-acoustic Emission upon Non-magnetic Precipitates in Single Crystal and Polycrystalline Incoloy 904 Alloy.

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5.7 Summary and Conclusions.

5.1 Introduction.

In the previous chapter we investigated the effects of cold-working in pure polycrystalline iron on B.E. and M.A.E. The main source of domain wall pinning was dislocations. In this chapter the effect of precipitation hardening is investigated. Incoloy 904 (51.0%Fe., 33.8%Ni., 14.0%Co., 1.2%Ti. by wt.) was chosen as a model material. This is a high strength, low thermal expansion nickel based superalloy used in machine components. The microstructure of nickel based superalloys is reviewed by G.P. Sabol and R. Stickler (1969). In particular the properties of titanium-hardened iron-nickel-cobalt alloys have been studied by Eiselstein (1966) and Angus et al. (1970).

Incoloy 904 has a second phase which is thought to have a simple morphology consisting of non-magnetic coherent spheres. These precipitates or non-magnetic inclusions grow as a random distribution on isothermal ageing. The volume fraction of these inclusions remains constant but their diameter can be varied by the length of the heat treatment. The M.A.E. and B.E. are used to monitor the precipitation of the second phase (γ') and, as in chapter 4, to lay a foundation for differentiating this dependence from other parameters such as stress.

5.2 The Material.

The Incoloy 904 system has been selected for its suitability as a model material in an investigation of the effects of inclusions on the magnetisation process. In particular, their effects on the B.E. and M.A.E. activity. Some of these requirements are discussed by Taylor (1981). The main requirements are;

- (1) A reasonably large saturation magnetisation, which should not be too dependent on small changes in the composition of the matrix material.

The saturation magnetisation is 125 kAm^{-1} and does not change very much on 5 wt.% changes in the composition of the matrix. The Curie temperature is $\sim 400^\circ\text{C}$ and so measurements can be made at room temperature.

- (2) A domain wall width, δ , which is in the range in which the inclusion diameter can be varied. For Incoloy 904, $\delta \approx 180$ nm.
- (3) Well defined easy axes of magnetisation, which are the $\langle 100 \rangle$ crystal directions in Incoloy 904.
- (4) Controllable levels of undesirable obstacles such as dislocation networks, carbide platelets and high angle grain boundaries. The formation of obstacles to domain wall motion, other than the non-magnetic γ' phase can be suppressed by careful preparation of the material and by reducing the carbon content.
- (5) An even distribution of non-magnetic inclusions in the matrix with a simple morphology.
- (6) A large magnetostriction to give good M.A.E. The magnetostriction constant is approximately 1.5×10^{-5} in Incoloy 904.

The main deviation of the Incoloy 904 system from the model requirements is the development of strain and the generation of dislocations for particle sizes $\sim \delta$ and larger. The γ' phase (Ni_3Ti) nucleates homogeneously and grows as spheres on ageing, there being only a small lattice mismatch with the matrix ($\ll 0.5\%$). At large diameters, d , the shape changes to cubic form, and for $d \gg 50$ nm strain is observed in the regions surrounding the inclusions. For $d \gg 300$ nm the inclusions become semi-coherent with interface dislocations. The range of inclusion sizes is limited by the necessity of using practical ageing times $\ll 1000$ hours. The maximum inclusion size is ~ 400 nm in the single crystals, ~ 600 nm in the polycrystals ($\approx 2 \times \delta$ and $3 \times \delta$ respectively).

From the Lifshitz-Wagner theory (Lifshitz and Slyzov, 1961; Wagner, 1961) the diameter of the inclusions is expected to be proportional to the cube root of the ageing time, $t_a^{1/3}$ and so the ageing times were chosen to be close to values $t_a(n)$, where $t_a(n) = n^3$ (n integer, t_a in hours).

5.3 Specimen Preparation.

5.3.1 Polycrystalline Rod Specimens.

Cylinders each with a diameter of 3 mm and a length of 50 mm were cut by spark erosion from a large bar obtained by Inco Europe Ltd. at the European Research and Development Centre, Birmingham. The specimens were electropolished at -5°C for 10 minutes with a current of 300 mA using a solution consisting of 10% perchloric acid, 45% acetic acid and 45% 2-butoxyethanol. This was done in order to remove surface damage, carbon deposits and other contaminants.

All the specimens were solution treated at 1050°C for 2 hours in vacuum and then water quenched in order to prevent any precipitate growth on cooling. It was important to prevent any oxidation of the alloy as this would lead to segregation of titanium to the surface, which would seriously effect the homogeneity of inclusion growth on ageing. The specimens were aged at 700°C in vacuum in order to grow the non-magnetic inclusions. There were one or two specimens for each ageing time, the times chosen being listed in table 5.1. Finally the specimens were centreless ground to 2.35 mm diameter and short lengths were spark cut from the ends for electron microscopy, reducing the length to 40 mm. This removes the surface of the heat treated material which may be depleted of titanium. The specimens were then electropolished again in the same way as before in order to remove the surface damage and to give a good finish with a diameter of 2.3 mm.

5.3.2 Single Crystal Rod Specimens.

The single crystals of Incoloy 904 had been previously grown by Inco Europe Ltd. The specimens were spark cut so that the axis of the cylinder lay along one of the $\langle 100 \rangle$ easy directions, the length being 40 mm and the diameter 2.3 mm. Solution treatment was carried out at 1050°C for 2 hours in vacuum followed by quenching in water, and then ageing at 700°C in vacuum for various times, as shown in table 5.2. These specimens were

previously prepared by R. Taylor for her own work (1981).

5.3.3 Polycrystalline T.E.M. Specimens.

The rod specimen ends were cut using a high speed silica carbide saw into discs of thickness $\sim 500 \mu\text{m}$. These were then ground on 1200 grade paper reducing the thickness to $\sim 150 \mu\text{m}$. The foils were then thinned using a commercial double-jet electropolisher, a 'Tenupol' made by Struers Ltd. This dished both sides of the T.E.M. disc simultaneously and had an automatic control for stopping the polishing just at the point of perforation. The polishing solution used was 20% perchloric acid in ethanol. The material ageing times looked at using T.E.M. were unaged, 27, 343 and 1013 hours.

5.4 Measurements.

Barkhausen event size distribution measurements were made (Section 2.2.2) using a magnetisation rate of $169 \text{ Am}^{-1}\text{s}^{-1}$ (2.0 mHz) over a current range of $\pm 2.35\text{A}$ which is sufficient to take the specimens close to saturation. Also, for the single crystals, event size distributions were measured from different regions of the hysteresis cycle. The B.E. activity profile was considered to be made up of 5 regions (section 5.5.6), the three peak regions and the two dips between the peaks (see fig.5.1). The event size distributions from these regions were measured using current ranges of;

- (a) $-1,000 - -700 \text{ mA}$, initial peak.
- (b) $-500 - -200 \text{ mA}$, region between initial peak and central peak.
- (c) $-150 - +150 \text{ mA}$, central peak.
- (d) $+200 - +500 \text{ mA}$, region between central peak and final peak.
- (e) $+700 - +1,000 \text{ mA}$, final peak.

For these measurements the magnetisation rate was $162 \text{ Am}^{-1}\text{s}^{-1}$ (30 mHz). The smallest single event detectable in Incoloy 904 is $1.7 \times 10^{-15} \text{ m}^3$ (see section 3.1.4).

B.E. activity profile measurements were made (section 2.2.3) using a magnetisation rate of $423 \text{ Am}^{-1}\text{s}^{-1}$ (5.0 mHz) over a current range $\pm 2.35 \text{ A}$. The signal amplification used was x1,000 for the single crystals and x10,000 for the polycrystals. The filter used to derive the signal envelope was 2.2Hz and 300mHz respectively.

M.A.E. activity profile measurements were made (method 1 and 2, sections 2.3.2 and 2.3.3) using the same current range as for the B.E. activity measurements. The results using method 1 are not presented in this thesis as they give no additional information. The method 2 measurements were made using a 20 Hz 150 mA r.m.s. modulation field. The plotter X scale sensitivity was 1 mV cm^{-1} .

Coercivity and hardness measurements were made as described in sections 2.4 and 2.6 respectively.

Observations of the Incoloy 904 microstructure were made using the Philips electron microscope (E.M.). The Lorentz microscopy was done using the high voltage electron microscope (H.V.E.M.). Domains were imaged using the Foucault technique (obtained by tilting the beam so that the magnetically split diffraction spot coincides with an edge of the objective aperture) and the domain walls by the Fresnel technique (obtained by defocussing the objective lens (Jakubovics, 1975)). In order to observe domain wall movements the magnetisation stage was used. This stage enables the specimen to be magnetised uniformly with a field range of $\pm 0.011 \text{ Tesla}$. The field direction lies in the foil plane in order to reduce demagnetising effects and the field inhomogeneity is $< 10\%$. In the Fresnel technique, domain walls appear bright in one of a pair of micrographs and dark in the other. In the Foucault technique the domain contrast is determined by the direction of their magnetic moments relative to the beam direction.

(I am grateful to J.P. Jakubovics and M.G. Hetherington for assistance with the H.V.E.M.)

5.5 Results.

5.5.1 Hardness Measurements from the Polycrystalline Material.

Fig.5.2 shows the hardness as a function of the ageing time. The peak in the hardness occurs after ~520 hours. H. Wiggins and Co. Ltd. (1974) report a time of only ~10 hours for an alloy containing 1.82%Ti and also some carbon. This would give rise to a faster Ni_3Ti growth and also carbide growth respectively. Mason and Rotherham (1972) report peak ageing after ~25 hours in a 1.5%Ti alloy, and this is consistent with the even longer time observed for the 1.2%Ti alloy.

5.5.2 Coercivity Measurements.

5.5.2.1 Polycrystals.

Fig.5.3 shows the magnetic coercivity as a function of the ageing time. The coercivity increases slowly from 90 Am^{-1} to 110 Am^{-1} at 125 hours. Then it increases rapidly until at 1000 hours it is 810 Am^{-1} . The general increase in the coercivity as a function of the ageing time can be understood in terms of progressively stronger pinning of the domain walls as the precipitates grow. The effects of precipitates upon the coercivity has been thoroughly investigated by others (Sagawa et al, 1976; Sasaki et al, 1970; Shilling and Soffa, 1977; and Uhl and Krauss, 1970).

5.5.2.2 Single Crystals.

Table 5.3 shows the coercivities of the single crystals obtained by Taylor. The coercivity increases with ageing time as the inclusions grow. The value for the unaged material is much smaller (21 Am^{-1}) than that of the polycrystals. This implies that the coercivity of the unaged polycrystalline specimens, fig.5.3, is almost entirely due to the grain boundaries.

5.5.3 T.E.M. Micrographs from the Polycrystalline Material.

The distributions of γ' inclusions after ageing for (a) 27 hours, (b)

343 hours and (c) 1013 hours obtained using the Philips electron microscope are shown in fig.5.4. The inclusions coarsen and become less numerous as a function of the ageing time. The material aged for 27 hours has a high density of 92 nm cubical inclusions. But the material aged for 343 hours has a duplex structure of inclusions. The smaller 22 nm inclusions are spherical whilst the larger less dense 260 nm inclusions have a dendritic morphology. The less dense 570 nm inclusions observed in the material aged for 1013 hours also have a dendritic morphology. More extensive observations of the microstructure in this alloy were carried out by Taylor (1981). The interaction with domain walls would be expected to be quite different for these different inclusion sizes and distributions.

5.5.4 Domain Wall Observations from the Polycrystalline Material.

Fig.5.5(a,b) show complimentary Fresnel micrographs (taken above and below focus) from material aged for 27 hours. The domain wall interacts weakly along its length with several inclusions, causing the wall to bend slightly at each inclusion. The streak contrast at each inclusion within the domains is due to the dipole field on each inclusion. Fig.5.6(a,b) show complementary Foucault micrographs from the same material. Two domains with magnetisation vectors anti-parallel are observed to run across the length of the picture. Also a series of 90° closure domains are observed to extend from the domain boundary into the right hand domain.

Fig.5.7 shows complementary Fresnel micrographs from material that had been aged for 343 hours. The lines that are dark in both images are bend contours associated with strain; domain walls are again identified by contrast reversal. At A a domain wall is pinned on a large inclusion. Also the same domain wall becomes confused at (B). This is a thin foil effect and does not concern us here.

Fig.5.8(a,b) show complementary Fresnel micrographs from material aged for 1013 hours. The domain walls are severely influenced by the very large inclusions. There are closure domains on the inclusions as can be

seen at (A). The complementary Foucault micrographs of fig.5.9(a,b) show a very complex structure of small domains and several closure networks. These closure domains form in order to reduce the free pole energy (Néel, 1944). Taylor (1981) also observed closure domains in the single crystal material for inclusion diameters >200 nm. These closure domains modify the pinning strength of the inclusions (Dijkstra and Wert, 1950; Goodenough, 1954; Tebble, 1955).

5.5.5 Observations of Domain Wall Movements.

The five Fresnel micrographs of fig.5.10 were taken with decreasing magnetic field from a specimen aged for 27 hours. As the field was reduced gradual changes in the domain walls took place (a-d), until $2.2 \times 10^{-3} \text{T}$ was reached; at that field there was a sudden and much more dramatic rearrangement of the domains (e), resulting in movements of the walls over distances much larger than the separation between inclusions. (The pointer enables the displacement of the wall to be estimated).

Fig.5.11(a,b) show two Fresnel micrographs obtained from a specimen aged for 343 hours. The material is at remanence in fig.5.11(a) This corresponds to the zero applied field position on the B.E. activity profiles. At A the white domain wall crosses a grain boundary giving rise to very large correlation domains. These correlation domains consist of large parallel domains separated by 180° domain walls. At higher fields closure domains tend to form at the grain boundaries (fig.5.11(b)). This would be expected to give rise to more 90° wall activity.

Fig.5.12 shows ten Foucault micrographs obtained from a specimen aged for 1013 hours. Again the domain structure is very much smaller and more complex. As the applied magnetic field is reduced the domain walls move in small jumps. The movements appear to be generally controlled by the closure domain networks around the inclusions rather than by the inclusions themselves.

The large increase in the coercivity for values of $t_a^{1/3}$ in the range

of 8 to 10 seems to be associated with the dramatic change in the domain structure. These small domains would also be expected to reduce the B.E. and M.A.E. activity, i.e. a smaller domain structure will generally give rise to smaller events, resulting in a smaller fraction being detected (see section 5.6.2 for an explanation of this).

The domain observations made by Taylor on the single crystal material indicated similar interactions (except, of course, the grain boundary effects).

5.5.6 Barkhausen Event Size Distributions.

A typical distribution for a polycrystalline specimen aged for 733 hours is shown in fig.5.13, obtained using the full current range. For the polycrystalline material the distribution spread and total count are shown in figs.5.14 and 5.15 respectively. The distribution spread remains fairly constant as t_a is increased, showing only a slight increase from 1.7 to 2.2 Volts. The total number of Barkhausen counts increases monotonically from ~160,000 for the solution treated specimens to about 210,000 at $t_a=216$ hours, before falling away to only 40,000 at $t_a=1013$ hours.

A typical distribution for a single crystal specimen aged for 1020 hours is shown in fig.5.16. Unlike the polycrystals the distribution spread increases for successively longer ageing times, being a maximum after 520 hours (fig.5.17). The total number of Barkhausen counts (fig.5.18) shows the same trends except now the peak is at the slightly longer time of 735 hours. Again the solution treated specimens give ~160,000 counts but the maximum number of counts is 600,000, very much larger than for any of the polycrystalline specimens.

Total Barkhausen counts and distribution spreads for the central peak region as a function of $t_a^{1/3}$ for the single crystals are shown in figs.5.19 and 5.20 respectively. These both show a peak at 520 hours which corresponds to the peak in the mechanical hardness (fig.5.2). Fig.5.21 shows Barkhausen event distributions obtained from the five current regions

(see 5.4). Notice that larger events occur from the initial peak region than from that of the final peak. The events which occur between the peaks in the profile are the smallest and least numerous.

5.5.7 Barkhausen Emission Activity Profiles.

5.5.7.1 Single Crystals.

Fig.5.22 shows the B.E. activity profiles for each ageing time. From these profiles and also from the profiles obtained from the polycrystalline material there are consistently three distinct regions of activity (these being previously mentioned in section 5.4). This is similar to the deformed polycrystalline iron results of the previous chapter (fig.4.6). There is a central peak in the activity at low fields in the coercive region, and also initial and final peaks in the activity at fields corresponding to just below the knees of the hysteresis loop. The initial peak in the activity, encountered as the field is reduced from close to saturation, is always larger than the final peak. All three peaks are found to depend on the ageing time and hence on the microstructure. Fig.5.23 shows a plot of the central peak height as a function of $t_a^{1/3}$ for the specimens. A slow increase in the peak height is observed which reaches a sharp peak at $t_a=520$ hours before falling away again. The unaged specimen has so little activity at low fields that no peak is observed at all. Only the two outer peaks were present but these were somewhat larger than usual.

A plot of the outer peak heights are also shown in fig.5.23. These have a similar trend to that of the central peak height except that the unaged specimen has increased activity.

5.5.7.2 Polycrystals.

The Barkhausen activity profiles obtained from several of the polycrystals are shown in fig.5.24. There is altogether less B.E. activity from the polycrystals, the graphs shown being plotted on a scale magnified 5 times compared with that for the single crystals. The three peaks are not

as well resolved but are still clearly discernable. All three peaks are found to depend on the microstructure but in the case of the polycrystals the central peak shows the strongest trend. A plot of the peak heights as a function of the parameter $t_a^{1/3}$ is shown in fig.5.25. The central peak rises to a maximum after ~300 hours before falling away rapidly after longer times. The initial peak first falls after ageing (similar to the single crystal unaged specimen), and then rises to show a less prominent maximum than the central peak at ~170 hours. The final peak shows a similar trend.

The peak in mechanical hardness at 520 hours (fig.5.2) indicates that slightly larger inclusions than in the case of maximum irreversible domain wall pinning (fig.5.25) have the strongest influence on dislocations.

5.5.8 Magneto-acoustic Emission Activity Profiles.

5.5.8.1 Single Crystals.

Fig.5.26 shows the M.A.E. activity profiles measured using method 2. In this material two peaks were always observed in the profiles. These peaks, as in the case of iron, occur at the same field as the outer peaks in the B.E. activity profiles. There is insufficient activity at low fields to produce a central peak although the activity is generally greater than zero. As in the B.E. activity profiles the initial peak is always larger than the final one. An interesting additional characteristic of these activity profiles is the presence of 'ledges' at slightly lower fields than the knee region. A plot of the zero field activity as a function of $t_a^{1/3}$ is shown in fig.5.27. (The coercivity is relatively small and can be ignored on this field scale, therefore the central dip corresponds to the activity at the steep part of the hysteresis loop). The trend shows a minimum at $t_a=520$ hours rather than the maximum observed in the B.E. activity central peak height plot for the same specimens. The unaged specimen shows very little activity at low fields in contrast to the unaged polycrystalline specimens (section 5.5.7.2). Fig.5.27 also shows the outer

peak heights as a function of $t_a^{1/3}$. These show a similar trend to the corresponding B.E. activity plot. Fig.5.28 shows the initial peak / central dip ratio. This shows a peak after 520 hours. The ratio is more accurate since it removes error from changes in the transducer coupling.

5.5.8.2 Polycrystals.

Fig.5.29 shows M.A.E. activity profiles for several of the polycrystals measured using method 2. The profiles are similar to those of the single crystals with two peaks and a central dip, although 'ledges' are not observed. The two peaks and the central dip amplitudes as a function of $t_a^{1/3}$ are plotted in Fig.5.30. The trend is very different from that of the corresponding low field B.E. activity (fig.5.25). There is a slight reduction in the activity up to $t_a^{1/3}=512$ hours after which the activity falls away much more rapidly. The initial and final peak heights and central dip showing similar trends so that the overall shape of the M.A.E. profiles changes very little as a function of the ageing time except for very long times. Here the ratio of the outer peak heights deviates substantially from unity as shown in fig.5.31.

5.6 Discussion.

5.6.1 The Unaged Material.

5.6.1.1 The Low Field Activity.

For both the single crystals and the polycrystals the general forms of both the B.E. and M.A.E. activity profiles are similar to those of iron. A central peak at zero field is present in the B.E. activity profiles but absent in the M.A.E. (figs.5.22, 5.24, 5.26 and 5.29), and so is likely to be mainly due to thermodynamically irreversible 180° domain wall motion. The unaged specimens exhibit very little B.E. and M.A.E. activity at low fields. They contain few pinning points and so the domain walls can move

relatively unimpeded through the material. It is not known what causes the residual activity but it could be due to a small amount of impurity precipitates such as carbides or nitrides. In the polycrystalline material, whose coercivity seems to be due almost entirely to grain boundaries, these may also play a role (the activity is 50% larger in the polycrystals).

