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Effect of grain size and orientation on the initial permeability of 36 wt % Ni–Fe alloys

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The effect of grain size and grain orientation on the initial permeability of a 36 wt % Ni–Fe alloy with additions of molybdenum, chromium and copper is reported. The initial permeability was found to increase with annealing temperature between 600° C and approximately 900° C due to the formation of a (1 2 3) [4 1 $\bar{2}$] primary recrystallization texture. Increasing the annealing temperature in the range 900 to 1100° C led to progressively lower permeabilities due to the growth of randomly oriented abnormal grains within the textured matrix. It is suggested that an increase in the misorientation between adjacent grains gives rise to an increase in the local magnetostatic energy, leading to much stronger pinning of magnetic domain walls, with a consequent decrease in permeability. Annealing at temperatures above 1100° C tends to increase the permeability. because of the increase in grain size.

1. Introduction

Interstitial elements such as carbon, oxygen, sulphur and nitrogen, together with second-phase particles and grain boundaries, are known to impede domain wall movement in the soft magnetic Ni–Fe alloys [1–6]. The final stage in the production of these alloys, therefore, involves a heat treatment at a high temperature in a reducing atmosphere which decreases the concentration of harmful interstitials and produces a strain-free fully recrystallized microstructure of large grain size, which gives a high relative initial permeability, μ_i.

Another factor which has been found to be important in the 50 wt % Ni–Fe alloy is the grain orientation [7–10]. The initial permeability is found to increase with an increase in the annealing temperature when the grains possess a random orientation. However, when a preferred orientation of the grains exists, the initial permeability does not begin to increase until the temperature is exceeded at which abnormal grain growth is initiated.

Inhibition of normal grain growth, and the subsequent onset of abnormal grain growth, has been observed to occur in sheet material when the grain size approaches the thickness of the sheet [11]. At this point the grain boundaries are perpendicular to the sheet surface and hence the driving force for grain growth is reduced.

2. Material and experimental method

A series of alloys based upon a commercial 36 wt % Ni–Fe alloy with additions of molybdenum, chromium and copper were used for this investigation, the nominal compositions being given in Table I. The material had been vacuum-induction melted and cold rolled into a strip of thickness 375 μm, the final reduction being ~ 60%. At every stage of manufacture precautions were taken to ensure that the level of impurities was kept to a minimum.

Heat treatment of the specimens for subsequent examination consisted of annealing for 4 h in a pure dry hydrogen atmosphere (dew point −70° C), at temperatures between 600 and 1200° C, followed by a furnace cool. Annular ring specimens of 25 mm o.d. and 17.5 mm i.d. were used for permeability measurements, and 25 mm × 25 mm specimens were punched or cut out from the strip for texture determinations.

The initial permeability was calculated from the inductance of a toroid consisting of a winding of ten turns of wire around a core formed from five rings, separated by paper laminations, in a

TABLE I Compositions of the research alloys

Alloy	Ni	Mo	Cr	Cu	Al	Mn	Si	Co	Fe
A	36.2	< 0.05	0.24	0.05	< 0.05	0.11	0.23	0.07	Bal
B	35.5	1.48	0.22	0.04	< 0.05	0.10	0.28	0.09	Bal
C	35.6	2.48	0.04	0.02	0.02	0.27	0.48	< 0.05	Bal
D	35.0	2.93	0.22	0.04	< 0.05	0.10	0.23	0.08	Bal
E	36.3	< 0.05	0.15	0.03	0.03	0.20	0.29	0.12	Bal
F	35.7	< 0.05	1.16	0.04	< 0.01	0.19	0.35	0.15	Bal
G	35.6	< 0.05	2.13	0.03	< 0.01	0.18	0.30	0.11	Bal
H	34.9	< 0.05	3.18	0.04	< 0.01	0.18	0.35	0.15	Bal
I	35.8	< 0.05	0.60	0.09	< 0.01	0.34	0.65	0.17	Bal
J	35.8	< 0.05	0.54	1.09	< 0.01	0.32	0.66	0.18	Bal
K	35.8	< 0.05	0.50	2.09	< 0.01	0.32	0.67	0.18	Bal
L	35.8	< 0.05	0.83	3.10	< 0.01	0.36	0.66	0.18	Bal

plastic former. A Wayne-Kerr model B224 a.c. bridge was used to measure the inductance with a field strength of $0.45\,\mathrm{A\,m^{-1}}$ at 1592 Hz. The bridge operated most accurately at 1592 Hz and, therefore, this frequency was chosen for comparing the initial permeability after different heat treatments. Additional experiments have shown that a linear relationship exists between the initial permeability measured at 50 and 1600 Hz, i.e.

