

Dependence of magnetic properties on atomic order in 77% Ni-14% Fe-5% Cu-4wt% Mo alloys

T. AKOMOLAFE

Department of Physics, University of Ilorin, Ilorin, Nigeria

G. W. JOHNSON

Metallurgy Department, University of Leeds, Leeds, UK

We have studied the effect of atomic order on the lattice parameter, a , Curie temperature, T_c , and the initial permeability, μ_i , of a series of molybdenum permalloys with varied Ni-Fe ratios and molybdenum concentrations. The ordering temperature range was between 380 and 600°C. The results of the effect of short-range order and long-range order on the lattice parameter after annealing for about 5 h between 420 and 470°C indicated a decrease of between 0.15 and 0.22 pm (0.04 to 0.06%) in the lattice parameter. No superlattice lines were detected. This may probably be due to the similarity in the atomic scattering factor of nickel and iron. It was also noted that both long-range and short-range ordering increased the Curie temperature of the ordered materials by about 1.4 to 6.6% due to the production of stronger and shorter Ni-Fe bonds. The number of Ni-Fe bonds, which controls the exchange force, was found to depend on the amount of order and molybdenum content in each material. The Curie temperature, which is a measure of the exchange force, is also an indirect means of measuring the degree of lattice ordering because the exchange integral is affected by metallurgical variables such as atomic ordering, composition, etc. Results of the isothermal annealing time on the initial permeability in the temperature range (380 to 600°C) indicated that between 380 and 460°C, maximum permeability was obtained at a critical degree of short-range ordering which is thought to correspond to a state when both the magnetostriction and anisotropy constants are close to zero. The maximum permeability was independent of ordering temperature in this range, although the time to reach this maximum decreases with increasing temperature. On annealing in the temperature range 460 to 500°C, the permeability reaches a maximum, the maximum permeability in this temperature range decreases with increasing temperature. As in the lower temperature range, the time to reach this maximum decreases with increasing temperature. Annealing between 500 and 600°C produced no maximum permeability. The permeability levelled off after an initial gradual increase. The activation energies for short-range order formation were found to be smaller (between 135 and 142 kJ mol⁻¹) in alloys with molybdenum concentrations above 4 wt% Mo (>2.5 at.% Mo) and higher activation energies (between 196 and 210 kJ mol⁻¹) in alloys with molybdenum concentrations below 4 wt% Mo (<2.5 at.% Mo) which suggests the formation of different ordered structures and mechanisms below and above 4 wt% Mo.

1. Introduction

The quaternary alloys of compositions near to 77% Ni-14% Fe-5% Cu-4wt% Mo, modifications of the binary stoichiometric composition Ni₃Fe commonly known as molybdenum permalloys, have unusually high permeabilities which are markedly influenced by composition and heat treatment. The changes in magnetic properties due to heat-treatment have been linked to changes in structure, internal strain, atomic order, etc. The attainment of high permeabilities of certain high-purity, strain-free complex Ni-Fe alloys in the region of Ni₃Fe are the result of simultaneously low magnetostriction and magnetocrystalline anisotropy constants [1-3]. The anisotropy is known to be very sensitive to lattice order and can be varied by

at least two orders of magnitude by changing the degree of order, but on the other hand, changes in order have less effect on the magnetostriction [4, 5], or on the magnetization [4, 6]. The degree of order is controlled by heat-treatment. In binary Ni-Fe alloys, the degree of order needed to achieve the highest permeability is extremely small and is attained by quenching the alloy from 600°C to room temperature [7-9], and this fact led Lewis [10] to suggest, as an explanation, that the quenching stresses, combined with the magnetostriction constants, yield a magneto-static energy equal and opposite to the anisotropy energy. The additions of non-ferromagnetic elements such as molybdenum, copper, tungsten, chromium, etc. to the basic binary Ni-Fe alloys resulted in increased

resistivity, permeability, ductility and simplified the heat treatment needed for optimum permeability. The high permeabilities obtainable from certain complex Ni-Fe alloys due to addition of molybdenum and copper are due to further reduction in the already small values of magnetostriction and anisotropy of the optimum binary alloy. The reduction is achieved by the introduction of non-ferromagnetic elements which decrease the proportion of magnetic nickel atoms, thereby reducing the density of the magnetic atom interactions which are responsible for the magnetostriction and the anisotropy.