5.6.1.2 The High Field Outer Peaks.

The two outer peaks in the B.E. and M.A.E. activity profiles occur close to the knees of the hysteresis curve and the initial peak is usually larger than the final one (figs.5.22, 5.24, 5.26, 5.29). This was also the case in plastically deformed polycrystalline iron (figs.4.6 ,4.12) and is believed to be due to the difference between domain wall creation and annihilation processes. In fig.5.21 it is apparent that the initial peak region has a larger distribution spread than the final peak region. Therefore the events which occur at this field are generally larger than those occurring at the corresponding reverse field. This is consistent with creation and annihilation of domain walls (section 4.6.1). The ratio of the largest event volumes (calculated as in section 3.1.4) for these specimens are a function of the ageing time. The pinning density calculated as in section 4.7.4 is not a useful function and is not equal to the inclusion density, for, as we shall discuss later, the individual inclusions do not always act as pinning sites. Their pinning strength is a function of their diameter (Tebble, 1955; Schroder, 1969; Nix and Huggins, 1964) and so as the inclusions grow with ageing, their number decrease but their pinning strength increases. (In the case of the iron, in the previous chapter, the assumption that the pinning strength per unit length of dislocation remains constant is a much more reasonable one.)

For the unaged specimens the size of the outer peaks is greater than might be expected from the trends illustrated in figs.5.23 and 5.25 of a fall with decreasing ageing time (for $t_a < 520$ hours). This is probably because in the absence of inclusions, there will be increased difficulty in nucleating domains (Goodenough, 1954), and on nucleation the walls will

make large movements both because there is increased energy stored and because there are no obstacles to their motion. Annihilation is not a nucleation process, but the absence of obstacles will again result in large areas of domain wall annihilating in a single event. The event size distribution spread for the unaged material (fig.5.17) is larger than that for material aged for $t_a < 520$ hours indicating that the events are in fact larger.

In the polycrystalline material the grain boundaries are important when considering the outer peak regions. The height of the B.E. initial peak is again larger for the unaged material than in specimens aged for a few hours. Now however there is little corresponding trend in the distribution spread (fig.5.14). The absence of the largest events may be because the maximum domain wall movement which can occur in a single event is limited by the grain size. For these specimens the maximum event size for $t_a < 520$ hours is 1.8V which corresponds to a maximum volume of $3 \times 10^4 \mu\text{m}^3$ (section 3.1.4), i.e. an approximate grain diameter of $40 \mu\text{m}$. The grain structure has been imaged in a scanning acoustic microscope (Briggs, 1985). The grain size near the edge of the specimen (where it is smallest, and from where the B.E. is detected) was found to be about $40 \mu\text{m}$. Although domain walls can sometimes pass through grain boundaries in some situations, (fig.5.11), this grain size is consistent with the maximum volume swept out by a domain wall.

From microscopy studies the general form of the B.E. and M.A.E. profiles are understood to be as follows;

- (1) On reducing the field from saturation domain walls nucleate and move through the material before becoming pinned. This results in an initial peak. (The magnetising stage on the H.V.E.M. could not produce a sufficiently high field to observe this directly).
- (2) As the field is reduced towards zero the domain walls join up to form a correlation domain network. These large mostly 180° domain walls result in large B.E. activity as they move past pinning points (fig.5.11), and give

rise to a central peak in the activity.

(3) As the field is increased towards saturation in reverse many domain walls annihilate giving rise to a final peak in the activity.

5.6.2 The Effect of Inclusions.

5.6.2.1 Single Crystals.

The effect of the inclusions can be considered as a function of the ageing time. The volume fraction of inclusions in the single crystals was constant (Taylor 1981), and so the effect of ageing is to make individual inclusions grow, but to decrease their number.

The effect this has on the B.E. activity is shown in figs.5.22 and 5.23. When the inclusions are small the domain walls do not make jumps from one inclusion to another, but rather move as plane walls, passing a large number of inclusions in a single movement (see figs.5.5 and 5.10). The initial effect of ageing is to reduce the initial peak height as domain walls can nucleate more easily when inclusions are present as described in section 4.6.2. The central peak grows because 180° domain wall pinning increases slightly. But the coercivity does not increase very much (table 5.3) because most of the movements of the domain walls are quasi-continuous. However as the inclusions become larger and fewer in number, e.g. after 343 hours, domain walls become more strongly pinned by individual inclusions (fig.5.7). Jumps now take place from one inclusion to the next; these give larger and more numerous events (fig.5.17) (the increased number is, of course, exaggerated in the B.E. total count because the increase in event size means in turn that events will be counted from a deeper volume below the surface). Both the initial and central peak amplitudes increase, though the initial peak remains the larger of the two. After 520 hours the inclusions are about the same size as the domain wall width and give rise to the largest B.E. activity.

For ageing times $t_a > 520$ hours, however, there is a decrease in the B.E. activity. The non-magnetic inclusions are now larger than the domain

wall width and the domains are small and their structure is complicated (figs.5.9 and 5.12). Taylor (1981) measured the inclusion diameter for the 735 and 1020 hour single crystals to be 300 nm and 300-400 nm respectively. The closure domains on these inclusions tend to dominate their interaction with the domain walls (fig.5.12). This gives rise to a much higher coercivity (table 5.3). However the domain walls are heavily pinned and their movements smaller (fig.5.17), resulting in a reduction in the B.E. activity at all fields (figs.5.22 and 5.23). The smaller events can only be detected from a small depth of the material and a much larger fraction will be below the detection threshold.

In the B.E. activity profiles of fig.5.22 all three peaks show similar trends with increasing ageing time. This is probably because the size of events associated with domain wall creation and annihilation are determined by the pinning strengths of the inclusions, just as the low field activity is. The trends in the M.A.E. activity however, vary (fig.5.27). The outer peaks show a maximum at $t_a=520$ hours just as the B.E. results do, and this implies that all the discussion above of the B.E. activity peak height trend applies for the 90° walls in the same way. On the other hand, at low fields, the M.A.E. activity is comparatively weak, and shows little variation with ageing time. This is because in these single crystal specimens the specimen axis was a $\langle 100 \rangle$ easy axis, so that most of the domain walls would be 180° at low fields.

In the M.A.E. activity profiles of Fig.5.26, ledges are visible (as indicated, for example, on the 125 hour profile) that are much less apparent on the B.E. activity profiles of the same specimens (fig.5.22). In the specimens with $t_a=27$ to 355 hours the B.E. activity at fields corresponding to the M.A.E. ledges is much smaller, relative to the outer peaks, than for the M.A.E. activity. This strongly suggests that 90° domain wall motion is predominantly responsible for the ledges. In fig.5.21 we observe that the distribution spread for the regions corresponding to the ledges ((b) and (e)) is smaller than that for the outer or central peaks.

This again indicates that 90° domain wall motion is largely responsible. These ledges do not occur in the unaged specimen, so they must arise from the presence of the inclusions.

There are two specimens which show anomalous behaviour. For the $t_a=520$ hour specimen there is a large increase in both the M.A.E. and B.E. activity and it is difficult to identify any ledges. Also the coercivity is anomalously low (table 5.3). With an ageing time of 735 hours, however, a new feature is apparent. In the B.E. activity profile (fig.5.22) there is a preliminary peak just discernable above the noise at negative current, and this corresponds to the first smaller peak in what has become a double outer peak in the M.A.E. The shape of this preliminary peak suggests that this is the normal initial peak observed in the profiles obtained from the specimens with a smaller t_a . Comparison of the B.E. and M.A.E. activity profiles for the 735 hour specimen suggests that most of the activity at fields less than that of the preliminary peak is due to 180° wall movements. This specimen has a number of odd features, for example the uniformity of the B.E. from -0.5 to $+0.75$ A. The behaviour of these two specimens remains to be explained!

It is not known to what extent the results obtained for the single crystal specimens, and in particular the appearance of ledges and double peaks, depend on the alignment of the easy axis with the external field direction. In polycrystals these details may be smoothed out over different crystal orientations, so that they are absorbed in the larger features, and furthermore the grain boundaries may modify the 90° domain closure structure.

5.6.2.2 Polycrystals.

Barkhausen Emission Activity.

The main trends in the B.E. activity profiles are similar to those of the single crystals. Some differences to note are;

- (1) The central peak is now usually larger than the initial peak (fig.5.25).
- (2) The low field activity is generally twice as large (figs.5.23 and 5.25).
- (3) The initial peak height is much smaller (figs.5.23 and 5.25).
- (4) The maximum activity occurs at a smaller value of t_a .

As described in the previous sections we have three inclusion size ranges of interest. (a) ($t_a \leq 27$ hours). These are small inclusions which perturb the domain wall but do not act as individual pinning sites (figs.5.5 and 5.10). Their presence aids domain wall nucleation and thus reduces the initial peak activity (fig.5.25). (b) (343–512 hours). These are larger inclusions which individually pin the domain wall (fig.5.7) and result in increased coercivity and B.E. activity. (c) ($t_a > 600$ hours). These are very large incoherent inclusions which result in a complex domain wall structure (figs.5.8 and 5.12). The coercivity becomes very large (fig.5.3) but the B.E. activity is reduced because of the small size of events, resulting in only a small fraction being detected (fig.5.12).

The initial peak (fig.22) in the polycrystalline material is small (Point (1) and (3) above) and the dependence on ageing time is weak in comparison with the single crystals. The grain boundaries cause domain wall nucleation to be much easier and to require less energy, so that when domain walls are created they not sweep out such large volumes of material. Therefore the initial peak is somewhat smaller and does not depend strongly on the size of the inclusions.

The maximum central peak activity occurs at $t_a = 216$ to 343 hours which is lower than the single crystal value of 520 hours (Point (4)). This may be because the δ' phase was observed to grow slightly more rapidly in the polycrystalline material, so that the inclusions achieve a diameter equal to the domain wall width after this shorter ageing time.

The B.E. at low fields gradually increases as a function of the ageing time (figs.5.24 and 5.25) in the same way as in the single crystals except that the maximum activity is somewhat less. For $t_a = 343$ hours it was 6.65 ± 0.75 mV whereas for the same t_a the single crystal value was 13 mV (Point

(2)), i.e. about twice as much. The reason that the polycrystal activity is generally less by a factor of 2 is probably due to the crystallographic orientation of the single crystals, their having an easy axis parallel to the field.

The Magneto-acoustic Emission.

The M.A.E. activity profiles show a more orderly trend with increasing ageing time than was the case for the single crystals. The profiles are smooth and consist of two simple peaks and a central dip as described in 5.5.7.2 (fig.29). The M.A.E. activity shows little dependence on the inclusion diameter for $t_a < 343$ hours (fig.5.30). This implies that the 90° walls are relatively unaffected by inclusions whose diameter is less than the wall width. The presence of a large number of small inclusions makes the domain wall motion slightly more reversible, and slightly reduces the activity from that of the solution treated material down to ~ 5.6 mV. The high field activity shows a similar trend for $t_a < 343$ hours. The reason for this insensitivity to small inclusions is probably because most of the 90° domain wall movements are associated with closure domains on the grain boundaries (fig.5.11(b)) and the grain size does not change significantly as a function of the ageing time.

For longer ageing times the activity falls off rapidly in the same manner as for the corresponding B.E. central peak height. The reason is probably again that events are small because the domains are smaller due to the presence of the large precipitates (figs.5.8 and 5.9) as for the 180° domain walls in the B.E. activity. The number of closure domains on the inclusions increases but the events associated with these domains are very small and are probably not detected.

The initial and final peak heights show a similar trend to the low field activity (fig.5.30). The closure domains on the large inclusions (fig.5.8) may well persist even at high fields and so the activity due to domain wall creation and annihilation is reduced since the domains are not completely annihilated. Domain wall creation will not occur to the same

extent since the closure domains on the inclusions will simply grow as the field is reduced from saturation. For these specimens the outer peaks in the activity become asymmetrical for large ageing times (fig.5.31) the initial peak being much larger than the final one. The reason for this is not known, although the data show a scatter rather than a trend (c.f. there was peak height asymmetry in the single crystals with $t_a=735$ and 1020 hours).

The amplitude of the M.A.E. activity of the polycrystals is generally slightly greater than that of the single crystals. For example, the initial peak, zero field and final peak amplitudes of the unaged specimens are 13.9, 7.0, 13.9 mV for the polycrystals and 11.2, 3.2, 9.6 mV for the single crystals. i.e. the polycrystals have a significantly higher amplitude around $H=0$ and slightly higher outer peak heights. In the polycrystals we may expect more 90° domain wall activity, first because of closure domains at the grain boundaries (fig.5.11), and secondly because in the single crystals the crystallographic orientation will more strongly favour 180° walls. (Where both the applied field and the rod axis are parallel to an easy axis. This will favour long domains parallel or antiparallel to the rod axis).

5.6.4 Applications.

In Incoloy 904 M.A.E. shows a quite different dependence on the microstructure from B.E. It is unaffected by precipitate sizes smaller than the domain wall width in polycrystals whereas B.E. is changed for sizes >100 nm (27 hours annealing time). For precipitate sizes greater than the domain wall width both B.E. and M.A.E. are strongly affected.

In a practical testing situation where the detection of precipitates may be important, the observed trend in peak heights of M.A.E. makes it possible to determine a single value for the precipitate size; whereas the B.E. peak height, not being a monotonic function of the ageing time (and hence the precipitate size), can give ambiguous values of precipitate size.

In alloys with an inhomogeneous microstructure, M.A.E. would also give a much more realistic measurement than B.E. which is dominated by surface effects. M.A.E. therefore has advantages for determination of the precipitate microstructure, though it is only sensitive to precipitate sizes greater than the domain wall width.

In the determination of stress in these two phase alloys (chapters 7 and 8), it may be difficult to determine the stress using only M.A.E., if the precipitate microstructure is also unknown, because the simple two peak structure of the M.A.E. profiles would be modified by both effects (chapter 8). However, if B.E. measurements are also made it may be possible to determine the stress level (section 8.7). The B.E. activity, particularly at low fields, is dominated by 180° domain wall motion and is found not to be so sensitive to stress. Therefore the precipitate size can in principle be determined and then allowed for in the M.A.E. activity, enabling the stress level to be determined. If the precipitate size is smaller than ~ 100 nm then the M.A.E. profile is insensitive to the precipitates, enabling the stress level measurement to be made using this measurement alone.

5.7 Summary and Conclusions.

M.A.E. and B.E. are affected by inclusions and can be used to determine the precipitate size within the range 100 to 600 nm, particularly if both signals are monitored. B.E. is more sensitive to small precipitate sizes than M.A.E., but for sizes greater than the domain wall width both signals are sensitive.

By comparing both signals the activity at low fields is found to be mostly due to 180° domain wall movements whereas the activity at high fields (near the knee) consists of both 180° and 90° domain wall movements. The trends in the B.E. and M.A.E. peak heights can be summarised as follows;

- (1) For the unaged material the B.E. initial peak height is large because of the difficulty in nucleating domain walls. The central peak

is very small as there is little domain wall pinning.

(2) For material with ~27 hours ageing (≤ 100 nm precipitates) the B.E. initial peak height is reduced as domain walls nucleate more easily. The central peak height increases due to increased domain wall pinning, although the pinning is not by individual inclusions.

(3) For polycrystalline material aged for 216–343 hours and 520 hours in the single crystals, the activity is a maximum. The inclusion diameter is comparable with the domain wall width (150–300 nm), and domain walls are pinned at individual inclusions.

(4) For material aged for >520 hours (300–600 nm precipitates) the B.E. activity decreases although the coercivity is high. The domains are small and their structure complex, resulting in small events. The presence of closure domains on the inclusions reduces the initial peak height, by aiding domain wall nucleation.

(5) For polycrystalline material aged for <343 hours (precipitate diameter <200 nm) the M.A.E. probably arises from closure domains on the grain boundaries and is not significantly influenced by the inclusions. The single crystal initial peak height follows a similar trend to that of the B.E., but is smaller (probably because most of the domain walls nucleate on the grain boundaries).

(6) For material aged for >500 hours (precipitate diameter >300 nm) the M.A.E. is reduced at all fields. The small complex domain structure results in small events.

Most of the differences between the single and polycrystals were understood in terms of;

(1) The grain boundaries, which reduce the size of the Barkhausen events and reduce the size of the B.e. initial peak and make it less sensitive to the precipitates probably because most domain walls nucleate at the grain boundaries. Also the M.A.E. is increased and insensitive to precipitates which are much smaller than the domain wall width because most of the 90° domain wall movements are associated with closure

domains at the grain boundaries (particularly at low fields).

(B) The single crystals having an easy axis parallel to the rod axis and the magnetic field direction which increases the B.e. activity around $H=0$ by a factor of 2, and enables ledges to be observed in the M.A.E. profiles. These are smoothed out in the polycrystalline material.

For the measurement of stress levels, a precipitate size of ≤ 100 nm would enable M.A.E. to be used, since it is insensitive to the small precipitates, otherwise B.E. measurements may also have to be made in order to isolate the effects of microstructure.

TABLE 5-1
Heat Treatment of Incoloy 904 Polycrystals

Number of Specimens	Ageing time at 700°C (hours)
2	0
1	1
1	8
2	27
1	64
2	125
2	216
2	343
2	512
2	733
2	1013
1	1731

TABLE 5-2
Heat Treatment of Incoloy 904 Single Crystals

Number of Specimens	Ageing time at 700°C (hours)
1	0
1	27
1	125
1	355
1	520
1	735
1	1020

Table 5.3

The magnetic coercivity of the single crystal
Incoloy 904 specimens. From Taylor, 1981.

Ageing Time (hours)	Coercivity (Am ⁻¹)
0	31.0 ± 1.6
27	41.4 ± 1.6
125	87.5 ± 6.4
355	103.5 ± 3.2
520	91.5 ± 2.4
735	157.6 ± 4.8
1020	335.8 ± 16.7

Figure 5.1 The current ranges used for Barkhausen event size distribution measurements are shown as (a)-(e) on a B.E. activity profile. The measurements were made using the same 300 mA range so that the variations in the resulting distributions reflect the true differences of the regions.

Figure 5.2 The room temperature hardness as a function of the cube root of the ageing time for the polycrystalline Incoloy 904 specimens.

Figure 5.3 The magnetic coercivity as a function of the cube root of the ageing time for the polycrystalline Incoloy 904 specimens.

(a)

27 hrs

0.5 μm

(b)

343 hrs

0.2 μm

(c)

1013 hrs

1 μm

Figure 5.4 T.E.M. micrographs of inclusions in Incoloy 904 alloy aged for (a) 27 hours, (b) 343 hours and (c) 1013 hours.

(a)

(b)

1 μm

Figure 5.5 Complimentary Fresnel micrographs of material aged for 27 hours showing a domain wall interacting weakly with several inclusions along its length.

(a)

(b)

Figure 5.6 Complimentary Foucault micrographs of material aged for 27 hours showing a domain network.

(b)

Figure 5.7 Complimentary Fresnel micrographs of material aged for 343 hours. At A a domain wall is pinned on a large inclusion. The complicated domain wall structure at B is a thin foil effect.

(a)

1 μm

(b)

Figure 5.8 Complimentary Fresnel micrographs of material aged for 1013 hours. The inclusions are surrounded by closure domains; for example at A.

(a)

(b)

2 μm

Figure 5.9 Complimentary Foucault micrographs of material aged for 1013 hours. The domains are very much smaller than in underaged material and are strongly identified with the inclusions.

Figure 5.10 These five Fresnel micrographs were taken of the same area at different magnetic fields from material aged for 27 hours. As the magnetic field was slowly reduced the domain walls moved relatively smoothly; micrographs (a)-(d) were taken as this occurred. Then as the field was reduced further a very large domain jump occurred over a distance far greater than the inclusion separation resulting in the new domain structure shown in (e). These large jumps are the most readily detectable and their presence indicates that greater B.F. and M.A.E. activity may be observed from the specimens aged for only short times as compared to those of very long times, where the domains are small.

(a) 8.7×10^3 Tesla

(b) 6.5×10^3 Tesla

(c) 5.5×10^3 Tesla

(d) 5.0×10^3 Tesla

(e) 2.2×10^3 Tesla

10 μm

(b)

Figure 5.11 Two Fresnel micrographs of material aged for 343 hours. The specimen is at remanance in (a) and a domain wall can be seen to cross a grain boundary at A. In (b) the applied field was 8.8×10^{-3} Tesla and here a closure domain occurs at the grain boundary. Closure domains tended to occur at the grain boundaries at high fields indicating that more M.A.E. activity may be expected to occur from the polycrystalline material than from single crystals and this at some non-zero field.

Figure 5.10 These five Fresnel micrographs were taken of the same area at different magnetic fields from material aged for 27 hours. As the magnetic field was slowly reduced the domain walls moved relatively smoothly; micrographs (a)-(d) were taken as this occurred. Then as the field was reduced further a very large domain jump occurred over a distance far greater than the inclusion separation resulting in the new domain structure shown in (e). These large jumps are the most readily detectable and their presence indicates that greater B.E. and M.A.E. activity may be observed from the specimens aged for only short times as compared to those of very long times, where the domains are small.