$$\mu_{\mathrm{i,\,50\,Hz}} = 2.44\mu_{\mathrm{i,\,1600\,Hz}}.$$

Preferred orientation of the grains was determined using a texture goniometer with Cu*Kα* radiation. From the chart recording a {1 1 1} pole figure was constructed. Prior to any texture determination the heat-treated specimen was etched in an acidic ferric chloride solution to remove ~ 25 μm; this ensured that the texture was representative of the bulk of the material.

The change in the grain structure with annealing temperature was followed by optical microscopy, specimens from the heat-treated rings being prepared by standard metallographic techniques and etched in an acidic ferric chloride solution. Measurement of the mean boundary separation was obtained by using a standard line intercept

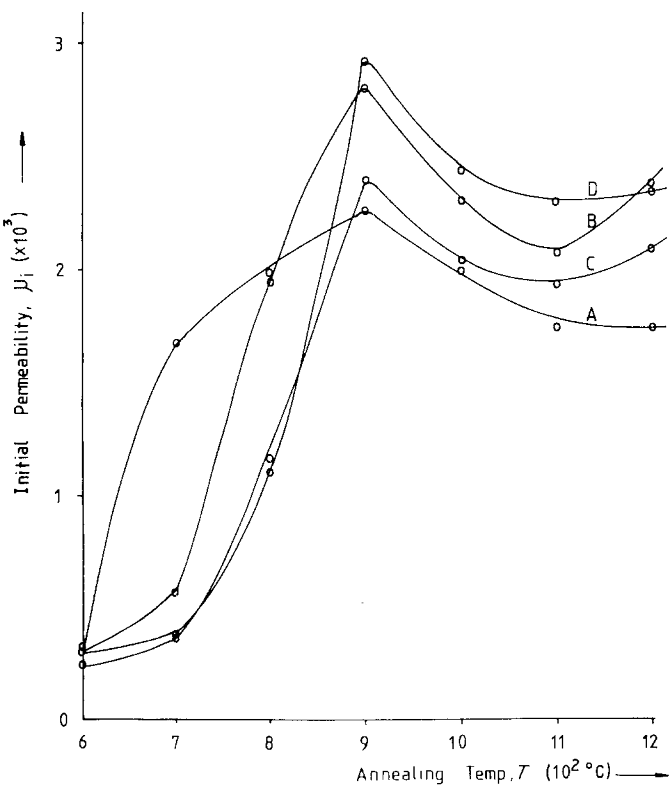

Figure 1 Effect of annealing temperature upon the initial permeability for the range of molybdenum additions.

method, and indirect determination of when recrystallization was complete was found by taking Vickers Hardness measurements using a load of 1 kg.

3. Experimental results

Figs. 1, 2 and 3 show the variation of initial permeability with annealing temperature for alloys with additions of molybdenum, chromium and copper, respectively. The variations of initial permeability, mean boundary separation, d, and Vickers hardness, H_V, with annealing temperature for the base alloy are compared in Fig. 4. All the permeability curves have the same general shape, with a maximum at around 900 to 1000° C, followed by a decrease in permeability and a levelling out, or rise, above 1100° C which depends upon the concentration of additional alloying element. It would therefore appear that the shape of the curves is primarily related to the heat treatment of the alloys and only secondarily to the alloying element.

In the cold-rolled condition the alloys had initial permeabilities of ~100 and there was very little change after annealing for 4 h until temperatures around 600° C had been exceeded. The shape of the Vickers hardness curve suggests that recrystallization is complete at ~800° C and this has been confirmed by optical microscopy (Fig. 5), together with transmission electron microscopy. In this low-temperature region up to 800° C the (0 0 1) [$\bar{3}$ 1 0] deformation texture (Fig. 6) became a (1 2 3) [4 1 $\bar{2}$] recrystallization texture (Fig. 7).