In the quaternary alloys, domain wall displacements make the dominant contribution to the initial permeability [11], and the rotational processes are relatively unimportant. According to Smoluchowski [12], three changes could occur to the Curie temperature when ordering takes place in a material. First, the Curie temperature may not be affected if the exchange integral does not change on ordering. Second, the Curie temperature is increased if the ordering increases the exchange integral. And third, a decrease in Curie temperature can be produced if ordering decreases the exchange integral.

In the work reported in this paper, which forms part of a more general investigation of the effect of heat-treatment on the magnetic properties of 77% Ni permalloys [13], the rates and the degree of ordering were investigated by measuring the changes in the lattice parameter, Curie temperature and the initial permeability on ordering at different temperatures for a series of quaternary alloys in which Ni-Fe ratios and molybdenum concentrations were altered. The measurements of activation energies for short-range ordering (s.r.o.) will be used to explain the ordering mechanisms in these alloys.

2. Materials and experimental techniques

Table I shows the compositions of some of the alloys used during the investigation. The alloys were all commercially produced, and apart from basic alloying elements, they contained varying amounts of added and impurity elements such as sulphur, silicon, manganese, magnesium, aluminium, etc. Silicon, magnesium and manganese are added as deoxidants; however, aluminium and manganese in addition, are also used as desulphurizers and for improving hot workability, respectively. The materials were supplied in the form of 375 μm cold-rolled sheets from which annular specimens of internal and external diameters of 17.5 and 25.4 mm, respectively, were punched by the use of a specially designed punch and die. Most of the

TABLE I

Mo	Cu	Mn	Fe	Ni
3.58	4.96	0.67	14.5	Bal.
3.77	4.99	0.66	14.2	Bal.
3.80	5.00	0.68	13.9	Bal.
3.96	4.86	0.67	13.9	Bal.
4.18 (A)	4.96	0.66	13.6	Bal.
4.18 (B)	4.94	0.71	13.4	Bal.
4.34	5.02	0.65	13.2	Bal.

specimen preparations for the permeability, lattice parameter and Curie temperature measurements were carried out before the initial high temperature anneals in order to reduce handling strains. The initial high-temperature anneals of 4 h at 1100°C in purified hydrogen with a dew point reduced to -60°C were carried out on all the materials prior to the ordering treatment to produce some purification (by the removal of interstitials such as oxygen, silicon and nitrogen) and microstructural changes such as recovery, recrystallization, grain growth, etc. The specimens were either furnace-cooled to room temperature, the cooling rate in the ordering temperature range, 600 to 400°C, being about 2°C min^{-1} or quenched from above the ordering temperature range by withdrawing them into a water-cooled jacket, the quenching rate being estimated to be $120^\circ\text{C min}^{-1}$. The ordering anneals were performed at preset temperatures in purified dry hydrogen with constant flow followed by quenching after specified ordering times.

The initial permeability, μ_i , was determined from measurements of the inductance of a toroid whose core was formed by a series of annular ring specimens. The standard toroid was formed of five heat-treated specimens separated by paper laminations placed in a perspex former around which ten turns of PVC-covered copper wire were wound.

In the measurement of Curie temperature the instrumentation was identical to that used in the initial permeability measurements, but in this case paper insulations and plastic perspex former could not be used. Consequently, the specimens were coated with a suspension of high-purity magnesium oxide in distilled water and allowed to dry. This prevented sticking and also provided electrical insulation. Enamelled copper wires were used for the windings and the leads. The windings held the rings together. Care was taken during the winding so as to minimize the strains on the specimens. The leads were taken out of the furnace through vacuum seals. In order to avoid explosion due to the possibility of gas leakage occurring at the points where the leads were taken out of the furnace, a nitrogen atmosphere was used. The Curie temperature was measured by monitoring the variation of the inductance of the toroidally wound annulus with temperature and noting the temperature at which it fell precipitously to the value of an air-wound toroid.