(a) 8.3×10^{-3} Tesla

(b) 7.7×10^{-3} Tesla

(e) 4.1×10^{-3} Tesla

(f) 4.0×10^{-3} Tesla

(c) 6.6×10^{-3} Tesla

(g) 2.8×10^{-3} Tesla

(i) 0.8×10^{-3} Tesla

(j) 0.6×10^{-3} Tesla

(d) 5.5×10^{-3} Tesla

(h) 1.1×10^{-3} Tesla

10 μm

Figure 5.13 A Barkhausen event size distribution obtained for polycrystalline Incoloy 904 aged for 733 hours at 700°C.

Figure 5.14 The distribution spread as a function of the cube root of the ageing time for the polycrystalline Incoloy 904 specimens.

Figure 5.15 The total Barkhausen count as a function of the cube root of the ageing time for the polycrystalline Incoloy 904 specimens.

Figure 5.16 A Barkhausen event size distribution obtained for single crystal Incoloy 904 aged for 1020 hours. at 700°C.

Figure 5.17 The distribution spread as a function of the cube root of the ageing time for the single crystal Incoloy 904 specimens.

Figure 5.18 The total Barkhausen count as a function of the cube root of the ageing time for the single crystal Incoloy 904 specimens.

Figure 5.19 The total Barkhausen count for the central peak region as a function of the cube root of the ageing time for the single crystal Incoloy 904 specimens.

Figure 5.20 The distribution spread for the central peak region as a function of the cube root of the ageing time for the single crystal Incoloy 904 specimens.

Figure 5.21 The Barkhausen event size distributions obtained for the five current regions given in section 5.4. Those shown here were measured from the single crystal Incoloy 904 specimen aged for 520 hours.

Figure 5.22 The B.E. activity profiles for the single crystal Incoloy 904 specimens

Figure 5.23 The initial, central and final peak amplitudes as a function of the cube root of the ageing time for the single crystal Incoloy 904 specimens.

Figure 5.24 Several of the B.E. activity profiles for polycrystalline Incoloy 904 specimens.

Figure 5.25 The initial, central and final peak amplitudes as a function of the cube root of the ageing time for the polycrystalline Incoloy 904 specimens.

Figure 5.26 The M.A.E. activity profiles for the single crystal Incoloy 904 specimens.

Figure 5.27 The initial peak, central dip and final peak amplitudes as a function of the cube root of the ageing time for the single crystal Incoloy 904 specimens.

Figure 5.28 The ratio of the M.A.E. initial peak to central dip amplitudes as a function of the cube root of the ageing time for the single crystal Incoloy 904 specimens.

Figure 5.29 Some of the M.A.E. activity profiles for polycrystalline Incoloy 904 specimens.

(C)

Figure 5.30 The M.A.E. (A) initial peak, (B) central dip and (c) final peak amplitudes as a function of the cube root of the ageing time for the polycrystalline Incoloy 904 specimens.

Figure 5.31 The ratio of the M.A.E. initial to final peak amplitudes as a function of the cube root of the ageing time for the polycrystalline Incoloy 904 specimens.

Chapter 6. The Effects of Neutron-irradiation and Subsequent Isochronal Annealing on Barkhausen and Magneto-acoustic Emission in iron and an iron-copper Alloy.

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6.1 Introduction.

In chapters 4 and 5 the effects of cold-working in polycrystalline alpha-iron and precipitation in Incoloy 904 alloy were investigated. In both of these systems it was assumed that there was one dominant type of obstacle to domain wall motion (in addition to the grain boundaries). In this chapter the effects of neutron irradiation upon very pure iron, and iron containing a small fraction of copper as an impurity, are considered. This system has several types of obstacles which may be important for domain wall pinning. There may be non-magnetic copper precipitates, dislocation loops and/or voids. Therefore the effects that these obstacles have on the B.E. and M.A.E. activity is likely to be more complex. However the results of the previous two chapters should aid the interpretation of the data; particularly for the cases of copper precipitation and dislocation loops (we may expect voids to act as obstacles in a similar way to precipitates, although they will behave differently as a function of ageing).

The chapter is divided into two sections. Section A is concerned with radiation damage in the very pure iron system; though even here the effects are generally dominated by the impurity levels. Section B is a study of the effects of irradiation of an iron-copper alloy. The aim of this study was to investigate the potential for using the B.E. and M.A.E. techniques for monitoring radiation hardening and embrittlement of pressure vessel steel nondestructively. The severity of this problem is known to be strongly correlated to copper impurities. Therefore the iron-copper binary alloy was used as a model system.

Section A.

6.2 Introduction to Pure Iron Study.

The damage structures formed in alpha-iron following neutron (Eyre and Bartlett, 1965; Smidt and Sprague, 1973), heavy-ion (Jenkins et al, 1978)

and electron (Little and Eyre, 1973) irradiations have mainly been characterized using transmission electron microscopy (T.E.M.). These studies have provided valuable information on the nature, geometry and distribution of the defect clusters created following irradiation to fairly high displacement doses. However, a major limitation is that low-dose damage structures ($< 1-3 \times 10^{19} \text{ n.cm}^{-2}$ ($> 1\text{MeV}$)) fall close to, or below, the resolution limit of T.E.M. (Robertson et al., 1982; Buswell, 1982). There is therefore considerable interest in exploring alternative analytical techniques capable of detecting the early stages of radiation damage production in alpha-iron.

In the present investigation, the B.E. and M.A.E. techniques are used to examine the low-dose neutron irradiation response of alpha-iron as a function of heat treatment, to provide additional insight into the underlying damage mechanisms. In particular the effects of interstitial impurities and prior cold work was studied.

6.3 The Experimental Preparation and Investigation.

6.3.1 Materials and Preparatory Heat Treatments.

The cylindrical specimens used for this investigation were 50 mm long and 5 mm in diameter, the starting material being obtained from Johnson-Matthey Chemicals Ltd. The main substitutional impurities and their concentrations are given in table 4.1. Also the levels of carbon and nitrogen are important and these are given as ~110 ppm (wt.) nitrogen and ~15 ppm (wt.) carbon.

The iron specimens were prepared in three conditions by the following heat treatments;

- (i) Strong annealing in vacuum (using silica tube) at 950°C for 16 hours followed by water quenching.
- (ii) Annealed as above but then cooled slowly in the furnace.
- (iii) No heat treatment (i.e. the as-received condition) leaving cold-work in.

The effect of heat treatment (i) is to retain residual carbon and nitrogen and other impurities in supersaturated solid solution, but the slow cooling in (ii) will result in the carbon and nitrogen precipitating as carbides and nitrides. These differences will be discussed in more detail later. The specimens were then neutron irradiated.

6.3.2 Irradiations and Heat Treatments.

The neutron irradiations were carried out in a hollow fuel element position in the PLUTO test reactor at Harwell. The temperature was maintained at $\sim 60^{\circ}\text{C}$ during irradiation by ensuring that they were in contact with the heavy water moderator and coolant. The irradiations covered a period of 3.4×10^5 sec. (~ 4 days), giving an integrated fission ($> 1\text{MeV}$) neutron dose of 2.9×10^{19} n.cm $^{-2}$, as measured by nickel and cobalt monitors.

B.E. and M.A.E. measurements were made before and after the irradiation. Also the irradiated specimens were isochronally annealed for 1 hour periods in 100°C increments over the temperature range 250° to 550°C , and B.E. and M.A.E. measurements made after each anneal.

6.3.3 The B.E. and M.A.E. Measurements.

Barkhausen event size distribution measurements were made before and after the neutron irradiation using a current range of ± 6.0 A, and a magnetization rate of $108 \text{ Am}^{-1}\text{s}^{-1}$ (0.5 mHz). The current range used here was larger than for the similar material of chapter 4 because the larger specimen diameter results in a larger demagnetizing factor.

B.E. activity profile measurements (carried out during the experiment as described above) were also made using the larger current range. The magnetization rate was $1080 \text{ Am}^{-1}\text{s}^{-1}$ (5 mHz) and the amplification used $\times 10^4$. M.A.E. activity profile measurements (method 2) were made using the same current range with a sweep rate of $320 \text{ Am}^{-1}\text{s}^{-1}$ and a 150 mA rms 20 Hz modulation field.

6.4 Results.

6.4.1 The Barkhausen Activity.

The B.E. activity profiles obtained before and after the neutron irradiation for the furnace-cooled, water-quenched and cold-worked conditions of alpha-iron are shown in fig.6.1. As expected the cold-worked material (fig.6.1(c)) has high activity at low fields, the profile being similar to those obtained in chapter 4 for the unannealed cold-worked material, indicating pinning of domain walls. There is some low field activity present from the water quenched material but this is quite small. The furnace-cooled material (fig.6.1(a)) exhibits quite high activity at low fields. The barkhausen event size distributions, fig.6.2, also show these results in a more quantitative manner. In particular the total Barkhausen count is much higher and the maximum event size larger in the cold-worked and furnace-cooled material. The effect of ambient temperature neutron irradiation is to significantly reduce the low field B.E. activity from the furnace-cooled material (fig.6.1(d)), and also to slightly reduce it from the water-quenched material. The activity from the cold-worked material is little effected by the irradiation.

The effects of post-irradiation annealing over the temperature range 250° to 550°C on the B.E. activity profile for the cold-worked iron are shown in fig.6.3. Small changes in the low field activity are apparent at 250°C. However, the dominant recovery effects take place above 350°C, so that by 550°C the profile appears to revert to that typical for the fully-annealed dislocation free condition. The low field activity as a function of the annealing temperature is shown in fig.6.4. Here we can clearly see the thermal recovery of the cold-work. Also shown on the figure are the annealing data for the unirradiated material investigated in chapter 4 (here the absolute activity is normalised so that the initial value is set equal to that observed from the larger specimens used in this study). Comparison of the two curves demonstrates that the effect of neutron irradiation is to (a) lower the overall temperature range for the recovery

of the cold-work in alpha-iron, and (b) to considerably broaden the temperature range over which recovery takes place.

6.4.2 The Magneto-acoustic Activity.

The magneto-acoustic activity measurements support the observations made using B.E. In particular the initial peak / low field ratio for the furnace cooled iron falls rapidly after the irradiation, fig.6.5, indicating that there is a large reduction in the pinning density (see chapter 4, fig.4.14). The ratio is fairly close to unity and thereafter remains fairly constant with temperature. The initial peak / final peak ratio for the cold-worked iron as a function of annealing temperature is shown in fig.6.6. This falls with temperature becoming equal to unity at 550°C. In section 4.7 it was reasoned that this parameter is monotonically related to the dislocation density. The data obtained from the unirradiated material discussed in chapter 4 is also shown on the figure, and again it is apparent that the overall temperature range for recovery of cold-work is reduced.

6.5 Discussion.

The effects of 60°C neutron irradiation on the B.E. and M.A.E. activity in the annealed and cold-worked alpha-iron containing low levels of interstitial impurities can be understood as follows: In the annealed material the irradiation causes the dissolution of precipitates and in the cold-worked material enhanced dislocation recovery. These effects arise from the point defect supersaturation induced by the radiation. In order to establish these interpretations, the essential features of radiation damage production and nitride / carbide precipitation processes in alpha-iron are now considered in general terms, before applying the conclusions to the interpretation of the B.E. and M.A.E. data.

6.5.1 Radiation Damage in Alpha Iron.

The basic mechanisms of damage production in alpha-iron following ambient temperature neutron irradiations are well established (e.g. reviews by Little, 1976, 1984; and Nichols, 1981). The majority of the point defects formed are concentrated in cascades, consisting of a vacancy-rich core surrounded by the associated self-interstitials. A fraction of the damage is also generated outside the cascades initially as single point defects. The self-interstitials aggregate rapidly to form clusters and dislocation loops, whilst spontaneous collapse of the cascade centres can take place to give vacancy clusters (i.e. dislocation loops or possibly microvoids). The temperature for vacancy migration in alpha-iron is not well established, but occurs either in the range $\sim -70^{\circ}$ to -50°C (Hautojärvi et al., 1983) or $\sim 220^{\circ}$ to 320°C (Woo and Frank, 1983). Consequently the dispersion of single vacancies created outside the cascade centres may be considered either truly immobile or subject to slow clustering at ambient temperatures.

The interstitial solutes carbon and nitrogen can play a significant role in the radiation damage processes in alpha-iron. Theoretical and experimental studies indicate trapping of both vacancies and self-interstitials by solute atoms, which leads to the removal of these elements from solid solution by the formation of point defect-impurity complexes. Self-interstitial complexes may also form but these will be unstable at ambient temperature; however, vacancy-carbon or vacancy-nitrogen complexes, with a binding energy of $\sim 0.4\text{eV}$ (Arndt and Damask, 1964; Mondino and Seeger, 1977) are stable at room temperature and only dissociate above $\sim 200^{\circ}$ – 250°C .

In summary the damage structure present in the 60°C neutron irradiated alpha-iron in the present study is that of a spectrum of vacancy clusters in the form of dislocation loops or microvoids and a second population of interstitial dislocation loops, together with a dispersion of single vacancies. Also there is the possibility of the formation of vacancy-solute

complexes, depending on the availability of carbon and nitrogen.

A second aspect which we need to consider in the irradiation response of alpha-iron is the stability of carbide and nitride precipitates. Both the dissolution of pre-existing precipitates and the formation of additional precipitates by neutron irradiation are possible. The dissolution of small precipitates within cascade centres could occur, releasing the carbon and nitrogen into solution. These solutes are then free to interact with point defects to form complexes. Alternatively, irradiation may enhance precipitation by providing nucleation sites in the form of point defect clusters, as detected in the early studies of Hull and Mogford (1961).

6.5.2 Nitrides and Carbides in Alpha Iron.

Two stable forms of nitride can precipitate from alpha-iron containing residual solute levels. The equilibrium phase is f.c.c. γ' -Fe₄N, which is thermally stable to ~500°C. The second form is the intermediate or metastable phase b.c.t. α'' -Fe₁₆N₂, with thermal stability to ~170°C, above which reversion to γ' occurs (Jack, 1951a,b; Ferguson and Jack, 1983, 1984). The carbon can substitute into α'' as Fe₁₆(C,N)₂; however, γ' can only incorporate ~0.2 wt. %C, and therefore at temperatures above α'' stability the carbon precipitates separately as equilibrium ϵ -Fe₃C.

Supersaturated solid solutions of nitrogen in alpha-iron are known to be susceptible to quench-ageing when at room temperature. The nitrogen atoms tend to cluster which is then followed by the formation of α'' -Fe₁₆N₂ (Ferguson and Jack, 1983).

It follows from the above information that in alpha-iron containing nitrogen interstitial solute (as is the case in this study), furnace cooling would mainly result in γ' -Fe₄N precipitation, together with some ϵ -Fe₃C. The precipitates form both intra- and intergranularly and can be quite coarse ($\leq 0.5 \mu\text{m}$). However, in water quenched alpha-iron which is subsequently stored at room temperature, the principle precipitate phases

are $\alpha''\text{-Fe}_{16}\text{N}_2$ or $\alpha''\text{-Fe}_{16}(\text{C},\text{N})_2$. These precipitates form intergranularly as planar particles with sizes ≤ 70 nm.

6.5.3 Precipitate Dissolution Under Irradiation.

The observed high pinning at low fields observed in the unirradiated B.E. activity profiles (fig.6.1) can now be understood in terms of carbonitride precipitation processes. In the furnace cooled material the high activity (fig.6.1(a)) is consistent with the presence of a significant distribution of coarse $\gamma'\text{-Fe}_4\text{N}$ particles and possibly from the Fe_3C phase. The much weaker pinning observed in the low field activity of the water quenched material (fig.6.1(b)) may be associated with the finer distribution of less stable $\alpha''\text{-Fe}_{16}(\text{C},\text{N})_2$ phase, which is produced by room temperature quench-ageing prior to measurement. The strong pinning in the cold-worked condition (fig.6.1(c)) is mainly due to the high dislocation density (chapter 4), but additional pinning due to nitride precipitation on the dislocations is also possible.

The effects of the neutron irradiation was to eliminate most of the pinning from both the furnace cooled and the water quenched specimens (figs.6.1 and 6.5). This implies that there has been precipitate dissolution by irradiation. Specifically, the results suggest that both metastable fine-scale $\alpha''\text{-Fe}_{16}(\text{C},\text{N})_2$ (if this was detected) and the more stable coarse $\gamma'\text{-Fe}_4\text{N}$ precipitates, together with any Fe_3C present, all dissolve under irradiation to $2.9 \times 10^{19} \text{ n.cm}^{-2}$. Also the absence of any post-irradiation precipitation by room temperature quench ageing is consistent with the nitrogen being retained in solution by the formation of vacancy-solute complexes as described in section 6.5.1. Alternatively the nitrogen may be retained in solution in the form of solute atmospheres around the irradiation-induced dislocation loops, by a mechanism analogous to strain-ageing. These processes have been postulated previously to explain depletion of nitrogen from solid solution as was indicated by internal friction measurements (Little and Harries, 1970).

A further aspect is that the point defect clusters and dislocation loops created by the irradiation appear to be on too fine a scale to cause measurable domain wall pinning in their own right.

The mechanisms of irradiation-induced precipitate dissolution, particularly at relatively high displacement doses, have been considered in the literature (Nelson et al., 1972; Ghoniem, 1984). However, the detailed aspects of dissolution appropriate to nitride precipitates in alpha-iron at low neutron doses requires some further consideration. The typical dimensions of α'' and γ' particles are much greater than the size of cascade centres, and so the early proposals of dissolution of small precipitates within cascades as discussed by Pugh (1964) and Damask (1967) can be discounted. Both α'' and γ' are ordered phases and so dissolution by irradiation-stimulated disordering, as proposed by Nelson et al. (1972) for nickel-base alloys is possible. The strong attraction of nitrogen atoms both for single vacancies and for small dislocation loops (i.e. trapping and strain-ageing processes, respectively) would have the effect of raising the effective nitrogen solubility in the irradiated condition, and consequently enhance the driving force for precipitate dissolution.

6.5.4 Irradiation-Enhanced Dislocation Recovery.

The B.E. and M.A.E. results (figs.6.4 and 6.6) clearly demonstrate the role of neutron irradiation in enhancing dislocation recovery processes during thermal annealing.

The observed response must be a consequence of the availability of additional vacancy supersaturation present during the annealing of the radiation damage structure. Vacancies can be 'injected' into the matrix to aid dislocation recovery processes (dislocation climb, polygonization and migration into grain boundaries) from two sources: (i) The break-up of vacancy-interstitial solute complexes; and (ii) the thermal emission from cascade centres (these existing as vacancy dislocation loops or microvoids). The thermal dissociation of vacancy-carbon or vacancy-nitrogen

atom pairs is believed to occur at $\sim 200^{\circ}$ – 300°C (Fujita and Damask, 1964; Wutting et al., 1968; Seidel, 1969). These temperatures are significantly below the temperature range of 300° – 400°C for major recovery and annealing out of radiation damage in the form of defect clusters or dislocation loops (Eyre and Bartlett, 1965).

Overall therefore, by proposing vacancy contributions from both damage components, radiation assisted recovery of cold-work is expected to take place over a broad temperature range from $\sim 200^{\circ}$ to $>400^{\circ}\text{C}$, and this is in accordance with the B.E. and M.A.E. measurements. The relatively small recovery at $\sim 250^{\circ}\text{C}$ compared with the dominant effects in the range 300° – 400°C probably reflect the relative vacancy contributions from the two sources.

6.6 Conclusions of Pure Iron Study.

The effect of 60°C neutron irradiation on alpha-iron containing nitride and carbide precipitates is to reduce the low field B.E. and M.A.E. activity (in particular the M.A.E. initial peak / low field ratio). This is consistent with the dissolution of γ' - Fe_4N and ϵ - Fe_3C precipitates, which are the likely obstacles to domain walls, in furnace cooled material; and dissolution of α'' - Fe_{16}N_2 or α'' - $\text{Fe}_{16}(\text{C},\text{N})_2$ precipitates in water quenched material.

The B.E. and M.A.E. techniques also indicate that subsequent isochronal annealing of neutron irradiated cold-worked alpha-iron leads to enhanced dislocation recovery as compared to the unirradiated material investigated in chapter 4. In particular complete annealing out of cold-work appears to have taken place almost 100°C below that of unirradiated specimens and the recovery process itself occurs over a much broader temperature range. This is believed to be due to additional vacancy supersaturation arising from the break-up of vacancy-interstitial solute complexes, and the thermal emission from cascade centres and dislocation

loops.

Section B.

6.7 Introduction and Irradiation Hardening in Pressure Vessel Steel.

The pressure vessels used for pressurised water reactors (P.W.R.) are constructed out of high toughness quenched and tempered low alloy MnMoNi ferritic steels. The P.W.R. operates at $\sim 290^{\circ}\text{C}$ and during its lifetime of ~ 30 years parts of the vessel receive a neutron dose of $\sim 3 \times 10^{19} \text{ n.cm}^{-2}$ ($>1 \text{ MeV}$).