A decrease in the initial permeability compared with the value at about 900° C was observed after annealing at temperatures in the range 900 to 1100° C, which coincided with the appearance of large abnormal grains (Fig. 8), ~350 μm diameter, on the surface of the sheet, and extending across the sheet thickness normal to the surface. Together with the appearance of these abnormal grains, it was found that the mean boundary separation was approximately constant between 900 and 1000° C before increasing between 1000 and 1100° C. In this region the recrystallization texture was found to deteriorate (Fig. 9), until no texture could be detected at 1100° C.

Annealing at very high temperatures caused the initial permeability and boundary separation to increase.

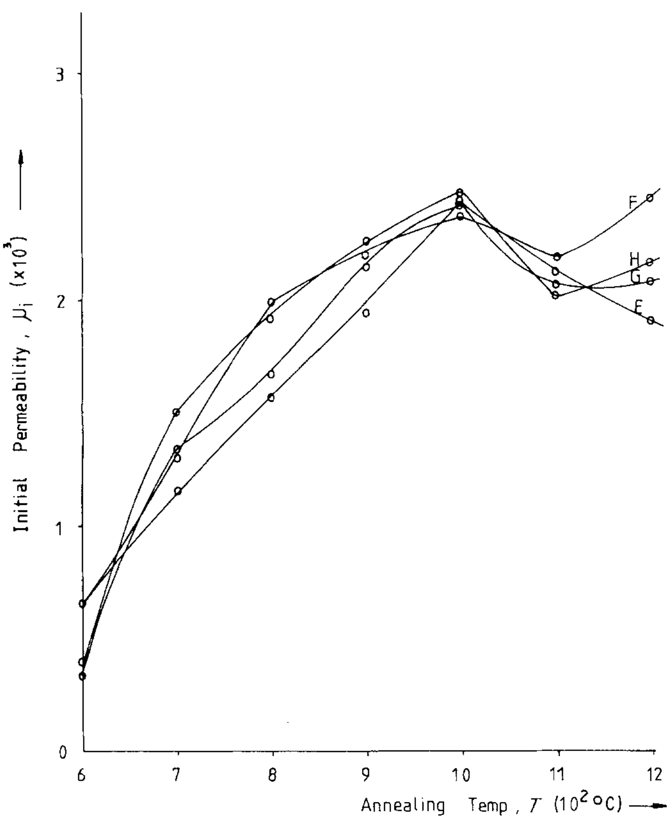

Figure 2 Effect of annealing temperature upon the initial permeability for the range of chromium additions.

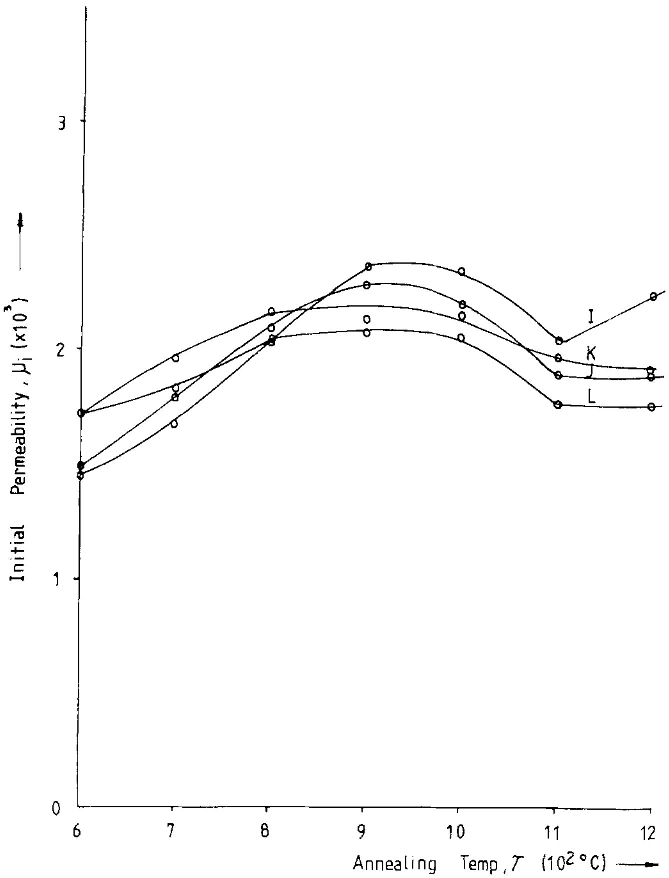

Figure 3 Effect of annealing temperature upon the initial permeability for the range of copper additions.