Measurements were made with a Wayne Kerr Universal Bridge and a null detector tuned to 1592 Hz, the natural frequency of the internal oscillator of the Wayne Kerr Bridge.

3. Results and discussion

As pointed out previously (Section 2), the aim of this investigation was firstly to attempt to monitor the formation, development and rate of ordering in these alloys, and secondly, to investigate the influence of order on magnetic properties. Alloys of varying molybdenum concentrations with different Ni-Fe ratios were used.

3.1. Lattice parameters determination

In order to confirm the presence of long-range order

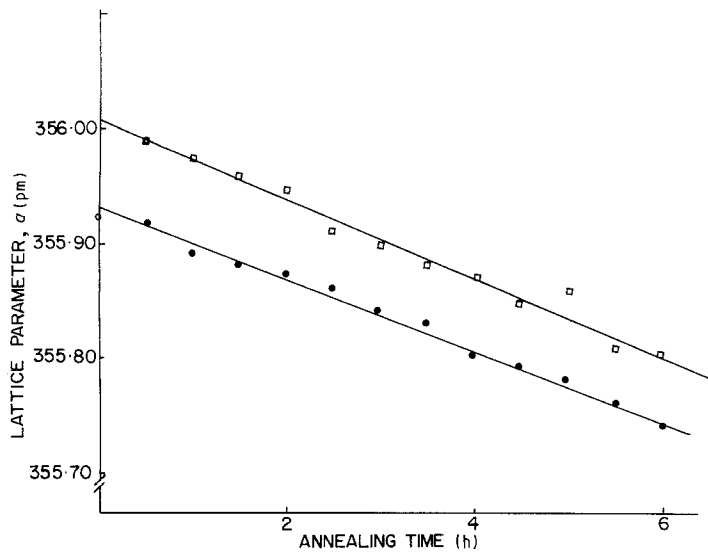


Figure 1 Graph of lattice parameter, a , against ordering time. (\square) 4.18 wt % Mo(A), (\bullet) 3.77 wt % Mo, annealing temperature 420°C.

(l.r.o.) in these materials, X-ray measurements were made to detect superlattice lines and to investigate the influence of order on lattice parameter. Even after annealing times of up to 40 h at 490°C, no superlattice lines were detected but there was a decrease in the lattice parameters. The inability to detect superlattice lines in these materials could be due to the similarity in the X-ray scattering factors of nickel and iron, an insufficiently long annealing period, or an incorrect cooling rate of the furnace used, about 2°C min⁻¹. This natural cooling rate of 2°C min⁻¹ for the furnace is too rapid for significant l.r.o. detection by superlattice lines to be obtained. Various workers who have succeeded in detecting superlattice lines in these materials by X-ray diffraction techniques, [6, 14] and neutron diffraction [15, 16] have employed an annealing period of over 100 h and a cooling rate of about 0.1°C h⁻¹.

Figs 1 and 2 which give the results of the investigation show a linear decrease in the lattice parameter of between 0.15 and 0.22 pm (0.04 to 0.06%) after annealing times of about 5 h. CuK α radiation was used. From Fig. 1 it can be seen that the lattice parameter is greater for an alloy with higher molybdenum concentration as would be expected because the atomic radius of molybdenum is greater than those of nickel and iron, even though the two materials were both annealed at 420°C. The slopes of the lines in this figure show that the 4.18% Mo (A) alloy orders slightly

more rapidly than the 3.77% Mo alloy, confirming the observations that there is a difference in ordering rate and possibly ordered structure, for compositions above and below 4 wt % Mo. Comparison of the slopes of the lines in Fig. 2 for the 3.58% Mo alloy annealed at 420 and 470°C shows that ordering proceeds more rapidly at 470°C than at 420°C as inferred from the permeability measurements to be discussed later.

The magnitude of the decrease in the lattice parameter of some Ni-Fe alloys is seen to depend on composition as obtained by these results and this has been supported by the work of earlier researchers on these materials. While a decrease of 0.15 to 0.22 pm in lattice parameters were obtained during these investigations, Taylor *et al.* [17] recorded a decrease of about 0.14 pm (0.04%) on some quaternary alloys, Lutts and Gullen [18] obtained a decrease of about 0.16 pm (0.017%) in lattice parameter of quenched and annealed materials using alloys of composition 73.35 and 74.45 wt % Ni.