This irradiation environment can give rise to significant changes in the mechanical properties of these steels (Steele, 1975; Nichols and Cowan, 1971; Little, 1976), such as an increase the yield strength (irradiation hardening) and the ductile-to-brittle transition temperature (radiation embrittlement). These phenomena are known to be associated with the presence of residual levels ($\sim 0.1\text{--}0.35\%$) of copper in the steels (Hawthorne et al., 1975 and 1979; Smidt and Sprague, 1973), although the mechanism is not well understood.

The role of copper in the radiation hardening and embrittlement process may be explained in terms of either: (a) stabilisation of point defect clusters (dislocation loops or microvoids), formed at the centre of displacement spikes by neutron-atom collisions, by the segregation of copper; or (b) precipitation strengthening by the f.c.c. ϵ -copper phase, in which the copper (normally immobile under thermal conditions at 290°C) simply precipitates from supersaturated solid solution as a result of the enhanced diffusion associated with the radiation induced vacancy flux. The first mechanism is suggested by transmission electron microscope (T.E.M.) and field ion microscope (F.I.M.) observations. Thus T.E.M. revealed enhanced retention of voids and dislocation loops in Fe-0.3%Cu relative to pure iron following high dose ($4.5 \times 10^{20} \text{ n.cm}^{-2}$) 280°C -neutron irradiation (Smidt and Sprague, 1973), whilst a high density of $\sim 0.6 \text{ nm}$ diameter

defects, which could be copper stabilised microvoids, have been detected by F.I.M. in neutron irradiated Fe-0.34%Cu (Brenner et al., 1978). Arguments advanced for irradiation induced copper precipitation (Odette, 1983; Fisher et al., 1984) are based on (i) the strong age-hardening response of copper in alpha-iron, in which peak strengthening (during 500°C thermal ageing) is achieved at Cu particle sizes below the resolution of T.E.M. (Hornbogen and Glen, 1960; Goodman et al., 1973a,b), together with the fact that (ii) the low equilibrium solubility of copper in alpha-iron at 290°C (< 0.02%) (Salje and Feller-Kniepmeier, 1977) implies high supersaturations at the reactor operating temperature for typical copper impurity concentrations. Whilst the full details of these mechanisms are still under discussion, it is clear that the microstructural features induced by irradiation, either in the form of precipitates or defect clusters, act as strong obstacles to dislocations and give rise to the observed radiation hardening response. These obstacles may also result in domain wall pinning, and so be capable of detection by the Barkhausen and magneto-acoustic emission techniques.

6.8 The Iron-Copper Alloy System.

In order to simulate the irradiation effects on pressure vessel steels an Fe-0.2%Cu alloy was used as a model system. Specimens of this alloy were irradiated with a high neutron flux over a period of one week so that the total dose received was equivalent to >24 years of pressure vessel service.

An Fe-0.7%Cu alloy was used to investigate the effects of copper precipitation by thermal ageing at 500°C and 630°C on the B.E. and M.A.E. signals. Goodman et al. (1973a) found that thermal ageing at 500°C gave a copper precipitate diameter proportional to the square root of the ageing time, t_a , and so $t_a^{1/2}$ was used as the ageing time parameter for the 500°C data.

6.9 Measurements.

Barkhausen event size distributions were made (section 2.2.2) using a

magnetisation rate of $43\text{Am}^{-1}\text{s}^{-1}$ (0.5 mHz) over a current range of $\pm 2.4\text{A}$. Some measurements were made at 5mHz using a current range of $\pm 300\text{mA}$ ($\text{dH}/\text{dt}=54\text{Am}^{-1}\text{s}^{-1}$) in order to look at the low field region. B.E. activity profile measurements were made (section 2.2.3) using a magnetisation rate of $432\text{Am}^{-1}\text{s}^{-1}$ (5.0 mHz) over the same current range. The signal amplification used was $\times 10,000$ and the envelope derived using a 300 mHz filter. M.A.E. activity measurements were made using method 2 only (section 2.3.3) using the same current range as for the B.E. activity measurements. A 20 Hz 150 mA modulation field was used and the signal envelope derived using a 300 mHz filter.

6.10 Experimental Preparation and Investigation.

6.10.1 Iron-7% Copper Alloy Specimens.

Twenty cylindrical Iron-0.7% Copper specimens of length 40mm and diameter 2.3mm were provided by A.E.R.E. Harwell. These were lightly polished and heat treated at 850°C for 18 hours in vacuum (all heat treatments were carried out in vacuum unless otherwise stated) and water quenched to retain the copper in solution. The specimens were then isothermally aged at 500°C or 630°C for various times (table 6.1) in order to grow the copper precipitates. They were then electropolished, using the same solution and conditions as in chapter 4, in order to remove the surface layer which may be copper depleted (critical for B.E. measurements). B.E. and M.A.E. activity profiles together with Barkhausen event size distributions, using the smaller $\pm 300\text{mA}$ range, were measured from the specimens. In addition the precipitate hardening was monitored using room temperature Vickers hardness tests at 10 kg load.

6.10.2 Iron-0.2% Copper Alloy Specimens.

Twenty iron-0.2% copper specimens with the same dimensions as the iron-0.7% copper specimens were provided by A.E.R.E. Harwell. The ends of these

were lightly polished and they were solution treated and electropolished as in section 6.10.1. B.E. and M.A.E. activity profiles were then measured from all the specimens, and Barkhausen event size distributions, using both the $\pm 2.4\text{A}$ and $\pm 300\text{mA}$ ranges were taken from six of them. Three of these specimens were then neutron irradiated at 290°C for a period of 120 hours in a hollow fuel element in the Harwell PLUTO reactor to a dose of $2.4 \times 10^{19} \text{ n/cm}^2$ ($>1 \text{ MeV}$). B.E. and M.A.E. activity profiles and Barkhausen event size distributions, using both current ranges, were then measured from these. Four further specimens were heat treated at 290°C for 120 hours, as thermal controls, and B.E. and M.A.E. activity profiles measured. Then all seven of these specimens were isochronally annealed for 1 hour periods in 50°C increments over the temperature range 400°C – 650°C , and B.E. and M.A.E. activity profiles were measured after each anneal. The mechanical hardness was monitored throughout as in section 6.10.1 (the radioactive specimens were wrapped in tantalum foil for heat treatments and the resulting thin oxide layer was removed by electropolishing).

In order to further investigate the low temperature thermal effects, which were found to occur, some additional heat treatments were carried out on the solution treated material. Three specimens were heat treated at 290°C , two of these for 10 hours and one for 1 hour. Two specimens were heat treated at 350°C for 120 hours. In order to isolate the effect of the copper a pure iron (Johnson Matthey Specpure) specimen was heat treated at 290°C for 10 hours. After each heat treatment B.E. and M.A.E. activity profiles and hardness measurements were made.

Thirty discs of diameter 2.3mm and thickness $500\mu\text{m}$ were cut, lightly polished and solution treated as in section 6.3.1. These were then isothermally treated at 500°C in vacuum for times ranging from 0–1706 hours (table 6.3). Hardness measurements were then taken in order to monitor precipitate hardening in the iron-0.2% copper alloy.

6.11 Results.

6.11.1 The Iron-0.7% Copper Alloy System.

6.11.1.1 Hardness Measurements.

Fig.6.7 shows the hardness as a function of the ageing time, t_a , at 500°C. The peak precipitate strengthening occurs after ~100 hours. The specimens aged at 630°C showed no significant change in mechanical hardness implying that they were all mechanically overaged.

6.11.1.2 Barkhausen and Magneto-acoustic Emission Activity Profiles.

B.E. activity profiles for each ageing time at 500°C are shown in fig.6.8. The profiles do not change very dramatically over the ageing time range which would be expected due to the small size of the precipitates in these specimens (chapter 5). The activity at low fields is shown in fig.6.9, as a function of $t_a^{1/2}$. The activity increases after only 20 minutes and then it decreases slowly up to ~500 hours before increasing again after the longest ageing times.

Fig.6.10 shows B.E. activity profiles for the specimens aged at 630°C. After 100 hours there is a large increase in the activity, particularly at low fields resulting in a central peak, implying a precipitate diameter which is comparable in size to the domain wall width. In fig.6.11 the magnitude of the low field activity is plotted as a function of t_a , showing >200% increase in the activity after 100 hours.

M.A.E. activity profiles for each ageing time at 500°C are shown in fig.6.12. The two peak heights and the low field activity are shown in fig. 6.13 as a function of $t_a^{1/2}$. There is a significant increase in the initial and final peak amplitudes after 20 minutes (19 and 20% respectively) but the activity follows no clear trend for longer ageing times. The low field activity does not change significantly with ageing time.

Fig.6.14 shows the M.A.E. activity profiles for the specimens aged at 630°C. There is a general increase in the activity at all fields and in

fig.6.15, where the low field activity and the two peak amplitudes are plotted, as a function of t_a , we observe a 5% increase in the low field activity after 10 hours and a 35% increase after 100 hours, The two high field peak heights show even larger increases.

6.11.1.3 Barkhausen Event Size Distributions.

The Barkhausen event size distribution for an unaged specimen is shown in fig.6.16 (± 300 mA range). The total number of events plotted is 15300. The total count as a function of $t_a^{1/2}$ is shown in fig.6.17. As in the case of the B.E. activity profiles there is a large increase after just 20 minutes ageing followed by a gradual reduction almost back to its original value after several hundred hours. For the longest ageing time the total count increases dramatically to 47,600 as the precipitates coarsen. In fig.6.18 the threshold intercept is plotted as a function of $t_a^{1/2}$. This shows the same trend as for the total count and indicates that the Barkhausen pulses are a minimum size after ~500 hours.

6.11.2 The Iron-0.2%Copper Alloy System.

6.11.2.1 Hardness Measurements.

Fig.6.19 shows the hardness changes induced by neutron irradiation, together with the effects of 290°C-thermal ageing and isochronal annealing. These data demonstrate that irradiation produces marked strengthening with a hardness increase >100%, and that maximum recovery of the hardness change occurs over the range 550°-650°C. The control specimens exhibit a small hardening peak centred at ~550°-600°C. The discs aged at 500°C showed no significant change in hardness over the whole 1,600 hour period.

6.11.2.2 Barkhausen and Magneto-acoustic Emission Profiles.

Fig.6.20 shows the B.E. activity profiles for one of the irradiated specimens. After the neutron irradiation the profile has a very large sharp initial peak and at low fields the curve is smoothed implying

homogeneous pinning of 180° domain walls. After the progressive anneals the profile starts to revert to an irregular shape at low fields but the sharp initial peak remains even after the 650°C anneal. The amplitude of the two peaks and the activity at zero field is plotted in fig.6.21 (the data being averaged for the three specimens). The high field peaks are much larger after irradiation, but then they become progressively smaller after each anneal until at 550°C the final peak is back to its pre-irradiation size. At the higher temperatures the two peaks grow slightly larger again. The low field activity increases slightly after irradiation and then gently increases until after the 500°C anneal. At 550°C there is a significant decrease in the activity (39%), followed by a further increase (27%) at 600°C and a decrease at 650°C .

The M.A.E. activity profiles for one of the irradiated specimens is shown in fig.6.22. These are similar to the iron-0.7% copper M.A.E. activity profiles except the outer peaks are less well pronounced. Fig.6.23 shows the same three parameters as for the B.E. activity profiles. These results are similar in their overall characteristics to those of the B.E. activity (fig.6.21). The main differences are:

- 1) The outer peak/central dip ratio is much smaller (usually the case for M.A.E. activity profiles).
- 2) The increase in the outer peak height amplitudes after irradiation is much smaller, and the low field activity is actually reduced.
- 3) The peaks at 500° and 600°C occur for all three parameters, not just in the low field region.

Fig.6.24 shows the B.E. activity profiles for one of the control specimens. After the 290°C heat treatment for 5 days the activity has increased quite dramatically at all fields, particularly in the initial peak region. However, unlike the irradiated material, the profile is jagged implying inhomogeneous pinning. After the progressive anneals the profile

almost reverts back to its original shape. The amplitude of the two outer peaks and the activity at low fields is plotted in fig.6.25. All three of these parameters decrease monotonically as a function of the annealing temperature.

The M.A.E. activity profiles for one of the control specimens is shown in fig.6.26. Again the activity increases at all fields after the 290°C heat treatment. In order to see the trends more clearly fig.6.27 shows the initial peak, low field and final peak amplitudes (the M.A.E. profile amplitudes were adjusted slightly in order to allow for small changes in the transducer coupling). Besides the initial increase after 5 days at 290°C we observe a significant increase at 500°C followed by a decrease at 550°C and a further increase at 600°C. This is similar to that observed for the irradiated specimens although the increases are not so large (from 450°-500°C the increase in the final peak amplitude is 9.6% as compared to the 15.7% observed for the irradiated material and from 550°-600°C it is 8.2% as compared with 19.2%). After the 650°C anneal the activity is reduced although it does not revert back to its original pre-irradiation value.

Figs.6.28 and 6.29 shows some of the B.E. and M.A.E. profiles obtained from specimens heat treated at 290°C for 1, 10 and 120 hours. From these profiles it is clear that the increased activity is not due to precipitation since it is greatly increased after only 1 hour. The activity from the pure iron control treated for 10 hours at 290°C is similarly increased, fig.6.30, and so the effect is not due to the presence of the copper.

In an attempt to simulate the effects of neutron irradiation (an increased copper mobility due to a greater vacancy concentration) specimens were heat treated at the higher temperature of 350°C for 120 hours. The B.E. and M.A.E. profiles before and after this treatment are shown in fig.6.31. We observe increased M.A.E and B.E. activity which is similar to the profiles aged at 290°C. At low fields the profiles are jagged implying

inhomogeneous pinning, and so the effects of irradiation are quite unlike thermal effects.

6.11.2.3 Barkhausen Event Size Distributions.

The average total Barkhausen event count before and after irradiation using both current ranges is shown in table 6.3. The increase in the total number of counts and the threshold intercept (not shown) after irradiation measured from the whole current range implies an increase in the pinning density. The total number of counts measured from the ± 300 mA range is reduced.

6.12 Discussion.

6.12.1 The Effect of Copper in Solution.

The solution treated material had a mechanical hardness of 64 and 85 for 0.2 and 0.7% copper respectively, indicating a solution hardening effect. On comparing figs.6.8, 6.12, 6.24, 6.26 and 6.30 we see that the addition of copper produces increases in both the B.E. and the M.A.E. at all fields. For example for 0.0, 0.2 and 0.7% copper in solution the B.E. low field activity is 0.30, 1.29 and 2.20 mV and for the M.A.E. it is 2.08, 2.71 and 3.51 mV respectively. So the M.A.E. and particularly the B.E. activity are very sensitive to copper in solution. The domain wall pinning is much more severe as is directly indicated by a >120% increase in the total Barkhausen count for only 0.2% copper in solution (compared to a pure iron specimen).

6.12.2 The Iron-0.7% Copper Alloy System.

The characteristic age hardening curve at 500°C (fig.6.7) shows peak strengthening after ~100 hours whereas the B.E. activity (fig.6.9) shows a 60% increase after only 20 minutes followed by small reductions up to several hundred hours. The increase after 20 minutes was unexpected but is believed to be due to the reduction in the vacancy concentration quenched

in at 850°C (see next section), or possibly due to stress relief at 500°C. Besides this early increase the low field B.E. activity is roughly inversely related to the mechanical hardness. The large increase at 1,700 hours indicates that the particle size is becoming large enough to cause domain walls to become pinned at individual inclusions (chapter 5). A similar weaker trend is evident for the M.A.E. activity outer peak amplitudes (fig.6.13) for ageing times less than 400 hours. However after 1,700 hours the activity does not increase. This was found to be the case in the Incoloy 904 alloy. In that study the M.A.E. was found to be relatively insensitive to the presence of precipitates with diameters smaller than the domain wall width, whereas the B.E. activity was markedly increased for precipitate sizes $>10\text{nm}$. Thus is consistent with Goodman et al.'s measurements (1973a). They reported particle diameters of 1.6, 2.4, 6, 10 and 15nm for ageing times of 1, 3, 10, 50 and 120 hours at 500°C in an Fe-1.4%Cu alloy. So, other than the large increase in B.E. and M.A.E. activity after ageing for 20 minutes, the precipitation of a large density of small copper precipitates is reflected by a reduction in the B.E. low field activity and total count (for the low field region, $\pm 300\text{mA}$ range) and a similar reduction in the threshold intercept. But the M.A.E. is relatively unaffected. For ageing times above 1,000 hours, however, the B.E. activity is markedly increased, and this is similar to that observed in the Incoloy 904 alloy after ageing for a few hours at 700°C.

On ageing at 630°C we are in the regime where the particles are of the order of the domain wall width (full wall width $\sim 60\text{nm}$ in iron). Hornbogen (1964) reports particle diameters of 15 and 30nm for an Fe-0.9%Cu alloy aged for 10 and 100 hours respectively at 600°C, and a particle diameter of 80nm for the same alloy aged for 96 hours at 700°C. These larger precipitates strongly pin the 180° domain walls giving rise to a peak in the low field B.E. activity profile after 100 hours (fig.6.10). The 90° domain wall activity is also increased as indicated by the increase in the

M.A.E. activity at all fields (figs.6.14 and 6.15). This is again similar to the trend observed in the Incoloy 904 alloy on ageing. In this iron-copper alloy, however, the ageing time is not long enough to reach peak activity. This data implies that the 600°C anneal in the Fe-0.2%Cu system will be important in detection of copper precipitates.

6.12.3 The Iron-0.2% Copper Alloy System.

6.12.3.1 Neutron Irradiation and the 290°C Heat Treatments.

After the neutron irradiation the hardness increased by 70 points. (fig.6.19). This radiation hardening is well established and is understood in terms of the formation of point defect clusters and/or copper precipitates (section 6.7). The hardness of the control specimens was not significantly changed after 120 hours at 290°C.

The increase in the M.A.E. activity (figs.6.26 and 6.27) and to an even greater extent the B.E. activity (figs.6.24 and 6.25) at all fields after the 290°C heat treatment was an unexpected result. In order to investigate this effect heat treatments were carried out for 1 and 10 hour periods. The M.A.E. and B.E. activity almost reached their maximum after only 1 hour (figs.6.28 and 6.29) and this result together with the inhomogeneous pinning implied by the jagged profile at low fields indicates that the effect was not due to copper precipitation. The similar increase observed in the pure iron specimen after 10 hours (fig.6.30) confirmed this. One explanation of this phenomena is that the vacancy supersaturation quenched-in at 850°C effects the domain wall motion causing the walls to make smaller jumps. This vacancy concentration will be greatest at the surface of the specimen (subject to maximum quench rate) and this is reflected in the surface sensitive B.E. activity, but it is not understood how the vacancies modify the domain wall motion.

The irradiated specimens gave increased B.E. activity at all fields (figs.6.20 and 6.21) but the shape of the profile was quite different than for the control specimens. The smooth shape implies homogeneous pinning of

180° domain walls. The 37% increase at low fields in the B.E. activity was not mirrored for the M.A.E. and this result together with the above is consistent with the formation of a high density of small copper precipitates (i.e. this was observed to occur in the Fe-0.7%Cu alloy, section 6.12.2). However, there is also the possibility of a large density of microvoids giving rise to these effects. The accompanying reduction in the low field Barkhausen event total count (table 6.3) and the threshold intercept confirm these possible interpretations (section 6.11.2.3).

The specimens aged at 350°C for 120 hours did not produce B.E. and M.A.E. profiles like those after irradiation indicating (fig.6.31) that the simple increase in vacancy concentration achieved during irradiation is not the only cause for radiation embrittlement.

The large increase in the B.E. outer peak amplitudes (figs.6.20 and 6.21) was not observed in the thermally aged Fe-0.7%Cu alloy and is probably not due to the presence of small precipitates. The earlier work on the effects of plastic deformation of pure iron (chapter 4) on B.E. and M.A.E. also gave large increases at these higher field regions. Consequently these increases may be due to the presence of a high density of dislocation loops formed during irradiation. The isochronal annealing will give further insight into this.

6.12.3.2 Isochronal Annealing.

The monotonic decrease in the B.E. initial and final peak and the low field amplitudes (figs.6.24 and 6.25) in the control specimens as a function of annealing temperature did not occur in the M.A.E. activity (fig.6.27). This may be because the effect is limited to a thin surface layer (section 1.4). If the increased quenched-in vacancy concentration at the surface also causes copper micro-clustering (being too small to be detectable by M.A.E.), this may be unstable and dissolve at successively higher temperatures resulting in the observed reduction in B.E. activity.