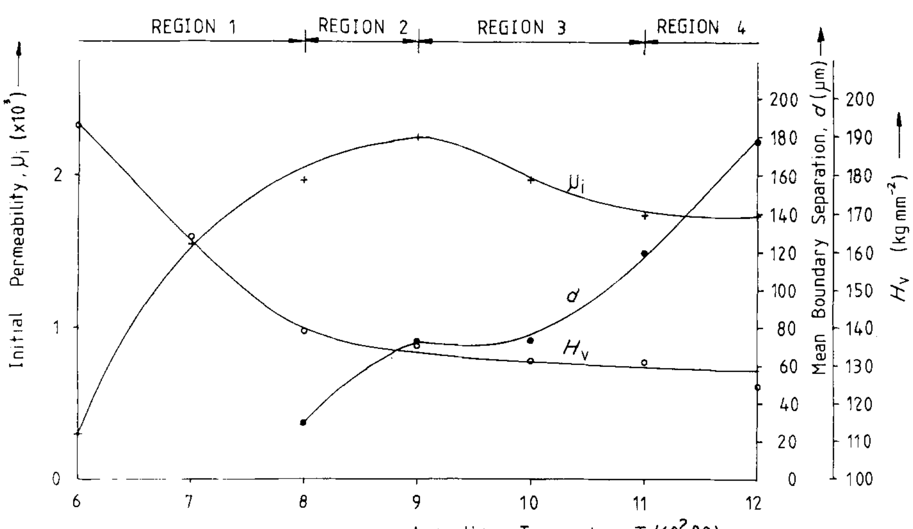

Figure 4 Variation of the initial permeability, mean boundary separation and H_V with the annealing temperature for 36 wt% Ni–Fe.

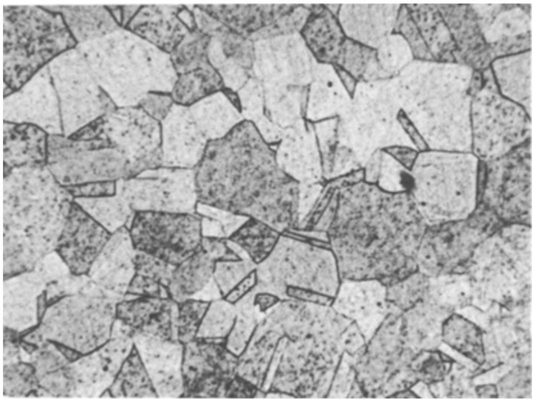

Figure 5 Recrystallized grain structure after annealing at 800° C for 4 h, × 153.

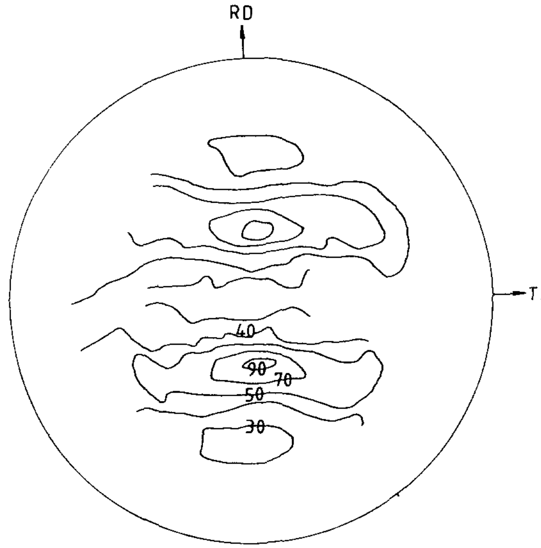

Figure 7 (1 2 3) [4 1 $\bar{2}$] recrystallization texture of 36 wt% Ni–Fe after annealing at 800° C for 4 h.