3.2. Curie temperature measurements

Although no superlattice lines were detected by X-ray diffraction, the existence of order formation in these alloys was deduced from the variation of lattice parameter, magnetic properties (to be discussed) and resistivity measurements [13]. Hence it was decided to investigate the influence of atomic ordering and

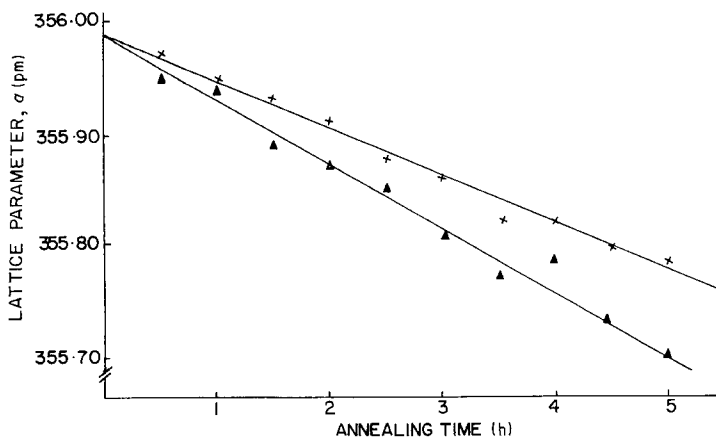


Figure 2 Graph of lattice parameter, a , against ordering time. 3.58 wt % Mo, (\times) annealed at 420°C, (\blacktriangle) annealed at 470°C.

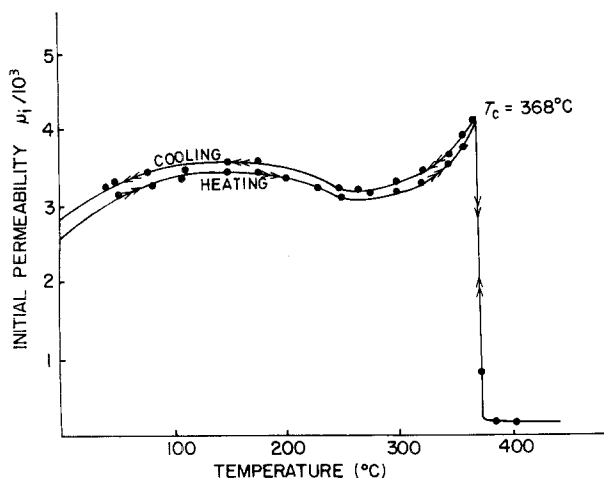


Figure 3 Graph of initial permeability, μ_i , against temperature, T , 3.80 wt % Mo, furnace cooled.

composition on the Curie temperature. Five compositions 3.77% Mo, 3.80% Mo, 4.18% Mo (A), 4.18% Mo (B) and 4.34 wt % Mo were used after either slowly cooling or quenching from 600°C to room temperature following the initial anneals of 4 h at 1100°C. The Curie temperature, T_θ , was determined by measuring the permeability as the ring specimens were slowly heated to above T_θ and then slowly cooled. An extremely sharp change in permeability occurs at the Curie temperature.

Figs 3 and 4 show the results obtained for the slowly cooled and quenched alloy with 3.80 wt % Mo concentration, respectively. Common to the two figures are two maxima, one at the lower temperature, while the second occurred at the Curie point.

The first maximum at lower temperature is attained by gradual increase in permeability. The reason for this maximum (only observable when measurements were made under a very small field) is the crystal anisotropy which approaches zero at a lower temperature than does the spontaneous magnetization. If the thermal expansion is taken into consideration, Carr [19] predicted that the relationship between the ferromagnetic anisotropy constant, temperature and