There are significant changes in both the B.E. and M.A.E. profiles in the irradiated material on annealing (figs.6.20, 6.21, 6.22 and 6.23). The steep increase in M.A.E. activity over the temperature range 400°-500°C together with the well defined peak at 500°C followed by the drop at 550°C (fig.6.23) is analogous to the effects of cold work (chapter 4). This implies a mechanism based on the annealing out of dislocation loops. The B.E. outer peaks show a gradual reduction up to 550°C (fig.6.21) which is also probably due to the annealing out of dislocation loops. The reduction in the B.E. low field activity amplitude at 550°C may be due to one or both of the following mechanisms. First, it could again be due to annealing out of dislocations. (The increase in B.E. at 500°C from its solution value is 52.3% and the same parameter for M.A.E. is 50.3%). Second, the growth of small precipitates formed during irradiation would result in a reduction in the low field B.E. activity (1 hour at 550°C corresponding to 20-100 hours at 500°C, see section 6.12.2). The strong peak at 600°C in the M.A.E. initial and final peak and low field amplitudes together with the peak in the B.E. low field amplitude are consistent with the coarsening of copper precipitates formed during irradiation. This implies that the increase in the B.E. observed after irradiation was due to copper precipitation rather than to microvoids. As in the case of the 630°C thermal ageing (sections 6.11.1.2 and 6.12.2), we are in the regime where each precipitate has a strong pinning effect on the domain walls giving rise to increased B.E. and M.A.E. activity at all fields. The increase in the low field B.E. and M.A.E. activity at 600°C, from the solution value, is similar (39.9 and 38.6% respectively). However, the increase is not sufficient to give rise to a central peak. Clearly, the precipitate sizes responsible for peak B.E. and M.A.E. activity do not correspond to that for peak strengthening in this material. Thus, precipitate coarsening at 600°C leads to a loss of hardening, but enhanced domain wall pinning as the particle diameter to domain wall width approaches unity.

The control specimens also showed smaller less pronounced peaks at

500° and 600°C in the M.A.E. activity (fig.6.23). This implies a low dislocation density present, possibly due to the quench from 850°C, and some limited copper precipitation on annealing.

6.13 Conclusions of Iron-Copper Alloy Study.

The results of the B.E. and M.A.E. studies imply that two independent irradiation strengthening mechanisms are operative in Fe-0.2%Cu alloy after 290°C-neutron irradiation, one based on dislocation loop formation and a second attributable to copper precipitation. The effect of dislocation loops on B.E. and M.A.E. appears to be similar to that of dislocations as discussed in chapter 4. The M.A.E. technique appears to be capable of detecting and distinguishing between domain wall pinning caused by both types of obstacle, but only during post-irradiation annealing. This is a consequence of the dependence of the sensitivity of the M.A.E. activity on the size of defect relative to the domain wall width as was found in chapter 5. The B.E. technique is more sensitive to small defects (as was the case for precipitates in Incoloy 904, chapter 5) and revealed the presence of the precipitates in the as-irradiated condition. Correlations between B.E., M.A.E. and other detection methods capable of characterising matrix defects at sizes < 1-2nm (e.g. small angle neutron scattering) could be useful in further validating the above interpretation and providing a calibration of the technique. At present no nondestructive examination technique exists and B.E. and M.A.E. show potential for use as a tool in monitoring embrittlement levels in nuclear pressure vessels.

Table 6.1

The ageing times at (a) 500^o and (b) 630^oC for Fe-0.7%Cu.

(a)

<u>Ageing time (hours)</u>	<u>No. of specimens</u>
0	2
0.33	2
8	2
39.5	2
144	2
785	2
1673	2

(b)

<u>Ageing time (hours)</u>	<u>No. of specimens</u>
(0)	(2)
1	1
10	2
100	2

Table 6.2
 The ageing times at 500°C for Fe-0.2%Cu.

Ageing time (hours)	No. of specimens
0	1
1	1
4	1
10	1
24	2
34	2
57	2
100	2
169	2
242	2
386	2
674	2
942	2
1346	2
1706	2

Table 6.3
 The total Barkhausen count before and after neutron irradiation for ± 300 and $\pm 2,400$ mA ranges for Fe-0.2%Cu.

	± 300 mA range	± 2400 mA range
Before irradiation	12242	134486
After irradiation	7540	153355

(1) Before n-Irradiation

(2) After n-Irradiation

Figure 6.1 The B.E. activity profiles. (1) Before neutron irradiation for (a) furnace cooled, (b) water quenched and (c) cold worked alpha-iron. (2) After neutron irradiation for (d)-(f) as (a)-(c) respectively.

Figure 6.2 The Barkhausen event size distributions for (a) furnace cooled, (b) water quenched and (c) worked worked alpha-iron.

Figure 6.3 The B.E. activity profiles for the cold worked alpha-iron before and after neutron irradiation and after each post-irradiation anneal.

Figure 6.4 The effects of neutron irradiation and subsequent isochronal annealing on the low field B.E. amplitude for the cold worked alpha-iron. The dotted curve shows this data for the unirradiated material investigated in chapter 4 (the values being normalised to allow for the different specimen areas).

Figure 6.5 The effects of neutron irradiation and subsequent isochronal annealing on the M.A.E. initial peak / low field ratio for the furnace cooled alpha-iron. The inset shows a typical M.A.E. activity profile obtained from this material.

Figure 6.6 The effects of neutron irradiation and subsequent isochronal annealing on the M.A.E. initial peak / final peak ratio for the cold worked alpha-iron. The dotted curve shows this data for the unirradiated material of chapter 4.

Figure 6.7 The room temperature hardness as a function of $t_a^{1/2}$ for Fe-0.7%Cu aged at 500°C.

Figure 6.8 Several of the B.E. activity profiles for Fe-0.7%Cu aged at 500°C.

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Figure 6.11 The B.E. low field amplitude as a function of the ageing time at 630°C for Fe-0.7%Cu.

Figure 6.12 The M.A.E. activity profiles for Fe-0.7%Cu aged at 500°C.

Figure 6.13 The M.A.E. initial peak, low field and final peak amplitudes as a function of $t_a^{1/2}$ for Fe-0.7%Cu aged at 500°C.

Figure 6.14 The M.A.E. activity profiles for Fe-0.7%Cu aged at 630°C.

Figure 6.15 The M.A.E. initial peak, low field and final peak amplitudes as a function of the ageing time at 630°C for Fe-0.7%Cu.

Figure 6.16 An example of a Barkhausen event size distribution for an Fe-0.2%Cu specimen. This was measured over the smaller range of ± 300 mA for solution treated material.

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Figure 6.18 The threshold intercept as a function of $t_a^{1/2}$ for Fe-0.7%Cu aged at 500°C.

Figure 6.19 The effects of neutron irradiation, thermal ageing and isochronal annealing on the room temperature hardness of Fe-0.2%Cu.

Current / A

Current / A

Figure 6.20 The B.E. activity profiles for Fe-0.2%Cu before and after neutron irradiation and after isochronal anneals.

Figure 6.21 The effects of neutron irradiation and isochronal annealing on the initial peak, low field and final peak amplitudes of the B.E. activity profiles of Fe-0.2%Cu.

Figure 6.22 The M.A.E. activity profiles for Fe-0.2%Cu before and after neutron irradiation and after isochronal anneals.

Figure 6.23 The effects of neutron irradiation and isochronal annealing on the initial peak, low field and final peak amplitudes of the M.A.E. activity profiles of Fe-0.2%Cu.

Figure 6.24 The B.E. activity profiles for Fe-0.2%Cu before and after 290°C heat treatment for 120 hours and after isochronal anneals.

Figure 6.25 The effects of the 290°C heat treatment and isochronal annealing on the initial peak, low field and final peak amplitudes of the B.E. activity profiles of Fe-0.2%Cu.

Figure 6.26 The M.A.E. activity profiles for Fe-0.2%Cu before and after 290°C heat treatment for 120 hours and after isochronal anneals.

Figure 6.27 The effects of the 290°C heat treatment and isochronal annealing on the initial peak, low field and final peak amplitudes of the M.A.E activity profiles of Fe-0.2%Cu.

Figure 6.28 The B.E. activity profiles for Fe-0.2%Cu aged for (a) 1, (b) 10 and (c) 120 hours at 290°C.

Figure 6.29 The M.A.E. activity profiles for Fe-0.2%Cu aged for (a) 1, (b) 10 and (c) 120 hours at 290°C.

Figure 6.30 (a) B.E. and (b) M.A.E. activity profiles for pure iron before and after 10 hour heat treatment at 290°C.

Figure 6.31 (a) B.E. and (b) M.A.E. activity profiles for Fe-0.2%Cu before and after 120 hour heat treatment at 350°C.

Chapter 7. The Dependence of Barkhausen and Magneto-acoustic Emission upon Tensile Stress in Various Materials.

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7.1 Introduction.

There is at present a strong motivation to develop nondestructive measurement techniques such as B.E. and M.A.E. for the measurement of residual stresses (Donaldson and Pasley, 1967; Gardner et al, 1971; Altpeter et al., 1981; Kusanagi et al. 1979a,b; Shibata and Ono, 1981). However the relationships of both B.E. and M.A.E. to stress and microstructure are complex and not yet sufficiently well understood to separate stress and microstructure effects. This stress-microstructure dependence is not peculiar to this technique, the ultrasonics technique of measuring acoustic velocity also suffers from this (Ruud, 1982).

In this chapter the stress dependence of B.E. and M.A.E. activity is measured from a number of materials. In order to do this a new magnetising solenoid was constructed (section 2.1.2). This enabled measurements to be made at various tensile loads 'in situ'. The B.E. activity measurement technique was also modified. Only a few different materials were investigated and so the results presented in this chapter do not achieve the aim of being able to separate the microstructure from the stress effects. However they demonstrate that different materials and microstructures give rise to a different stress dependence. In chapter 8 one particular alloy is investigated in great detail in order to find suitable parameters for the measurement of stress.

7.2 Laser Calibration and Specimen Gripping.

When a specimen is under tensile stress there is the possibility of the acoustic signal changing due to a change in the clamping (Edwards et al., 1985). This can arise because the longitudinal resonant modes change due to a change in the effective length of material supporting the acoustic waves (section 3.2.1). In order to investigate this the acoustic coupling as a function of the applied tensile stress was measured using the arrangement shown in fig.7.1. A 13 mJ Neodymium-YAG laser was used as a standard acoustic source (Scruby et al., 1981). This produced a 10 ns pulse

of 1.06 μm radiation which was directed onto the side of a mild steel specimen mounted in the tensile testing machine. The acoustic signal was detected, using the 2.5 MHz damped piezo-electric transducer, and amplified and filtered in the usual way. The transient recorder was used in the pre-trigger mode in order to record the signal. Two bandwidths were used; the first of these was the largest obtainable i.e. 4.4 kHz-7.0 MHz (3 dB's), and the second was the same as that used for M.A.E. activity profile measurements i.e. 32-100 kHz (3 dB's). Measurements were not made to d.c. as large low frequency noise components made signal recording difficult. A typical signal obtained using both of these bandwidths is shown in fig.7.2(a,b). The amplitude of the first peak in the waveform was taken as a relative measure of the coupling. This amplitude together with the initial peak, central dip and final peak of the M.A.E. activity profiles obtained from the specimen as a function of the applied tensile stress is shown in figs.7.3 and 7.26. We observe that, although the M.A.E. initial peak amplitude decreases by ~50% at 205 Mpa load, the coupling does not change significantly. This implies that no calibration will be required in order to measure the M.A.E. activity as a function of the tensile stress.

The reason for this consistency in the coupling may be due to the method of gripping the specimen. i.e. measurements made by Edwards et al. (1985) used threaded specimens which meant that specimen gripping increased as the stress was increased. But in the arrangement used here the specimens were pre-tightened using collets so that specimen gripping did not change significantly with stress. There are however two drawbacks to this method of specimen support. First, the maximum stress which can be applied is limited by the collet gripping to ~200 MPa. Second, the ends of the specimen are under a constant compressive stress which may modify the results obtained. However simple cylindrical specimens can still be used and, as shown above, the acoustic coupling is independent of the applied stress.

7.3 The Effect of Stress on Domain Structure.

When materials are under elastic strain the preferred domain orientations are modified due to the effect of magnetostriction. In order for this to occur the applied stress must be comparable to the forces maintaining the alignment of the domains in the unstrained material. These forces are mainly due to the crystal anisotropy and therefore we require (Bozorth, 1951);

where λ is the magnetostriction constant.

$$\frac{3}{2} \lambda \sigma \sim k \quad \sigma \text{ is the applied stress.} \quad (\text{Eq}^n.7.1)$$

k is the first anisotropy constant.

Consequently for materials with a positive magnetostriction, the application of a tensile stress will cause the domains magnetised in directions closest to the stress direction to grow at the expense of neighbouring domains, in order to reduce the elastic energy. In the present experimental arrangement the magnetisation direction is parallel to the stress axis. Consequently as the tensile stress is increased in combination with a periodic magnetic field, there is a greater tendency for domains with their magnetization direction parallel to the stress axis, and therefore 180° domain wall movements, to become dominant. For materials with a negative magnetostriction the magnetic moment of the domains will tend to align close to 90° to the stress axis at low fields. This may give rise to more non- 180° domain walls although the effects may be more complex in a cycling magnetic field.

The magnetostriction, λ , will also depend on the domain distribution and therefore also be a function of stress. λ is given by;

$$\lambda = \frac{3}{2} \lambda_s \cdot (\{\cos^2 \theta\}_{av} - \{\cos^2 \theta\}_o) \quad (\text{Eq}^n.7.2)$$

where $\{\cos^2 \theta\}_o$ is the initial distribution of angles of the domain moments and $\{\cos^2 \theta\}_{av}$ is the distribution at any time, where θ is the angle between the direction of magnetisation and the direction in which the magnetostriction is measured at that time.

A large tension will orientate the domains at 90° to the stress axis

in nickel. Consequently when a field is applied the magnetisation changes the average of $\cos^2\theta$ from 0 to 1 so that λ is $\frac{3}{2}\lambda_s$ (Bozorth, 1951). Fig.7.4 shows the saturation magnetostriction for nickel as a function of both compressive and tensile stresses (Kirchner, 1936). For nickel $\frac{2 \cdot k}{3 \lambda} = 80 \text{ MPa}$. Fig.7.4 indicates that a stress of this value is able to change the saturation magnetostriction most of the way to its high stress value of 60×10^{-6} .

In addition to the changes in the domain wall structure which is most easily observed as a change in the magnetostriction constant on application of stress there is the possibility of changes in the domain volumes. Smaller domains will give rise to much smaller M.A.E. and B.E. signals (section 5.6.3).

Consequently in materials of positive magnetostriction such as steel, iron and Incoloy 904 the low field B.E. may be expected to increase and the M.A.E. to decrease as a function of increasing stress (fig.7.5), since domains parallel to the stress axis are favoured even at zero applied field, resulting in less 90° domain walls (predicted by Bloch (1977), and Theiner and Altpeter (1983)). In negative magnetostriction materials such as nickel the low field B.E. may decrease and the M.A.E. increase as a function of increasing tensile stress. However, these predictions neglect changes in the domain volumes.

7.4 Measurements.

7.4.1 Barkhausen Emission Activity Profiles.

Only a single detection coil was used for Barkhausen measurements (section 2.1.8) as it was not possible to locate a second coil above the specimen. This meant that there was additional noise across the coil which was mostly from the coil driving circuit (this would have been rejected in the two coil system). In order to reduce this a modified driving circuit was constructed (fig.7.6). The output of the function generator was filtered using a six-pole 5.2 Hz low pass Bessel filter (table 2.4) which

then controlled the current through the solenoid via a 2 A power buffer. The B.E. activity profile was then measured in the usual way (section 2.2.3). The current range chosen for the B.E. activity profile measurements was ± 1.35 A which was sufficient to include the whole field range where B.E. was found to occur. The magnetisation rate was $385 \text{ Am}^{-1}\text{s}^{-1}$ (5.0 mHz) and the signal envelope was obtained by using the 300 mHz filter.

7.4.2 Magneto-acoustic Emission Activity Profiles.

These measurements were made in the usual way (section 2.3.3) using a current range of ± 1.6 A and a magnetisation rate of $146 \text{ Am}^{-1}\text{s}^{-1}$. The current range was larger than that for the B.E. measurements because some M.A.E. was found to occur at higher fields. The use^{of} different current ranges for the B.E. and M.A.E. measurements was not ideal but it was necessary to use the minimum current range for B.E. in order to reduce (a) noise and (b) overheating of the driving circuit components. M.A.E. activity profile measurements were made using method 2 with a 25 Hz modulation field of 150 mA, and the signal envelope derived using a 300 mHz filter.

7.4.3 Materials.

The specimens used were all cylinders of length 40mm and diameter 2.3mm. The materials investigated were as follows;

- (1) Mild steel (as recieved).
- (2) Mild steel (annealed at 700°C for 18 hours). Yield point 354 MPa.
- (3) Pure iron (cold-worked).
- (4) Pure iron (annealed at 850°C for 18 hours). Yield point 96 MPa.
- (5) Pure nickel (as recieved).
- (6) Pure nickel (annealed at 1200°C for 18 hours). No yield point.
- (7) Incoloy 904 (solution treated).
- (8) Incoloy 904 (solution treated and aged at 700°C for 343 hours).

The magnetostriction constants for some of these materials are listed in

table 7.1. The yield point for specimens (2), (4) and (6) was measured by machining tensile specimens with dimensions as shown in fig.4.1, and then loading them in the 5 kN straining machine.

B.E. and M.A.E. activity profiles were measured for specimens of these materials as a function of the applied tensile stress and the peak heights measured in the usual way. (B.E. measurements were not made for annealed nickel).

7.5 Results and Discussion.

7.5.1 Barkhausen Emission Activity Profiles.

7.5.1.1 Profile Shape.

Figs.7.7 to 7.11 show the B.E. activity profiles as a function of the applied tensile stress for the materials (1)–(5) above. The 'as received', cold-worked and aged materials ((1),(3),(5),(8)) tend to have a fairly large pinning density resulting in a large central peak at low fields and smaller peaks or ledges at high fields close to the 'knees' of the hysteresis loop (figs.7.7, 7.9, 7.11 and 5.24). The annealed and solution treated material ((2),(4),(6),(7)) have a low pinning density resulting in low activity at low fields and two peaks in the activity at higher fields (figs.7.8, 7.10, 7.12 and 5.24(0 hours)). i.e. as discussed in sections 4.6.1 and 5.6.2 the low field activity is controlled by the pinning of 180° domain walls on the obstacles, whilst the activity at higher fields is more complex.

7.5.1.2 Stress Dependence.

In general the B.E. activity profiles show fairly small changes as a function of stress. Figs.7.13–7.18 show the peak height amplitudes as a function of the applied stress for the materials (1)–(5) and (8).

Consider first the low field activity. The only materials investigated which show a monotonic dependence upon stress is Incoloy 904 and cold-worked iron (figs.7.18 and 7.15). In these materials the low field B.E.

activity increases as a function of applied stress as predicted. In the case of 'as received' mild steel the sensitivity to stress falls off above 40 MPa at all fields (this could also be the case in iron since measurements were not made above 25 MPa). The annealed mild steel and iron (figs.7.14 and 7.16) show a quite different dependence upon tensile stress. In these materials the pinning density is low. The activity decreases with increasing stress, although the large decrease observed in the mild steel specimen for stresses below 10 MPa (fig.7.14) is associated with the removal of a jaggedness from the activity profile (fig.7.8). The absence of definite obstacles to domain wall pinning is known to give rise to inhomogeneous pinning (chapter 6) which causes a jagged profile. This implies that the application of stress causes the domain wall pinning to become more homogeneous. Hajko et al. (1973) suggests that, on loading, stress induced pinning sites form which are stronger than the existing pinning sites. A uniform distribution of such pinning sites could well account for the reduction in the low field activity in annealed mild steel by giving rise to much smaller Barkhausen jumps. This is a similar effect to that observed on irradiation of the iron-copper alloy (sections 6.4.2.2 and 6.5.2.1) where it is believed that a high density of copper precipitates and dislocation loops form.

The 'as received' mild steel shows a reduction in the initial peak height as a function of stress (fig.7.13). The activity from these outer peaks is thought to be largely due to 90° domain wall movements (chapters 4 and 5) and so we may expect these to behave in a similar way to that of the M.A.E. outer peak amplitudes.

The 'as received' nickel specimen shows a strong decrease in activity with increasing stress for stresses above 10 MPa (fig.7.17). This is in agreement with the expected trend (section 7.3) for negative magnetostriction materials. The increase in activity with stress up to 6 MPa, which also occurred in the acoustic measurements, is not understood.

7.5.2 Magneto-acoustic Emission Activity Profiles.

Figs.7.19 to 7.26 show the M.A.E. activity profiles as a function of the applied tensile stress for the materials (1)-(8) above. All the positive magnetostriction materials show a decrease as a function of stress as expected. The 'as received' nickel specimen (fig.7.23) showed a more complicated trend where the activity increases for low stresses the central peak reaching a maximum at ~30 MPa before decreasing again. (Kusanagi et al (1979) also reported this activity trend in nickel). The M.A.E. activity profiles for annealed nickel (fig.7.24) show a progression from a double to a single peak profile over a small stress range. On removing the load (the last activity profile shown), the activity increases again but remains as a single peak. This indicates that the specimen was plastically deformed on loading resulting in increased pinning at low fields. Annealing of plastically deformed polycrystalline iron also reduced the central peak with respect to the initial peak although the low field activity was not sufficiently large to produce a peak in the profile (fig.4.13). These results suggest that the initial peak / central dip amplitude could be used to monitor the plastic deformation (c.f. the same parameter measured from the B.E. profiles in cold-worked iron as a function of annealing temperature, fig.4.10, supports this).