4. Discussion

The initial permeability curve can be divided into four distinct regions, as illustrated for the base alloy in Fig. 4, and each region may be explained in terms of the microstructural changes which occur.

4.1. Region 1

In the as-received condition the material contains a high dislocation density and, because of magneto-elastic interactions between domain walls and the stress field around dislocations, domain wall movement will be difficult. Hence, the initial permeability will be small but, as the temperature is raised to ~600° C when recovery takes place with the formation of a subgrain structure, the initial permeability increases owing to a decrease in the dislocation density. The amount of recrystallization produced during the 4 h anneal increases with increasing temperature above 600° C and the permeability increases rapidly. Once the material has fully recrystallized, around 800° C, resistance to domain wall movement is then due to the grain and twin boundaries.

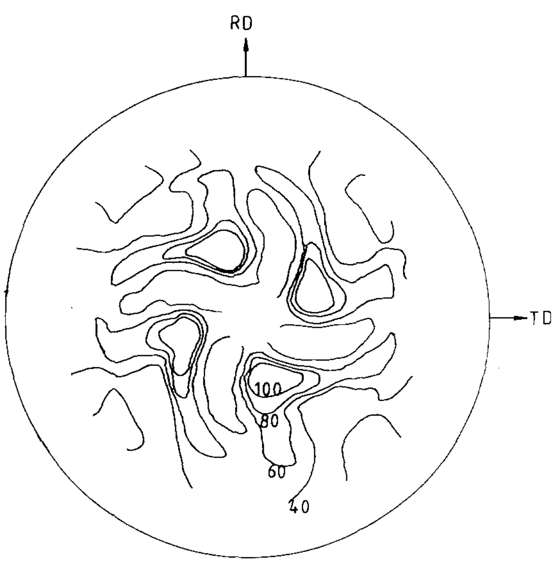

Figure 6 (0 0 1) [$\bar{3}$ 1 0] deformation texture of 36 wt% Ni–Fe.

4.2. Region 2

Between 800 and 900° C normal grain growth of the recrystallized grains leads to an increase in mean boundary separation, *d*, and a consequent increase in initial permeability, although the rate

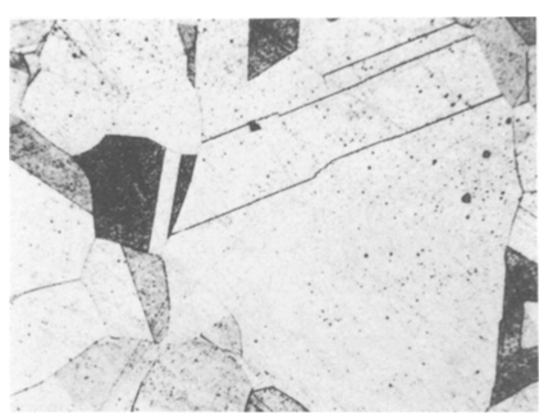

Figure 8 Large abnormal grain within the microstructure after annealing for 4 h at 1000° C, × 122.

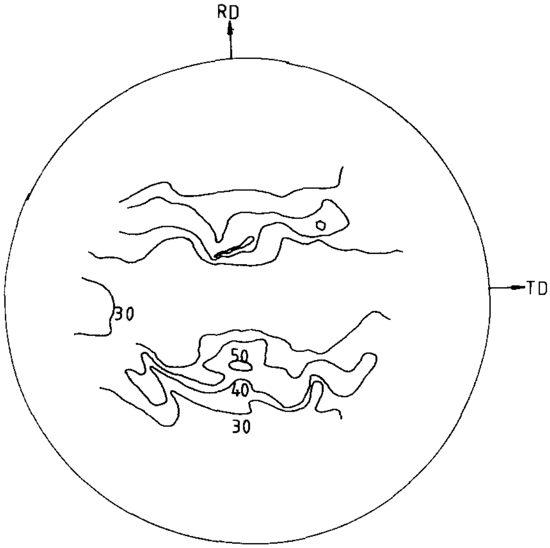

Figure 9 Deterioration of the recrystallization texture after 4 h anneal at 900° C.

of increase reduces and maximum permeability occurs after annealing at approximately 900° C, or somewhat higher for the chromium and copper alloys. One consequence of this normal grain growth is to cause the recrystallization texture to become weaker.