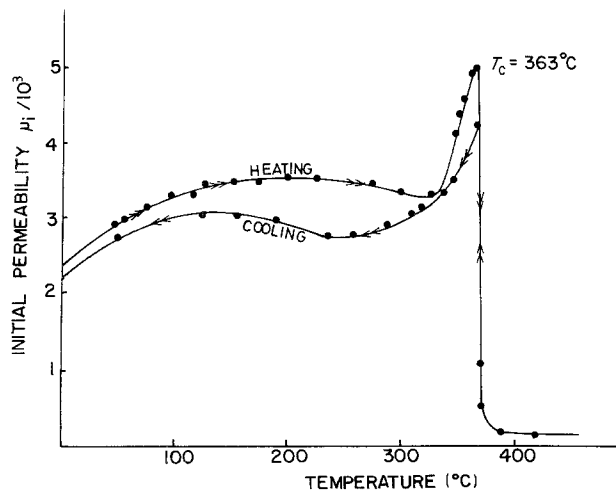


Figure 4 Graph of initial permeability, μ_i , against temperature, T , 3.80 wt % Mo quenched from 600°C.

saturation magnetization is of the form:

$$K_1(T) = K_1(0, 0) \cdot \left(1 - c \frac{T}{T_\theta}\right) \left(\frac{M}{M_0}\right)^{10} \quad (1)$$

where $K_1(T)$ is the first crystal anisotropy constant at temperature T , $K_1(0, 0)$ is the anisotropy constant for zero temperature and zero strain, T_θ is the Curie temperature, M is the saturation magnetization at temperature T , M_0 is the saturation magnetization at absolute zero and c is a constant.

At this first maximum, $K_1(T)$ is zero and $c = T_\theta/T$. The results of this investigation gave values of c which vary from 1.73 to 2.03, in fair agreement with the value of 1.74 obtained by Carr. The wide variation is due to the broadness of the maximum and to different degrees of order in each specimen.

The second maximum at the Curie point was much sharper than the first maximum at a lower temperature. The rapid rise in permeability as the second maximum is approached is due to both the anisotropy and magnetostriction tending to zero as the Curie temperature is approached. Usually both the anisotropy and magnetostriction decrease with increasing temperature and attain zero values at the Curie temperature. Because the observed anisotropy is a function of saturation magnetization (Equation 1), and as the saturation magnetization is zero at the Curie temperature, it then becomes obvious that the crystal anisotropy should be zero at the Curie temperature. Above the Curie point the specimens became paramagnetic, the permeability falls very rapidly and sharply to a very small value. No intermediate readings were possible. The materials were then heated to 30°C above the Curie temperature, left at this temperature for about 30 min, and measurements repeated during the cooling process. In all cases, the permeability showed hysteresis effects. The permeability of the slowly cooled alloys was always greater during cooling than during the heating process. The opposite was found for the quenched alloys as shown in Fig. 4, the permeability being higher during heating than during cooling (with the exception of the 4.18 wt % Mo (B) alloy). The increase in permeability during cooling (for furnace-cooled materials) could be due to slow diffusion processes which take place at temperatures slightly higher than the Curie temperature, and lead to an increase in s.r.o. In the case of the quenched specimens, which have a smaller amount of s.r.o. initially, the rate of ordering may be less, and the increased order insufficient to overcome a hysteresis effect which would be expected to lower the permeability on cooling through the Curie temperature.

Table II shows the effect of heat-treatment and

TABLE II

Material	Curie temperature (°C)	
	Furnace-cooled	Quenched at 600°C
3.77 wt % Mo	373	350
3.80 wt % Mo	368	363
4.18 wt % Mo (A)	355	346
4.18 wt % Mo (B)	357	347
4.34 wt % Mo	353	343

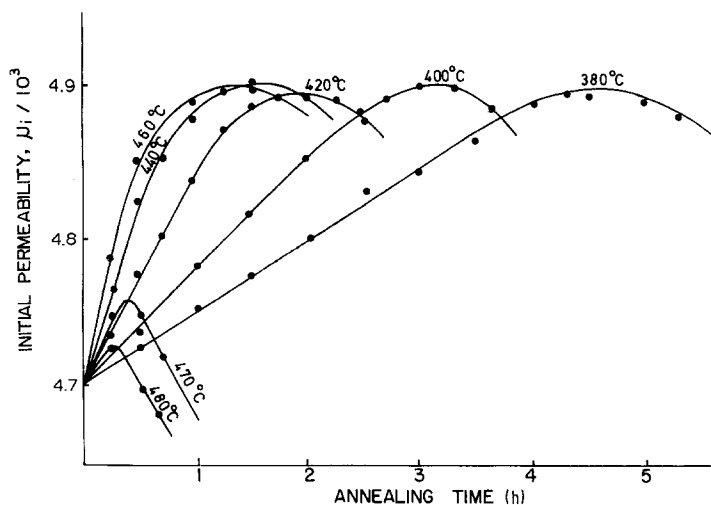


Figure 5 Graph of initial permeability, μ_i , against ordering time.