Figs.7.27 to 7.34 show the initial peak, central dip and final peak as a function of the applied tensile stress for each of the materials. The specimens with the higher pinning density ((1),(3),(5) and (8)) show the strongest dependence upon stress, although in the cases of annealed mild steel and nickel the stress range investigated was smaller than that for the corresponding 'as received' specimens. This was because the material elastic limit is much lower in the annealed material. However, from figs.7.33 and 7.34, it is apparent that the microstructure (in this case non-magnetic inclusions) modifies the stress dependence quite considerably, particularly for stresses <20 MPa, in this case the aged material showing the ^rstrongest dependence.

Table 7.2 shows the change in the low field activity at 23.6 MPa from the zero stress value in the four materials with the higher pinning density. Both the B.E. and M.A.E. measurements are shown. This table indicates that the dependence of M.A.E. and B.E. on stress is entirely material dependent.

7.7 Summary and Conclusions.

The results can be summarised as follows;

(A) Positive magnetostrictive materials.

(1) The B.E. dependence on stress is complex and generally quite weak though in many materials the low field activity increases with stress.

(2) The M.A.E. activity always monotonically decreases with increasing stress.

(B) Nickel.

(1) Both the B.E. and M.A.E. activity increase with increasing stress for small stresses (<10 MPa for B.E. and <30 MPa for M.A.E.).

(2) For higher stresses both the B.E. and M.A.E. activity decrease.

For elastic strains B.E. tends to have a more complex dependence than M.A.E. and is of course limited to a small surface layer (10-100 μm). Although the magnetisation process used in this M.A.E. experiment involves a 25 Hz frequency component, thereby limiting measurements to a ~0.1-1.0 mm surface layer, measurements could be made using method 1 (section 3.2.2).

From the results presented in this chapter it is clear that further work is necessary in order to be able to separate microstructural from stress effects. In the following chapter an extensive study is made on a mild steel alloy in a number of different heat treated conditions. This should determine the extent to which the state of stress can be inferred from B.E. and M.A.E. measurements.

Table 7.1

The magnetostriction constants for iron, nickel
and Incoloy 904.

Material	λ_{100} ($\times 10^{-6}$)	λ_{111} ($\times 10^{-6}$)
Iron	20.7	-21.2
Nickel	-45.9	-24.3
Incoloy 904	15.	?

Table 7.2

The change in the low field B.E. and M.A.E.
activity at 23.6 MPa from that at zero stress in
the materials with the higher pinning density.

	Iron	Steel	Nickel	Incoloy
B.E.	7%	22%	27%	40%
M.A.E.	-50%	-20%	7.6%	-5%

Figure 7.1 The experimental arrangement used to investigate the acoustic coupling as a function of applied tensile stress.

Figure 7.2 The acoustic signals obtained using a laser pulse source. (A) bandwidth 4.4 kHz to 7.0 MHz. (B) bandwidth 32 to 100 kHz. Note that the time scale is 10x greater in (B).

Figure 7.3 The amplitude of the first peak in the acoustic signal of fig.7.2 as a function of the applied tensile stress. The upper line is for the larger bandwidth. This result confirms that the acoustic coupling remains constant throughout the tensile loading experiment.

FIG.7.4 Magnetostriction of nickel measured under tension or compression. Note limits of zero and $3/2$ of magnetostriction at zero stress.

Figure 7.5 The predicted stress dependence of the low field B.E. and M.A.E. signals for positive magnetostriction materials.

Figure 7.6 The experimental arrangement used to drive the straining machine mounted solenoid for B.E. measurements.

Figure 7.7 The B.E. activity profiles at different applied loads for mild steel (ground finish with no heat treatments). The profiles here and throughout this chapter are put one above the other for clarity, the noise offset being negligible).

Figure 7.8 The B.E. activity profiles at different applied loads for mild steel heat treated at 700°C for 18 hours.

Figure 7.9 The B.E. activity profiles at different applied loads for cold-worked pure iron. This is one of the specimens of chapter 4 which has 4.9% plastic strain induced by tensile loading.

Figure 7.10 The B.E. activity profiles at different applied loads for pure iron annealed at 850°C for 18 hours.

Figure 7.11 The B.E. activity profiles at different applied loads for pure nickel (ground finish with no heat treatments).

i)

ii)

Figure 7.12 (A) B.E. and (B) M.A.E. activity profiles for pure nickel annealed for 18 hours at 1200°C.

Figure 7.13 The B.E. initial, central and final peak amplitudes as a function of the applied tensile stress for mild steel (ground finish with no heat treatments).

Figure 7.14 The B.E. initial peak, low field and final peak amplitudes as a function of the applied tensile stress for mild steel annealed for 18 hours at 700°C.

Figure 7.15 The B.E. initial, central and final peak amplitudes as a function of the applied tensile stress for cold-worked pure iron, plastically strained by 4.9%.

Figure 7.16 The B.E. initial peak, low field and final peak amplitudes as a function of the applied tensile stress for pure iron annealed for 18 hours at 850°C.

Figure 7.17 The B.E. central peak amplitude as a function of the applied tensile stress for pure nickel (ground finish with no heat treatments).

Figure 7.18 The B.E. central amplitude as a function of the applied tensile stress for polycrystalline Incoloy 904 aged at 700°C for 343 hours.

Figure 7.19 The M.A.E. activity profiles at different applied loads for mild steel (ground finish with no heat treatments). As was the case for B.E. the M.A.E. profiles are put one above the other here and throughout this chapter for clarity, the true noise offset for each profile being ≈ 52 mV.

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Chapter 8. The Dependence of Barkhausen and Magneto-acoustic Emission upon Tensile Stress and Microstructure in 4360 Steel.

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8.1 Introduction.

The ability to measure residual levels of stress in steel structures is an area of growing interest. The traditional methods used are hole drilling (Mathar, 1934) and ring coring (Wolf and Sauer, 1974), but these are relatively expensive and the techniques partially destructive. Several nondestructive techniques are presently being developed including X-ray diffraction (SAE, 1971; Maeder et al., 1981) and ultrasonic velocity (Crecraft, 1968; Green, 1973). The X-ray diffraction technique gives reliable results but is limited to surface measurements ($< 13 \mu\text{m}$) due to the 'soft' X-rays used. Ultrasound measurements, on the other hand, are generally made through the material thickness. However the velocity is generally dependent on a number of other parameters such as dislocation density and crystallographic texture and more fundamental research is required in order to be able to separate these effects (Ruud, 1982). It is worth mentioning, however, that some recent progress using the ultrasonic velocity technique (measuring velocity parallel and perpendicular to the principle stress axis) has enabled a stress measurement to be made independently of microstructural effects (Thompson et al., 1983; Lee et al., (in press)) Although some workers have used B.E. and M.A.E. to measure residual stresses (Karjalainen, 1980; Theiner et al., 1979), there is again the problem of being able to separate the microstructure and stress effects from the signals.

In this chapter B.E. and M.A.E. measurements are presented as a function of both microstructure and stress. The aim is to be determine the salient features of the signals characteristic of various microstructural parameters with a view to be able to use M.A.E. to measure stresses without prior knowledge of the microstructure. The different microstructures were obtained by homogenisation and tempering, normalisation, and full annealing heat treatments on the same material. Optical microscopy was used to observe these structures.

8.2 The Material.

The material used was 4360 steel. This is a mild steel (composition shown in table 8.1), used for general engineering components, such as crankshafts, sprockets and small arms parts. Fig.8.1 shows the isothermal transformation diagram for a steel of composition 0.5%C, 0.91%Mn (Woolman and Mottram, 1969). The Fe-C phase diagram (fig.8.2) indicates the appropriate temperature ranges for homogenisation, normalisation and annealing heat treatments (Krauss, 1980). These were subsequently chosen to be 1000°, 900° and 850°C respectively. These treatments can produce many different microstructures which could contain any of the following main forms; (a) martensite, (b) pearlite, (c) ferrite and (d) cementite. All of these can be expected to have different effects on the B.E. and M.A.E. signals which would complicate the determination of the stress value.

8.3 Specimen Preparation and Heat Treatments.

8.3.1 Preparation of Rod Specimens.

34 rods of length 40mm and diameter 2.3mm were machined from 5mm diameter rods of 4360 steel. The ends of these were then ground on 1200 grade silicon carbide paper to ensure good transducer coupling. All the rods were then heat treated in silica tube in vacuum as described in 8.3.3. There was generally two specimens for each heat treatment. They were then electropolished at 0°C for 30 seconds with a current of 300mA using a solution consisting of 10% Perchloric acid, 45% acetic acid, and 45% 2-butoxyethanol. This removed any contaminants arising during the heat treatments (particularly the dark oxide layer which formed on the water quenched specimens), and produced a good surface finish.

8.3.2 Preparation of Disc Specimens.

36 discs of diameter 5 mm and thickness ~0.6mm were cut from a rod using a high speed cutting wheel. These were then ground on silicon carbide papers to 1200 grade, before being heat treated in silica tube in vacuum as

described in section 8.3.3. The discs were then lightly polished and hardness measurements made. They were then mounted in transparent plastic blocks and the exposed surface reground in order to remove the indentations. Each specimen was then polished using 6µm and then 1µm diamond polish on polishing wheels. They were then etched in a 4% Nital solution for ~10 seconds and optical micrographs taken of the microstructure.

8.3.3 Heat Treatments of Rods and Discs.

8.3.3.1 Quenched and Isochronally Tempered Specimens.

Several of the rods and discs were homogenised at 1000°C for 1 hour in vacuum and then water quenched (the silica tube being broken on quenching). This ensured that the brittle martensitic structure was formed. The specimens were then cleaned as described above before being tempered for 100 minutes in vacuum and air cooled. The tempering temperatures for each specimen are listed in table 8.2.

8.3.3.2 Quenched and Isothermally Tempered Specimens.

Several more specimens were homogenised at 1000°C for 1 hour as in 8.3.3.1. These were then cleaned before being tempered at 650°C in vacuum and air cooled. The tempering times are listed in table 8.2. These heat treatments were done in preference to using a higher upper tempering temperature in section 8.3.3.1 in order to ensure that the austenitic phase did not form.

8.3.3.3 Normalised Specimens.

Other specimens were austenised in vacuum at 900°C for various times and air cooled. The times chosen are listed in table 8.2. This was done in order to produce a uniform structure of pearlite and ferrite.

8.3.3.4 Annealed Specimens.

A fourth group of specimens were heat treated at 850°C for various times (table 8.2) and furnace cooled. During the furnace cooling the temperature was reduced by 15°C every 15 minutes until at 640°C the furnace was turned off and the specimens left to cool in the furnace. This was done in order to produce a pearlite / ferrite structure as 8.3.3.3, but in this case the ferrite should be coarse-grained and the pearlite have a coarse interlamellar spacing.

8.4 Measurements.

8.4.1 Measurements at Zero Applied Tensile Stress.

B.E. activity profiles were made using the table mounted solenoid as described in section 2.1.3 using a current range of $\pm 2.4\text{A}$ ($\pm 21,600\text{Am}^{-1}$), and a magnetisation rate of $432\text{Am}^{-1}\text{s}^{-1}$ (5mHz). The activity envelope was obtained using a 300mHz filter and the X/Y plotter was set to 10mV/div on X.

M.A.E. activity profiles were made using method 2, the coil and the current range being the same as above. The experimental arrangement was as described in section 2.3.3 except that the signal was amplified a further $\times 10$ and the plotter sensitivity reduced by a factor of ten (10mV/div. on X). This was found to be more stable. The modulation field used was a 25Hz 200 mA rms triangular waveform, the higher amplitude being used because some of the specimens gave only a small signal. The slow sweep magnetisation rate was $128\text{Am}^{-1}\text{s}^{-1}$ (1.48mHz), and the signal envelope was filtered using a 300mHz filter.

Coercivity measurements were made as described in section 2.4. Hardness measurements were made on the disc specimens before they were polished and set in resin. These were then used to take optical micrographs of the microstructures obtained for each heat treatment.

8.4.2 Measurements as a Function of Applied Tensile Stress.

B.E. and M.A.E. activity profiles were measured as a function of tensile stress for most of the rod specimens using the experimental arrangement described in sections 7.4.1 and 7.4.2 respectively. For the B.E. measurements the current range was $\pm 1.3\text{A}$ ($H=\pm 20,000\text{Am}^{-1}$) and the magnetisation rate $400\text{Am}^{-1}\text{s}^{-1}$. This range was sufficient to include all the B.E. detectable, but does not go to specimen saturation. The M.A.E. measurements were made using a 25Hz 200 mA rms modulation field and a current range of $\pm 1.6\text{A}$ ($H=\pm 24,600\text{Am}^{-1}$). The slow sweep magnetisation rate was $146\text{Am}^{-1}\text{s}^{-1}$ (1.48mHz).

Measurements were taken with both increasing and decreasing load at various load values. Therefore two B.E. and M.A.E. activity profiles were measured at each load. (In some of the specimens a marked difference was observed between the activity profile obtained at any particular load after the load had been increased than for that obtained at the same load after the load had been decreased from a greater value. However this effect is not discussed in this thesis.)

8.5 Results and Discussion (I)-Microstructure Effects.

8.5.1 Isochronally Tempered Specimens.

8.5.1.1 Optical Micrographs.

The micrographs in figs.8.3 and 8.4 indicate the characteristic microstructure after each heat treatment. The as-quenched (untempered) material shows a typical martensitic structure. Many small plates ($6.6\mu\text{m}$ long, $1.0\mu\text{m}$ wide) are visible, which are characteristic of a cross section through martensite plates. After tempering at 150°C epsilon-carbides should form but these would not be visible at this magnification. At 250°C several large black precipitates (diameter $\sim 2\mu\text{m}$) are visible which may be overaged epsilon-carbides, and these redissolve after 350°C . After tempering at 500°C the original austenite grain boundaries become more visible indicating that cementite may be precipitating at these boundaries

(austenite grain diameter $\sim 22\mu\text{m}$). At 560°C the compositional change is much more apparent. Many areas appear lighter as the carbon concentration is reduced and more cementite precipitates. After tempering at 625°C the cementite particle density increases. Also white bands appear on the grain boundaries. These are probably the growth of ferrite grains. At 650°C a well established ferrite and cementite structure exists.

8.5.1.2 Hardness and Coercivity Measurements.

Fig.8.5 shows the mechanical hardness as a function of the tempering temperature. The 'as quenched' material has a hardness of 690 ± 15 which is typical for the martensitic structure in a 0.5%C steel. The hardness remains fairly constant after the 150°C temper which is characteristic of the combined effect of the hardening due to the formation of epsilon carbide and the softening due to the carbon depletion of the martensite matrix. At higher temperatures the material becomes markedly softened. In the early parts of the third stage of tempering both the solution of the epsilon carbides and the removal of carbon from the martensite result in softening, but also there is a hardening component due to the precipitation of cementite. As the temperature increases there is a further softening expected due to the growth and coalescence of cementite particles which occurs when the steel has attained a simple ferrite and cementite structure.

Fig.8.5 also shows the coercivity as a function of the tempering temperature. The 'as quenched' material has a coercivity of $1180 \pm 80 \text{ Am}^{-1}$ (this reading had the largest error primarily due to the low reading obtained from one of the specimens). This falls gently as the tempering temperature is increased until at 500°C it was $937 \pm 6 \text{ Am}^{-1}$. At higher temperatures the coercivity falls more rapidly, as the cementite / ferrite structure forms, until at 650°C it is only $422 \pm 3 \text{ Am}^{-1}$.

8.5.1.3 Barkhausen Emission Activity Profiles.

The B.E. activity profiles and the initial and central peak amplitudes as a function of the tempering temperature are shown in figs.8.6 and 8.7 respectively. The central peak amplitude increases rapidly going through a large maximum at 600°C as the martensitic structure transforms to cementite. An initial peak forms at 500°C which is the temperature where cementite is first observed to form (section 8.5.1.1). At higher temperatures this initial peak follows a similar trend to that of the central peak, and becomes the larger of the two peaks at 600°C and above. This peak occurs at the same field as the M.A.E. initial peak and so is probably associated with 90° domain wall movements. A smaller final peak would be expected to occur from 90° domain wall movements at a field corresponding to the M.A.E. final peak but this cannot be resolved from the central peak.

8.5.1.4 Magneto-acoustic Emission Activity Profiles.

The M.A.E. activity profiles as a function of the tempering temperature are shown in fig.8.8. The signal was too small to measure for tempering temperatures below 350°C. This implies that martensite structure does not allow M.A.E. to occur. This is because martensite only has one easy magnetic axis so that only 180° domain walls can exist. Therefore all the B.E. activity in the martensite is associated with 180° domain wall movements. The initial peak, central dip and final peak amplitudes as a function of the tempering temperature are shown in fig.8.9. At temperatures above 350° the M.A.E. activity increases monotonically at all fields. It is interesting to note that the initial / central and initial / final peak ratios, shown in fig.8.10, both show a peak at a similar temperature to that of the B.E. activity. Also the initial / final peak ratio is unusually large.

8.5.1.5 Summary.

(1) The B.E. activity.

(a) The activity increases at all fields as the martensite structure recovers and cementite particles precipitate.

(b) The martensite structure gives rise to a simple central peak in activity. This is due to 180° domain wall movements.

(c) The cementite structure is associated with a large initial peak which is often larger than the central peak. The initial peak chiefly arises from 90° domain wall movements.

(2) The M.A.E. activity.

(a) No M.A.E. activity occurs from the martensite as this structure only allows 180° domain walls to exist.

(b) As cementite particles precipitate the M.A.E. activity increases at all fields.

(c) The initial / final peak ratio is large in cementite (1.4-2.3).

8.5.2 Isothermally Tempered Specimens.

8.5.2.1 Optical Micrographs.

Fig.8.11 shows the microstructure of the material tempered at 650°C . After tempering for 30 minutes the structure is similar to the 625°C temper in 8.5.1.1. Light regions of ferrite can be observed at the original austenite grain boundaries, and a large density of cementite precipitates can be seen. After 100 minutes there is both more ferrite and cementite. In particular a decoration of black cementite particles can be seen at the austenite grain boundaries. The 300 minute temper gives rise to more ferrite, and the decoration of the martensite plate boundaries with cementite particles. After 1000 minutes the microstructure appears to remain the same.

8.5.2.1 Hardness and Coercivity Measurements.

Fig.8.12 shows the hardness as a function of the tempering time at

650°C. After 30 minutes it is 269 ± 4 and this falls almost linearly with the logarithm of time until after 1000 minutes it is 196 ± 3 . In the third stage of tempering where aggregates of cementite and ferrite are involved there is a simple relationship between the temperature and time (Reed-Hill, 1973);

where t is the time to attain a certain hardness.

$$1/t = A \cdot \exp(-Q/RT)$$

Q is an empirical activation energy.

A is a constant.

-(Eqⁿ.8.1)

Therefore the observed hardness relationship corresponds to an almost linear dependence of hardness on temperature, which is consistent with the high temperature region of fig.8.5.

Fig.8.12 also shows the coercivity as a function of the tempering time. This remains fairly constant over the time range 10-100 minutes at $\sim 426 \text{ Am}^{-1}$, but falls away for longer times until after 1000 minutes it is $365 \pm 10 \text{ Am}^{-1}$. This linear region was not observed in fig.8.5 but the coercivity of the specimens tempered for 10 minutes at 650°C is already much smaller than that for the specimens tempered at 625°C for 100 minutes ($612 \pm 12 \text{ Am}^{-1}$).

8.5.2.3 Barkhausen Emission Activity Profiles.

The B.E. activity profiles as a function of the tempering time are shown in fig.8.13. The high activity observed after 10 and 30 minutes is similar to that of the specimen tempered for 100 minutes at 600°C (fig.8.6). The activity then falls dramatically after 100 minutes at 650°C which is surprising but consistent with the sharp decline in activity observed as a function of tempering temperature (fig.8.7). The activity then increases again after 300 minutes and remains high after 1000 minutes. The exact form of the activity profile varies slightly each time it is measured and the initial and central peak amplitudes are generally not well defined. However these peak amplitudes have been estimated and are shown in

fig.8.14 (note the curves are not intended to be an accurate interpolation!). At each tempering time the peak amplitudes of both specimens agree to within ~10% (except the central peak amplitude for the specimens tempered for 10 minutes), which implies that the dip at 100 minutes is genuine.

8.5.2.4 Magneto-acoustic Emission Activity Profiles.

The M.A.E. activity profiles and the initial peak, central dip and final peak as a function of the tempering time are shown in figs.8.15 and 8.16 respectively. The activity remains fairly constant with tempering time, showing a small minimum at 100 minutes. However the initial / central peak ratio (fig.8.17(a)) and the initial / final peak ratio (fig.8.17(b)) both show that the specimens tempered for 100 minutes have an anomalously large ratio. The general trend is a decrease in both ratios as a function of the tempering time (the initial / final peak ratio falls to unity). These trends are consistent with those observed in fig.8.10 for temperatures above 600°C.