4.3. Region 3

Optical microscopy showed that at around 900° C the grains extended across the thickness of the sheet, and a plateau occurs in the mean boundary separation curve in Fig. 4. This suggests that normal grain growth is inhibited which is in agreement with other workers [11]. At this temperature large abnormal grains were observed on the sheet surface. The permeability reaches its maximum value at about 900° C and then decreases with the onset of abnormal grain growth. As the annealing temperature is increased up to 1100° C the recrystallization texture gradually deteriorates until no texture could be detected at 1100° C. This reduction in preferred orientation is due to the replacement of the oriented primary recrystallized grains by randomly oriented abnormal grains. Between 900 and 1100° C the microstructure will constist of textured primary recrystallized grains and randomly oriented abnormal grains. An increase in the boundary separation should yield a higher value of initial permeability, but the opposite is observed, and the decrease in permeability must be due to the increasing misorientation between adjacent grains as the textured grain structure is progressively replaced by a random structure.

4.4. Region 4

Once the microstructure consists of only large randomly oriented abnormal grains, any further increase in annealing temperature will result in normal grain growth. Hence, as the boundary separation becomes very large the misorientation across a boundary will have less influence upon the ease of domain wall movement and consequently, the initial permeability becomes constant for the base alloy and tends to increase with annealing temperature for alloys with molybdenum, chromium or copper additions. Magnetostriction measurements have shown that those alloys possessing the smallest value of saturation magnetostriction constant give the greatest initial permeability value in this region [12].

When two grains are in contact there may be a discontinuity in the component of magnetization normal to the grain boundary. This will result in a change of magnetostatic energy per unit area of the grain surface between the grains. As the misorientation across a boundary increases then the magnetostatic energy will also increase. If a preferred orientation of the grains exists then the magnetostatic energy will be low because the misorientation between the easy directions of magnetization in adjacent grains is small. Increasing misorientation will lead to stronger domain wall pinning and, eventually, a closure domain would be expected to form at a grain boundary in order to reduce the magnetostatic energy to zero. Formation of closure domains will result in strong pinning of domain walls due to an increase in domain wall area and, therefore, total domain wall energy together with an increase in magnetoelastic energy.

An explanation of the observed decrease in the initial permeability in region 3 can be postulated in terms of the misorientation across the grain boundaries. At around 900° C, where only textured grains are present, the boundaries should all be of a low misorientation. As the temperature is raised and abnormal grain growth is initiated, then some of the boundaries will be higher in misorientation and they will offer greater resistance to the movement of domain walls. Gradually more abnormal grains will replace the primary recrystallized grains and therefore more high misorientation boundaries will be present. Since the abnormal grains have a random orientation then it would be expected that when they contact a textured grain a closure domain will form at the boundary. Eventually a temperature is reached where only randomly oriented abnormal grains exist, and although there should be only high misorientation boundaries present, the grain size is so large that the influence of the boundaries is diminished, owing to a decrease in the number of grain boundaries.

The effect of misorientation across a grain boundary upon the ease of domain wall movement in these alloys has been studied by Lorentz electron microscopy [12] and the observations were consistent with the ideas presented above. Consequently, for a high initial permeablity a material should have a microstructure consisting of either primary recrystallized grains which have undergone normal grain growth and still retain a preferred orientation, or large abnormal grains which have replaced the primary recrystallized grains and undergone normal grain growth. These two microstructures are ultimately due to the final heat-treatment conditions.

5. Conclusion

It is concluded that there are two grain structures which can lead to a high initial permeability. By developing a strong preferred grain orientation the local increase in magnetostatic energy at a boundary is reduced and hence the boundary offers less resistance to domain wall motion. Alternatively, a large recrystallized grain size will offer less resistance owing to a decrease in the number of grain boundaries at which domain wall pinning may occur.

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