composition on the Curie temperature. From these results it is seen that the slowly cooled alloys have slightly higher Curie temperatures than the quenched alloys. The reason for this is that the slowly cooled (furnace-cooled) alloys have a substantial degree of atomic ordering. The implication of this is that the ordering produces more of the stronger and shorter bonds between the nickel and iron atoms rather than between Ni-Ni and Fe-Fe atoms. Consequently, this causes the exchange integral, J_{ex} , to increase. This result is in a very good agreement with the prediction of Smoluchowski [12] who stated that the degree of order, which influences the numbers of the different types of bond, causes the exchange integral to change to a value given by

$$J_{ex} = 2J_{Ni-Fe} - (J_{Ni-Ni} + J_{Fe-Fe}) \quad (2)$$

where J_{ex} is the overall exchange integral in the alloy, J_{Ni-Ni} is the exchange integral due to Ni-Ni interactions, J_{Fe-Fe} is the exchange integral due to Fe-Fe interactions, and J_{Ni-Fe} is the exchange integral due to Ni-Fe interactions. Thus an ordering which favours the formation of stronger and shorter bonds between nickel and iron atoms will cause the overall exchange integral J_{ex} to increase.

Secondly, it is known that the approximate relationship between the Curie temperature, T_θ , and the exchange integral, J_{ex} , as given by Smoluchowski [12] is of the form

$$T_\theta = \frac{2ZS(S+1)J_{ex}}{3k} \quad (3)$$

where Z is the number of nearest neighbours, S is the electron spin quantum number ($=\frac{1}{2}$) and k is Boltzmann's constant.

From Equation 3 it is seen that the Curie temperature, T_θ , is proportional to the exchange integral, J_{ex} , and an increase in the exchange integral thus causes a higher Curie temperature. Hence the increase in the Curie point, as shown by these results for ordered alloys, is justified. From Table II it could be seen that as the molybdenum concentration increases the Curie point tends to decrease for both ordered and quenched alloys. This could be due to increasing concentrations of molybdenum reducing the number of Ni-Fe nearest neighbours, Z , and so reducing J_{ex} and the Curie temperature.

3.3. Permeability measurements

Before permeability measurements were performed the materials were given an ordering heat-treatment. The ordering heat-treatment consisted of an initial anneal of the materials at 1100°C for 4 h in purified dry hydrogen to produce some purification and microstructural changes. After the initial anneal the materials were isothermally annealed at preset temperatures in order to vary the degree of order in the materials. The ordering anneals were carried out between 380 and 600°C. The ordering temperature range was divided into two temperature zones: (a) below 500°C (i.e. temperatures below the order-disorder temperature); (b) between 500 and 600°C (temperatures above the order-disorder temperature).

The rate of order formation in these alloys was studied by monitoring the change in permeability with annealing time at various temperatures.

Figs 5 and 6 show the effect of ordering anneals between 380 and 600°C for an alloy containing 4.18 wt% Mo (A). All the materials used for the results in Fig. 5 were furnace cooled to room temperature after the initial anneal of 4 h at 1100°C. From the results in Fig. 5 the following points may be seen.

(i) The permeability reaches a maximum at different times at different temperatures. The maximum is independent of temperature at temperatures below 460°C but falls as the annealing temperature is increased above 460°C. On continuation of annealing after the maximum is reached, the permeability falls. The explanation for this behaviour is that as the degree of s.r.o. increases with annealing time permeability also increases, and at an optimum degree of s.r.o. as discussed earlier, the maximum in permeability is attained. On further annealing, the optimum degree of s.r.o. is exceeded and this causes the permeability to drop. Hence it could be concluded that at an optimum degree of order, both the anisotropy and magnetostriction are vanishingly small, hence the attainment of maximum permeability.