8.5.2.5 Summary.

(1) The B.E. activity.

(a) The activity from cementite is large.

(b) The profile is jagged with poorly defined initial and central peaks.

(c) As in 8.5.1.5 the structure is associated with a large initial peak which is larger than the central peak. The initial peak is mainly due to 90° domain wall movements.

(2) The M.A.E. activity.

(a) Although there are some changes in the microstructure the M.A.E. remains fairly constant at all fields.

(b) The initial / final peak ratio falls with tempering time (1.4-1.0).

8.5.3 Normalised Specimens.

8.5.3.1 Optical Micrographs.

Fig.8.18 shows the microstructure of the normalised material. Because the air cool was rather rapid, large regions of martensite were produced in addition to the simple ferrite / pearlite structure usually associated with normalisation (section 8.3.3.3). The martensite grain diameter (shown in table 8.3) increases with normalisation time in agreement with the expected increase in the austenite grain size. There are also dark regions of pearlite present and the ferrite is of the Widmanstätten form. The regions of ferrite become larger after 30 minutes although there are less of them. After 100 minutes there is less ferrite because the martensite grains have become very large. Consequently the most large regions of ferrite occur after the intermediate normalisation time (a point we will come back to later).

8.5.3.2 Hardness and Coercivity Measurements.

Fig.8.19 shows the hardness as a function of the heat treatment time at 900°C. This material is much softer than the purely martensite microstructure the observed hardness being 279 ± 2 after the 30 minute treatment. The hardness increases with the time which is consistent with an increasing martensite grain diameter (table 8.3).

The coercivity as a function of the heat treatment time is also shown in fig.8.19. This remains fairly constant at $\sim 440 \text{ Am}^{-1}$ after 10 and 30 minutes and then increases to $\sim 520 \text{ Am}^{-1}$ after 100 minutes. After 30 minutes the martensite grain size is larger and this would be expected to increase the coercivity. However the larger regions of ferrite probably counteract this. At 100 minutes there is even more martensite and less ferrite which increases the coercivity.

8.5.3.3 Barkhausen Emission Activity Profiles.

The B.E. activity profiles and the initial and central peak amplitudes

as a function of heat treatment time are shown in figs.8.20 and 8.21 respectively^e. The central peak amplitude (fig.8.21) increases with normalisation time and the full width of the profile becomes smaller. This dominant central peak is absent in the M.A.E. profiles and so must arise from 180° domain wall movements. The initial peak amplitude (fig.8.21) is a maximum after the 30 minutes heat treatment. It occurs at the same field as the M.A.E. initial peak and so must consist largely of irreversible 90° domain wall movements.

We can interpret the B.E. activity in the following way. As the external magnetic field is reduced from saturation, domain walls nucleate in the magnetically soft ferrite. These move quickly until they become pinned on the martensite and pearlite grains, giving rise to the initial peak in the activity. Therefore the initial peak amplitude is monotonically related to the size of the ferrite regions since this determines the maximum size of the Barkhausen events (8.5.3.1). Then as the field is reduced through zero the domain walls move through the martensite grains (irreversible 180° domain wall activity) giving rise to the central peak in the activity profile. Therefore the central peak amplitude is monotonically related to the martensite grain size (table 8.3).

The overall changes in the profile are small compared to the changes observed in the tempered material. This is probably because, unlike the effects of tempering, the type of pinning sites present does not change on heat treatment.

8.5.3.4 Magneto-acoustic Emission Activity Profiles.

The M.A.E. activity profiles and the initial peak, central dip and final peak amplitudes as a function of the heat treatment time are shown in figs.8.22 and 8.23 respectively^e. The activity is less than that observed from a cementite structure (fig.8.15). This is probably because the large fraction of martensite present restricts 90° domain walls to the regions of ferrite and pearlite. The activity goes through a small minimum

at 30 minutes which was unexpected since we might predict the same trend as that for the B.E. initial peak amplitude. However the difference is small and the M.A.E. readings are not as accurate in an absolute sense as the B.E. ones. Consequently in order to measure this small trend we need to take a ratio of parameters (this being more accurate). If the initial peak amplitude is divided by the central dip amplitude (fig.8.24(a)) the trend is the same as the B.E. initial peak and consequently the same as the B.E. initial peak / central peak ratio (the central peak being almost constant). The initial peak / final peak ratio, shown in fig.8.24(b), does not change with the heat treatment time. Like the cementite specimens it is quite large, viz. 1.45 ± 0.05 .

8.5.3.5 Summary.

(1) The B.E. activity.

(a) The activity does not change dramatically with heat treatment. The profile consists of a large central peak and a small initial peak.

(b) The central peak appears to be associated with irreversible 180° domain wall activity from the martensite (increasing with martensite grain size).

(c) The initial peak appears to be associated with domain wall movements in the ferrite.

(2) The M.A.E. activity.

(a) The activity is smaller than that associated with the cementite structures (as martensite is present), and again the profile does not change very much with heat treatment time.

(b) The initial peak / central dip ratio follows the same trend as that for the B.E. ratio.

(c) The initial peak / final peak ratio is quite large (1.45).

8.5.4 Fully-annealed Specimens.

8.5.4.1 Optical Micrographs.

Fig.8.25 shows the microstructure for the annealed material. This consists of grains of ferrite and pearlite as expected. There is much more ferrite than in the normalised material, probably because no martensite was able to form during the slow cooling. Both the ferrite and pearlite grain diameter increase with annealing time (table 8.3), which is consistent with an originally larger austenite grain size.

8.5.4.2 Hardness and Coercivity Measurements.

Fig.8.26 shows the hardness as a function of the annealing time. As expected the fully annealed material is very soft. The hardness remains constant at ~180 which may be due to there being the same pearlite fraction in each specimen. i.e. from the Lever equation the % pearlite is given as;

$$\% \text{pearlite} = \% \text{carbon} \times \frac{100}{0.87} \quad (\text{Reed-Hill, 1973}) \quad (\text{Eq}^n.8.2)$$

and so is independent of the heat treatment time. However it is surprising that the hardness does not change with the grain size.

Fig.8.26 also shows the corresponding coercivity measurements. These are observed to decrease as a function of annealing time. Theoretical models of the coercivity in pearlite/ferrite steels predict that the coercivity is inversely proportional to the pearlite grain size (Hetherington, 1985). This agrees fairly well with these measurements.

8.5.4.3 Barkhausen Emission Activity Profiles.

The B.E. activity profiles as a function of the annealing time are shown in fig.8.27. Again the form of the profiles does not change very much, which is probably because the nature of the pinning sites remains the same. This will be useful in the practical application of stress measurement. The amplitude of the central peak is similar to that of the normalised material but the initial peak is twice as large. This could be

because there is more ferrite present in the annealed material, the initial peak being again associated with the ferrite (measurements made on pure annealed ferrite resulted in a B.E. activity profile with an initial peak but no central peak (final profiles of fig.4.6)). The form of the profile is therefore understood as follows. On reducing the external magnetic field from saturation domain walls nucleate and move quickly through the ferrite before becoming pinned on the pearlite grain boundaries (observed by Hetherington et al., 1986 on the H.V.E.M.). Therefore the Barkhausen events from the initial peak region (both 90° and 180° domain wall activity) will be generally larger for larger ferrite grains. Consequently the B.E. initial peak increases for increasing ferrite grain size (table 8.3 and fig.8.28). On reducing the magnetic field through zero the domain walls will move through the pearlite resulting in the central peak. The amplitude of this peak remains fairly constant with annealing time (fig.8.28).

8.5.4.4 Magneto-acoustic Emission Activity Profiles.

The M.A.E. activity was greater at all fields than was observed for the normalised material (fig.8.29). The larger regions of ferrite in the annealed material again account for the greater activity observed in the outer peak regions. The high activity at low fields (almost producing a small peak in the profile) probably arises from 90° wall activity in the pearlite grains. The domain structure in pearlite is complex and consists of many small domains (Hetherington et al., 1986) and therefore the domain jumps will be small resulting in less M.A.E. activity than in ferrite (section 5.6). The initial peak, central dip and final peak amplitudes are shown in fig.8.30. These show little change as a function of the annealing time. However the initial peak / central dip ratio (which can be measured more accurately and is therefore more sensitive), shown in fig.8.31(a), increases as the ferrite grain size increases, in a similar manner as was observed in the normalised material. The initial peak / final peak ratio is shown in fig.8.31(b). This decreases with increasing annealing time.

8.5.4.5 Summary.

(1) The B.E. activity.

(a) The activity does not change dramatically with annealing time. The profile consists of a large central peak and an initial peak ~40% smaller.

(b) The initial peak appears to be associated with irreversible domain wall movements in the ferrite.

(c) The central peak can be associated with with wall movements in the pearlite.

(2) The M.A.E. activity.

(a) The activity is ~50% larger than that observed in the normalised material, and again the profile does not change very much with annealing time.

(b) The activity at low fields is larger than in any of the other structures and is associated with the pearlite.

(c) The initial peak / central dip ratio follows the same trend as the B.E. initial peak.

(d) The initial peak / final peak ratio is close to unity (1.13-1.09).

From the data presented above it is seen that the different microstructures give rise to different characteristics in the B.E. and M.A.E. profiles. Thus in order to make quantitative stress measurements it will be necessary to know which structures are present. When the dependence of B.E. and M.A.E. upon stress is analysed in the next section it will become apparent that for many of the basic types of microstructure an M.A.E. parameter can be chosen that is monotonically dependent on stress but almost independent of certain changes in the microstructure. Therefore a knowledge of the basic microstructure present will enable the appropriate M.A.E. parameter to be chosen to enable the stress to be measured. The B.E. activity is more sensitive to the microstructure, and the general form of the profile was characteristic of the basic type of microstructure present.

Consequently, if the microstructure is unknown then a measurement of B.E. will enable this to be determined qualitatively, and will then guide the choice of the M.A.E. parameter for stress measurement.

8.6 Results and Discussion (II)-Stress Effects.

8.6.1 Isochronally Tempered Specimens.

8.6.1.1 Barkhausen Emission Activity Profiles.

The B.E. activity profiles as a function of stress were found to be quite different for each tempering temperature. The profiles for a number of applied tensile loads are shown in fig.8.32 for a specimen tempered at 600°C. (The exact form of the profile at zero applied load is slightly different from that shown in fig.8.6. This is because both the magnetising coil and the detection coil systems are different from those used for the microstructure measurements.) In this specimen the B.E. activity generally decreases as a function of increasing stress at all fields. However the initial peak becomes progressively more prominent in the load range 0-35 kg. The initial peak and low field amplitudes as a function of the applied stress are plotted in fig.8.33. As the stress is increased from 0-10 MPa the activity decreases very rapidly. For higher stresses the central amplitude remains fairly constant. However the initial peak amplitude begins to increase reaching a maximum at ~60 MPa before decreasing rapidly for larger stresses. This stress dependence is different from that obtained in 'as received' mild steel (fig.7.13) and indicates that the B.E. is very sensitive to the microstructure.

The B.E. activity low field amplitude as a function of the applied tensile stress for a specimen tempered at each of the temperatures in table 8.2 (excluding 600°C (fig.8.33) and 650°C (fig.8.42(c))) are shown in figs.8.34 and 8.35. The low field B.E. activity increases monotonically with increasing stress for the 'homogenised only' specimen (fig.8.34(a)). After tempering at 250°C (fig.8.34(b)) and 350°C (fig.8.34(c)) the activity increases rapidly for stresses <20 MPa and then remains fairly constant at

higher stress values. However at 500°C (fig.8.35(a)) the B.E. activity decreases with increasing stress, and at 560°C (fig.8.35(b)) and 625°C (fig.8.35(c)) this inverse dependence becomes stronger. At ~500°C the martensitic structure is recovering and cementite particles are beginning to precipitate (section 8.5.1.1). Therefore it appears that for martensitic structures the B.E. generally increases as a function of stress whereas for structures consisting of cementite particles in a ferrite matrix it decreases. These different dependences upon the applied tensile stress may be understood as follows: The martensite only allows 180° domain walls to exist (this being experimentally observed and theoretically justified (8.5.1.4)). Therefore, according to the simple argument of section 7.3, we would expect the B.E. to increase with stress, as is observed. However in the cementite structures irreversible 90° domain wall activity was observed (8.5.1.4). This 90° activity increased with tempering temperature (indicated by the M.A.E. activity figs.8.8 and 8.9) and so we may expect the B.E. activity to gradually change from an increasing to a decreasing function of the stress. i.e. provided the activity from 90° domain walls becomes greater than that from 180° domain walls, the stress dependence will change from being controlled by 180° wall motion to 90° wall motion.

The main trends as a function of both microstructure and stress can be summarised for the central peak amplitude by plotting the data in the form of contour levels. In order to do this the data was interpolated in order to evaluate its value at equally spaced values of both stress and tempering temperature. This was done on a VAX computer by using a cubic spline fit technique. Then a suitable contour routine was used in order to calculate the contour positions. Fig.8.36 shows a contour map obtained in this way. (The measurements of the two specimens tempered at each temperature were averaged both here and in all following contour graphs). The following is observed:

- (1) The sharp maximum in activity at 600°C and zero applied stress.

- (2) The activity increases gradually with stress for tempering temperatures below 500°C . This is the martensitic region where only irreversible 180° domain wall activity is observed.
- (3) The activity decreases rapidly with stress for tempering temperatures above 500°C , the strongest dependence being at 600°C . This is the cementite region where much irreversible 90° domain wall activity is observed.

8.6.1.2 Magneto-acoustic Emission Activity Profiles.

The martensitic specimens could not be investigated since there was no observed M.A.E. signal from these. Several M.A.E. profiles measured at different applied loads are shown in fig.8.37 for the specimen tempered at 600°C . The M.A.E. decreases monotonically at all fields with increasing load. The shape of the profile also changes so that over the range 0-78 kg the initial peak / final peak ratio increases from 1.1 to 1.4. However the initial peak / central dip ratio remains almost unchanged at 3.1 to 2.9. The M.A.E. activity decreased with increasing stress for all of the specimens investigated. The initial peak, central dip and final peak as a function of stress for specimens tempered at 500° , 560° , 600° , and 625°C are shown in fig.8.38. The dependence upon stress is much simpler than B.E.. The reason for this could be that, whereas the B.E. activity consists of activity from both 90° and 180° domain walls, M.A.E. activity only occurs from the 90° domain walls. The universal decrease as a function of stress lends further support to the simple argument of section 7.3 for 90° wall activity in positive magnetostriction materials.

The main trends are summarised in the form of contour maps as before. The M.A.E. initial peak amplitude as a function of both stress and tempering temperature is shown in fig.8.39. This parameter is a strong function of both stress and microstructure as indicated by closely packed contours at $\sim 45^{\circ}$ angles to the axes. At 650°C the contours become almost vertical implying that in this high temperature region the initial peak

becomes insensitive to microstructure changes . The central dip and final peak amplitudes (not shown) show similar trends. The initial peak / central dip ratio, shown in fig.8.40, is almost independent of the applied stress for tempering temperatures below 600°C, whereas it is strongly dependent upon the microstructure. Above 600°C the dependence upon stress and microstructure changes (a point that will be discussed in section 8.6.2.2). In summary, for the isochronally tempered material:

- (1) There is no signal from the martensitic phase.
- (2) The signal increases with tempering temperature, as the proportion of martensite decreases.
- (3) The signal decreases with stress.
- (4) The sensitivity to stress and microstructure is comparable.

8.6.2 Isothermally Tempered Specimens.

8.6.1.1 Barkhausen Emission Activity Profiles.

The B.E. activity from the specimens tempered at 650°C is again a complicated function of both the stress and the microstructure. The activity profiles for several loads from a specimen tempered for 10 minutes is shown in fig.8.41. As the load increases the activity generally falls and the profile shape changes. The initial and central peak amplitudes as a function of the stress are shown in fig.8.42(a). The initial peak decreases rapidly as the stress increases; this sharp decrease is expected since the initial peak is mainly due to 90° domain wall movements (see sections 8.5.2.5 and 7.3). The central peak amplitude is almost independent of the stress and this is unexpected since this parameter decreased with increasing stress in material tempered at 625°C for 100 minutes. But a temper at 650°C for 10 minutes appears to be more severe than the 100 minute temper at 625°C as indicated by the coercivity measurements (8.5.2.1:-fig.8.12). Therefore the domain structure may be quite different.

The initial and central peak amplitudes as a function of the applied tensile stress for specimens tempered at 650°C for the times listed in

table 8.2 are shown in figs.8.42 and 8.43. These are only representative figures since different specimens showed slightly different trends as a function of the stress. From these it is apparent that the central peak amplitude does show small changes with stress (fig.8.42(b) (30 mins. temper) shows a decreasing trend, fig.8.42(c) (100 mins. temper) a decreasing followed by an increasing trend). These results suggest that another process is occurring besides a simple redistribution of domain wall types from 90° to 180° (e.g. the domain size could be changing). The initial peak amplitude decreases with stress for all the specimens consistent with 90° wall activity (note it is also consistent with a decreasing domain size).

The contour plot of the B.E. initial peak amplitude as a function of both stress and tempering time is shown in fig.8.44. The results are summarised as follows:

- (1) The amplitude increases with tempering time although the 100 minute temper is anomalously low (8.5.2.3).
- (2) The amplitudes at 300 and 1000 minutes are almost identical: consistent with the similar microstructures observed (8.5.2.1).
- (3) The amplitude decreases with increasing stress.
- (4) The stress dependence falls off above 80 MPa.

Fig.8.45 shows the corresponding plot for the central peak amplitude. The stress dependence is small as indicated by almost horizontal contours. In general we can conclude that the dependence of B.E. upon stress in tempered material is complex and very sensitive to the microstructure. Therefore B.E. is not very useful for stress measurement in these structures.

8.6.1.2 Magneto-acoustic Emission Activity Profiles.

The M.A.E. activity again showed a simpler dependence upon stress than B.E. Fig.8.46 shows several profiles at different loads obtained from the specimen tempered for 10 minutes. As for the isochronally tempered material

the activity decreases at all fields as the load increases. The initial peak, central dip and final peak amplitudes as a function of the stress for the specimens tempered at 650°C for the times listed in table 8.2 are shown in figs.8.47 and 8.48. Again the activity decreases at all fields as the stress increases. The central dip amplitude shows a modest decrease with stress (35% at 190 MPa in the specimen tempered for 10 minutes (fig.8.47(a))). However, in these cementite materials the M.A.E. outer peak amplitudes decrease more rapidly with stress than in any of the other structures investigated (52% in the same specimen at 190 MPa). Also, unlike B.E., the stress dependence is still strong at 190 MPa. The M.A.E. initial peak, central dip and final peak amplitudes as a function of stress and tempering temperature (contour plots not shown) all consist of closely packed vertical lines. This indicates that the stress dependence is dominant. Consequently, in a practical testing situation, if it is known that the structure is cementite then M.A.E. could effectively be used to measure the stress level. The M.A.E. initial peak / central dip ratio contour plot is shown in fig.8.49. In the isochronally tempered material (fig.8.40) this varied quickly with tempering temperature but was not strongly dependent upon the stress for temperatures below 600°C. However the reverse is now the case. This parameter is strongly dependent upon the stress level as compared to the tempering time (the contours being almost vertical). This indicates that this parameter could also be used to measure the stress value. Also the use of a ratio would avoid the need for an absolute calibration of the acoustic measurement system, especially the coupling of the specimen to the transducer. In summary, for the isothermally tempered material:

- (1) The sensitivity to tempering time is weak as compared to stress.
- (2) The signal decreases with stress at all fields.
- (3) The sensitivity to stress extends over the whole 180 MPa range.
- (4) The initial peak / central dip parameter appears to be particularly useful for measuring the stress level.

8.6.3 Normalised Specimens.

8.6.3.1 Barkhausen Emission Activity Profiles.

The stress dependence of the B.E. activity from the normalised specimens is rather complex. Fig.8.50 shows several profiles measured at different applied loads from a specimen treated for 30 minutes at 900°C. The dependence upon stress is not as large as that observed from some of the tempered material. The central peak height only changes slightly but the width of the peak appears to increase for larger loads. This indicates that the total Barkhausen activity increases slightly with load, but at fields other than the low field peak position. Fig.8.51 shows the central peak amplitudes as a function of stress for the specimens treated for the times listed in table 8.2. The specimens treated for 10 and 100 minutes show a sharp increase with stress as the stress increases from zero to 20 MPa. It is not clear whether this is a true effect since at very low stress in these grips the specimen and support are not perfectly rigid, so that there may be some fluctuations in the detected signal. At larger stresses the central peak amplitude decreases until it reaches a minimum. This minimum value occurs at ~100, ~80 and ~50 MPa for heat treatment times of 10, 30 and 100 minutes respectively. The amplitude then continues to increase for stresses up to 200 MPa. It is not clear why this should be the case but we would expect some decrease in the B.E. activity from the 90° wall movements in the ferrite and an increase from the 180° wall movements in the ferrite and martensite. From the results it appears that the domain structure in the ferrite is influenced at low stresses and in the martensite at high stresses. Hence an increase in the martensite grain size (table 8.4) will give rise to a greater general increase in the B.E. with stress.

The complicated microstructure in these specimens (section 8.5.3.1) is reflected in the complex stress dependence. The contour plot of this central peak amplitude as a function of both stress and heat treatment time, fig.8.52, summarises the results for the behaviour of the B.E. for

the normalised specimens:

- (1) The dependence upon both microstructure and stress is small (note contour scale).
- (2) The microstructure trend dominates; particularly at larger stresses.
- (3) The signal increases with heat treatment (for stresses above 10 MPa).
- (4) The stress dependence is complex. There is a minimum signal for a stress level in the range 50-100 MPa (the value being dependent upon the heat treatment); the activity increasing for larger stresses.

8.6.3.2 Magneto-acoustic Emission Activity Profiles.

In contrast to the complexity of the B.E. activity the M.A.E. activity is more simply dependent upon the applied load. Fig.8.53 shows several profiles for the specimen treated for 30 minutes at different loads. The M.A.E. outer peak amplitudes decrease with increasing load, as they did for the tempered material, in accordance with the expected behaviour of materials with positive magnetostriction. The initial peak, central dip and final peak amplitudes as a function of the applied tensile stress for the specimens tempered for 10, 30 and 100 minutes are shown in fig.8.54. All of these parameters decrease with stress, although the decrease is not as severe as that observed for the cementite structures. The initial peak / central dip ratio also decreases (indeed it appears to be smaller than 1.0 at 190 Mpa in the specimen treated for 10 minutes, but the small size of the signal at this stress makes it difficult to measure this ratio accurately). It is surprising that, despite the complexity of the microstructure, the stress dependence is fairly simple. This may be because the nature of the pinning sites does not change with heat treatment.

A contour plot of the M.A.E. initial peak amplitude, fig.8.55, and the initial peak / central dip amplitude, fig.8.56, as a function of the stress and the heat treatment time summarises some of these results for normalised material:

- (1) The microstructure dependence is weak.

- (2) The initial peak increases slightly with heat treatment time.
- (3) The signal decreases with increasing stress.
- (4) The initial peak / central dip ratio increases with heat treatment time.
- (5) This ratio generally decreases with stress (particularly for stresses larger than 80 MPa).

8.6.4 Fully Annealed Specimens.

8.6.4.1 Barkhausen Emission Activity Profiles.

The B.E. activity profiles obtained as a function of stress were slightly different from those measured at zero stress in the table-mounted solenoid. Fig.8.57 shows the activity profiles at various loads from a specimen annealed for 30 minutes. Additional peaks can be seen at the higher magnetic fields. The reason for the appearance of these peaks is not clear. Tentatively it is thought that they could arise from domain wall movements near the ends of the specimen where there are compressive forces due to the clamping. This may cause significant effects in these very plastically soft specimens, particularly at low loads where the clamping stress is much greater than the tensile stress.

The shape of the B.E. activity profile changes with stress, and the initial peak and central peak amplitudes for the specimens are shown in fig.8.58 (the additional peaks were not measured). These graphs indicate that the stress dependence is complicated. The central peak amplitude remains fairly constant with increasing stress implying that domain wall motion in the pearlite grains is little affected (section 8.5.4.3). However the initial peak shows an initial decrease up to 50, 60 and 80 MPa for annealing times of 30, 100 and 1000 minutes respectively followed by an increase for higher stresses. This is similar to the results from the normalised specimens and suggests that 90 and 180° domain walls in the ferrite are influenced in a complicated way by the applied stress. Therefore we cannot draw any conclusions about the micromagnetic mechanisms

in these specimens. However it is apparent that the B.E. signal from this material does not lend itself very readily to stress measurement.

8.6.4.2 Magneto-acoustic Emission Activity Profiles.

The M.A.E. activity, arising from only 90° domain wall movements, again shows a simpler dependence upon stress. Fig.8.59 shows several profiles measured from a specimen annealed for 30 minutes. The activity at high fields decreases with increasing stress. The initial peak, central dip and final peak amplitudes as a function of the stress for the specimens are shown in fig.8.60. There are similar reductions in the signal with increasing stress for all three annealing times. Also the initial peak / central dip amplitude falls from 1.40 to only 1.06 at 190 MPa. These results are summarised in the contour plots of the initial peak amplitude and the initial peak / central dip ratio for the fully annealed material shown in figs.8.61 and 8.62:

- (1) The sensitivity to stress is much greater than to microstructure.
- (2) The M.A.E. activity decreases at all fields with increasing stress.
- (3) The ratio generally decreases with stress (particularly above 60 MPa).

8.7 Implications for Stress Measurement.

It is clear from the data presented here and in the previous chapter that for the measurement of internal stress that some knowledge of the local microstructure is required. In addition the stress dependence of B.E. is quite complex in most materials and consequently this technique alone can only be easily used after very careful calibration using similar materials (King and Smith, 1978). However if both B.E. and M.A.E. can be measured then it should be possible to determine the stress level for a steel specimen with an unknown microstructure. A suggested procedure for doing this would be as follows:

- (1) Measure the B.E. and M.A.E. profiles from the specimen. Five (or six) independent parameters can be extracted, viz: The B.E. initial and

central (and sometimes final) peak amplitudes, and the M.A.E. initial peak, low field and final peak amplitudes. There are also several related parameters which can be useful, such as the M.A.E. initial peak / low field ratio etc.

(2) From the form of the B.E. profile (and to a lesser extent the M.A.E. profile), determine qualitatively which domain wall pinning sites are present (i.e. the microstructure). For example martensite gives rise to a single peak in B.E. but no M.A.E. at all. Ferrite / pearlite gives rise to an initial and central peak in the B.E. of a characteristic shape and an M.A.E. initial peak / low field ratio close to unity. Cementite gives rise to a high B.E. signal with an initial peak dominating the profile and a high M.A.E. signal (see section 8.5).

(3) With a knowledge of which microstructures are present in the specimen, the appropriate calibration can be used for the M.A.E. signal parameter. For the microstructures for which you can use M.A.E. to measure the stress, the ratio of the initial peak to the low field amplitude is usually the preferred parameter. Being a ratio, no calibration of the acoustic coupling and sensitivity is required.

(4) If a reliable acoustic coupling can be guaranteed then the height of the initial M.A.E. peak may be a preferable parameter, especially for lower stresses. For example in ferrite / pearlite and ferrite / pearlite / martensite microstructures this parameter gave the better stress dependence for stresses below 40 and 80 MPa respectively.

(4) Some structures do not appear to be amenable to stress measurement by M.A.E. In particular pure martensite gives no M.A.E. at all, and partially recovered martensite gives M.A.E. that depends sensitively on the microstructure but hardly at all on the stress. It may be that B.E. could be used to measure surface stress in these cases, with M.A.E. aiding the identification of the microstructure. For example in the partially recovered martensite the M.A.E. initial peak / low field ratio will indicate fairly precisely the microstructure, and then the B.E.

central peak could be used to measure the surface stress.

An important point to consider is that in real structures the stress or the microstructure, or both, may vary with depth in the material. It will be necessary to consider how to extend these techniques for application in such cases.

8.8 Summary and Conclusions.

Both the B.E. and M.A.E. signals are sensitive to the microstructural and stress state in mild steel. The activity occurring from different microstructural features in a given specimen is often observed at different fields in the profile. In general as the field is reduced from saturation, domain wall creation and movement will begin in the soft magnetic regions such as ferrite giving rise to an initial peak in both the B.E. and M.A.E. profiles. At lower fields the activity will occur from regions of pearlite and/or martensite. This gives a large central peak in the B.E. activity profile but almost no M.A.E., as the domain walls are mostly of the 180° type. Consequently the different microstructures will result in different B.E. and M.A.E. profiles. For example large regions of ferrite and cementite give greater high field B.E. and M.A.E. activity, whilst large regions of martensite severely reduce the M.A.E. activity. The greatest low-field M.A.E. activity in the microstructures investigated was found from the pearlitic morphologies.

The effect of tensile stress is always to reduce the M.A.E. signal at all fields in mild steel. This is consistent with the expected behaviour for 90° domain walls. The sensitivity to stress is generally much greater than to grain size. The B.E. stress dependence is more complicated, being quite sensitive to the microstructural state. For example in the martensitic structures B.E. increases with increasing stress whereas in cementite structures it decreases. In ferrite/pearlite and ferrite/martensite the low field dependence is not monotonic.

In order to use these techniques to measure the stress level in steel,

the microstructure must be determined. If this is not already known, then a combination of measurements of B.E. and M.A.E. can be used to determine the type of pinning sites that are present. Most of the useful information for this comes from the B.E., which has much stronger dependence on the microstructure than the M.A.E. has, though it should be remembered that the B.E. is confined to a layer close to the surface. Once the kind of microstructure is known, then in most cases the stress can be determined from the M.A.E. alone. The initial peak / low field ratio is generally the most useful parameter. Considerable variations in the microstructure are liable to occur in precisely those regions, such as welds, where knowledge of internal stress is of greatest importance. Provided that the fundamental form of the microstructure does not change (i.e. the same phases are present), then this ratio is rather insensitive to changes in the grain size and in the proportions of the different phases, and this, together with the low attenuation of M.A.E signals through steel, makes this a potentially reliable parameter for the non-destructive measurement of internal stress.

Table 8.1
The composition of 4360 steel.

	Fe	C	Si	Mn	S	P
% wt.	Balance	0.45 - 0.55	0.05 - 0.35	0.7 - 1.0	0.06max	0.06max

Table 8.2
The heat treatment of the 4360 steel rod and disc specimens.

Tempering Temperature (°C)	Tempering Time (mins)	Normalisation Time at 900°C (mins)	Full Annealing Time at 850°C (mins)	No. of rods	No. of discs
Quenched only	-	-	-	2	2
150	100	-	-	0	2
350	100	-	-	2	2
500	100	-	-	2	2
560	100	-	-	1	2
600	100	-	-	2	0
625	100	-	-	2	2
650	10	-	-	2	0
650	30	-	-	2	2
650	100	-	-	2	2
650	300	-	-	2	2
650	1000	-	-	2	2
-	-	10	-	2	2
-	-	30	-	2	2
-	-	100	-	2	2
-	-	-	30	1	1
-	-	-	100	2	2
-	-	-	1000	1	1

Table 8.3

(A) The martensite grain diameter as a function of the normalisation time and (B) the ferrite and pearlite grain diameter as a function of the annealing time.

(A)

Normalisation Time (mins.)	Martensite Grain Diameter (μm)
10(a)	14.8
10(b)	17.6
30(a)	24.9
30(b)	33.7
100(a)	44.4
100(b)	51.2

(B)

Annealing Time (mins.)	Ferrite Grain Diameter (μm)	Pearlite Grain Diameter (μm)
30	11.5	15.1
100(a)	19.4	16.6
100(b)	22.6	17.4
1000	17.3	25.1

ISOTHERMAL TRANSFORMATION

Chemical Composition	
C	Mn
0.50	0.91

McQuaid Ehn grain size 7—8 (ASTM).
Austenitizing Temperature 910°C.

Figure 8.1 The isothermal transformation diagram for a 0.5%C 0.91%Mn steel (Woolman and Mottram, 1969).

Figure 8.2 A portion of the Fe-C phase diagram with temperature ranges for process annealing, recrystallization annealing, stress relieving and spheroidization indicated. (Krauss, 1980).

20 μm

(c) 250°C temper

20 μm

20 μm

(d) 350°C temper

20 μm

Figure 8.3 Optical micrographs of isochronally tempered material. (a) Untempered, (b) 150°C temper, (c) 250°C, and (d) 350°C temper.

20 μm

(c) 625°C temper

20 μm

(d) 650°C temper

20 μm

20 μm

Figure 8.4 Optical micrographs of isochronally tempered material. (a) 500°C temper, (b) 560°C, (c) 625°C and (d) 650°C temper.

Figure 8.5 The room temperature hardness and magnetic coercivity measurements as a function of the tempering temperature for the isochronally tempered specimens.

Figure 8.6 The B.E. activity profiles for the isochronally tempered specimens.

Figure 8.7 The B.E. initial and central peak amplitudes as a function of the tempering temperature for the isochronally tempered specimens.

Figure 8.8 The M.A.E. activity profiles for the isochronally tempered material.

Figure 8.9 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the tempering temperature for the isochronally tempered specimens.

(A)

(B)

Figure 8.10 (A) The M.A.E. initial peak / central dip ratio and (B) the M.A.E. initial peak / final peak ratio as a function of the tempering temperature for the isochronally tempered specimens.

(a) 30 minutes temper

20 μm

(b) 100 minutes temper

20 μm

(c) 300 minutes temper

20 μm

(d) 1,000 minutes temper

20 μm

Figure 8.11 Optical micrographs of the isothermally tempered material. (A) 30 minutes, (B) 100 minutes, (C) 300 minutes and (D) 1,000 minutes temper.

Tempering time / mins.

Figure 8.12 The room temperature hardness and magnetic coercivity measurements as a function of the tempering time for the isothermally tempered specimens.

Figure 8.13 The B.E. activity profiles for the isothermally tempered specimens.

Figure 8.14 The B.E. initial and central peak amplitudes as a function of the tempering time for the isothermally tempered specimens.

Figure 8.15 The M.A.E. activity profiles for the isothermally tempered specimens.

Figure 8.16 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the tempering time for the isothermally tempered specimens.

Figure 8.17 (A) The M.A.E. initial peak / central dip ratio and (B) the M.A.E. initial peak / final peak ratio as a function of the tempering time for the isothermally tempered specimens.

(A) 10 minutes

20 μm

(B) 30 minutes

20 μm

(C) 100 minutes

20 μm

Figure 8.18 Optical micrographs for the material normalised at 900°C for (A) 10 minutes, (B) 30 minutes and (C) 100 minutes.

Normalisation time / mins.

Figure 8.19 The room temperature hardness and magnetic coercivity measurements as a function of the heat treatment time for the normalised specimens.

Figure 8.20 The B.E. activity profiles for the normalised specimens.

Figure 8.21 The B.E. initial and central peak amplitudes as a function of the heat treatment time for the normalised specimens.

Figure 8.22 The M.A.E. activity profiles for the normalised specimens.

Figure 8.23 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the heat treatment time for the normalised specimens.

Figure 8.24 (A) The M.A.E. initial peak / central dip ratio and (B) the M.A.E. initial peak / final peak ratio as a function of the heat treatment time for the normalised specimens.

(A) 30 minutes

100 μm

(B) 100 minutes

100 μm

(C) 1,000 minutes

100 μm

Figure 8.25 Optical micrographs for the material annealed at 850°C for (A) 30 minutes, (B) 100 minutes and (C) 1,000 minutes.

Annealing time / mins.

Figure 8.26 The room temperature hardness and magnetic coercivity measurements as a function of the annealing time for the fully annealed specimens.

Figure 8.27 The B.E. activity profiles for the annealed specimens.

Figure 8.28 The B.E. initial and central peak amplitudes as a function of the annealing time for the fully annealed specimens.

Figure 8.29 The M.A.E. activity profiles for the annealed specimens.

Figure 8.30 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the annealing time for the fully annealed specimens.

Figure 8.31 (A) The M.A.E. initial peak / central dip ratio and (B) the M.A.E. initial peak / final peak ratio as a function of the annealing time for the fully annealed specimens.

Figure 8.32 The B.E. activity profiles at different applied loads for a specimen tempered at 600°C for 100 minutes.

Figure 8.33 The B.E. initial and central peak amplitudes as a function of the applied tensile stress for a specimen tempered at 600°C for 100 minutes.

Figure 8.34 The B.E. central peak amplitude as a function of the applied tensile stress for (A) an untempered specimen, (B) a specimen tempered at 250°C and (C) a specimen tempered at 350°C for 100 minutes.

500 °C 100 mins.

560 °C 100 mins.

625 °C 100 mins.

Figure 8.35 The B.E. central peak amplitude as a function of the applied tensile stress for specimens tempered at (A) 500°C, (B) 560°C and (C) 625°C for 100 minutes.

Figure 8.36 A contour representation of the B.E. central peak amplitude as a function of both the applied tensile stress and the tempering temperature for the isochronally tempered specimens.

Figure 8.37 The M.A.E. activity profiles at different applied loads for a specimen tempered at 600°C for 100 minutes. Note that the profiles are displaced one above another for clarity.

500 °C 100 mins.

500 °C 100 mins.

600 °C 100 mins.

625 °C 100 mins.

Figure 8.39 A contour representation of the M.A.E. initial peak amplitude as a function of both the applied tensile stress and the tempering temperature for the isochronally tempered specimens.

Figure 8.40 A contour representation of the M.A.E. initial peak / central dip ratio as a function of both the applied tensile stress and the tempering temperature for the isochronally tempered specimens.

Figure 8.41 The B.E. activity profiles at different applied loads for a specimen tempered at 650°C for 10 minutes. Note that the profiles are displaced one above another for clarity.

650 °C 10 mins.

(A)

650 °C 30 mins.

(B)

(C)

Figure 8.42 The B.E. initial and central peak amplitudes as a function of the applied tensile stress for specimens tempered at 650°C for (A) 10 minutes, (B) 30 minutes and (C) 100 minutes.

Figure 8.43 The B.E. initial and central peak amplitudes as a function of the applied tensile stress for specimens tempered at 650°C for (A) 300 minutes and (B) 1,000 minutes.

Figure 8.44 A contour representation of the B.E. initial peak amplitude as a function of both the applied tensile stress and the tempering time for the isothermally tempered specimens.

Figure 8.45 A contour representation of the B.E. central peak amplitude as a function of both the applied tensile stress and the tempering time for the isothermally tempered specimens.

Figure 8.46 The M.A.E. activity profiles at different applied loads for a specimen tempered at 650°C for 10 minutes.

650 °C 10 mins.

(A)

650 °C 30 mins.

(B)

(C)

Figure 8.47 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the applied tensile stress for specimens tempered at 650°C for (A) 10 minutes, (B) 30 minutes and (C) 100 minutes.

650 °C 300 mins.

(A)

650 °C 1000 mins.

(B)

Figure 8.48 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the applied tensile stress for specimens tempered at 650°C for (A) 300 minutes and (B) 1,000 minutes.

Figure 8.49 A contour representation of the M.A.E. initial peak / central dip ratio as a function of both the applied tensile stress and the tempering time for the isothermally tempered specimens.

Figure 8.50 The B.E. activity profiles at different applied loads for a specimen heat treated at 900°C for 30 minutes.

900 °C 30 mins. AC (A)

900 °C 10 mins. AC (B)

900 °C 100 mins. AC (C)

Figure 8.51 The B.E. central peak amplitude as a function of the applied tensile stress for specimens heat treated at 900°C for (A) 10 minutes, (B) 30 minutes and (C) 100 minutes.

Figure 8.52 A contour representation of the B.E. central peak amplitude as a function of both the applied tensile stress and the heat treatment time for the normalised specimens.

Figure 8.53 The M.A.E. activity profiles at different applied loads for a specimen heat treated at 900°C for 30 minutes.

(A) 900 °C 10 mins.

(B) 900 °C 30 mins.

(C) 900 °C 100 mins.

Figure 8.54 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the applied tensile stress for specimens heat treated at 900°C for (A) 10 minutes, (B) 30 minutes and (C) 100 minutes.

Figure 8.55 A contour representation of the M.A.E. initial peak amplitude as a function of both the applied tensile stress and the heat treatment time for the normalised specimens.

Figure 8.56 A contour representation of the M.A.E. initial peak / central dip ratio as a function of both the applied tensile stress and the heat treatment time for the normalised specimens.

Figure 8.57 The B.E. activity profiles at different applied loads for a specimen annealed at 850°C for 30 minutes.

(A) 30 minutes

(B) 100 minutes

(C) 1,000 minutes

Figure 8.58 The B.E. initial and central peak amplitudes as a function of the applied tensile stress for specimens annealed at 850°C for (A) 30 minutes, 100 minutes and (C) 1,000 minutes.

Figure 8.59 The M.A.E. activity profiles at different applied loads for a specimen annealed at 850°C for 30 minutes.

(A) 30 minutes

(C) 1000 minutes

(B) 100 minutes

Figure 8.60 The M.A.E. initial peak, central dip and final peak amplitudes as a function of the applied tensile stress for specimens annealed at 850°C for (A) 30 minutes, (B) 100 minutes and (C) 1,000 minutes.

Contour No.	Amplitude/mV
1	10.00
2	12.67
3	15.33
4	18.00
5	20.67
6	23.33
7	26.00
8	28.67
9	31.33
10	34.00
11	36.67
12	39.33
13	42.00
14	44.67
15	47.33
16	50.00

Figure 8.61 A contour representation of the M.A.E. initial peak amplitude as a function of both the applied tensile stress and the annealing time for the fully annealed specimens.

Figure 8.62 A contour representation of the M.A.E. initial peak / central dip ratio as a function of both the applied tensile stress and the annealing time for the fully annealed specimens.

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