(ii) Between 460 and 490°C the maximum permeability attained decreases with increasing annealing temperature and the time to attain the maximum permeability also decreases with increasing annealing temperature. The reason for the fall in maximum permeability is due to the fact that s.r.o. is no longer

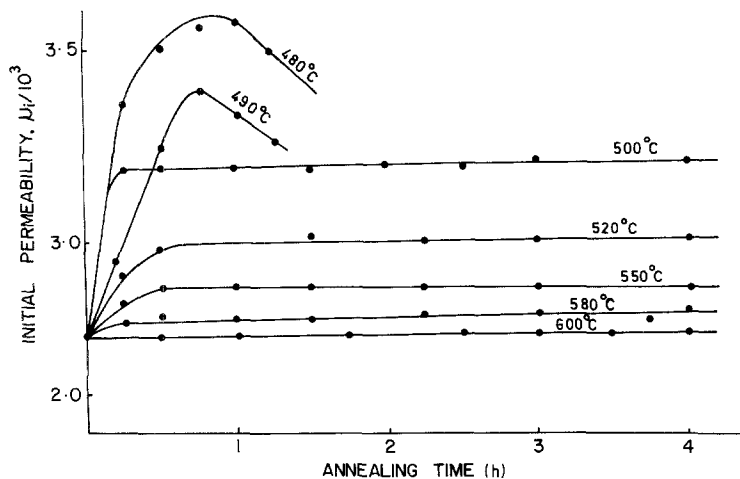


Figure 6 Graph of initial permeability, μ_i , against ordering time.

the only controlling factor on permeability (as in the lower temperature zones). The decrease in maximum permeability (in this region) could be due to the pinning of the domain walls by microdomains of l.r.o. which might have nucleated and grown [17]. When the microdomains reach a critical size the permeability reaches a maximum which is less than that at the lower temperature range where only s.r.o. occurs. This proposed occurrence of l.r.o. is consistent with the change in resistivity on annealing [13]. Also, when the materials are heated to 600°C for a few minutes (the temperature above the order-disorder transformation), furnace cooled to room temperature, and then reannealed in the temperature range 400 to 460°C, the initial permeability increased to its expected maximum. This is consistent with the destruction of l.r.o. in the materials when they are heated above the order-disorder temperature. These results are consistent with the works of Bozorth [1-3], Richards *et al.* [20], Enoch *et al.* [21-23] and other workers who argued that in high-purity complex Ni-Fe alloys the attainment of high permeability is possible at a critical degree of order.

Fig. 6 shows the effect of annealing time on the initial permeability in the annealing range 460 to 600°C. All the specimens were quenched at 600°C after the initial anneals of 4 h at 1100°C so that the initial amount of s.r.o. would be small. The ordering anneals were similar to those described above, i.e. the materials were annealed at preset temperatures

followed by rapid quenching. Below 500°C the material reaches a maximum permeability and then the permeability decreases (already discussed). Between 500 and 600°C no such maximum was obtained. The permeability increased initially and then levelled off. The levelling off is due to the attainment of an equilibrium degree of s.r.o. which is less than that required to obtain maximum permeability in the alloys. The equilibrium degree of s.r.o. decreases with increasing annealing temperature and at 600°C there is a negligible amount of order formation. As the annealing temperature was higher than the order-disorder transformation temperature, no l.r.o. formation is expected in this temperature zone. In order to justify the assumption that permeability is controlled by s.r.o. some quenching experiments were performed. Above 600°C (i.e. between 600 and 1100°C) the permeability of the alloy increased with quenching temperature. This is because as the quenching temperature increases the concentration of quenched-in vacancies in the alloys also increases and this promotes the formation of atomic ordering. Although the permeability increases with quenching temperature the maximum permeability in this region is less than that obtained in the temperature range 380 to 490°C. The reason for this is that the amount of s.r.o. produced by quenched-in vacancies due to quenching at high temperature is about 60% of that produced by diffusion due to the process of slow cooling [24]. Secondly, there will be vacancy loss during quenching

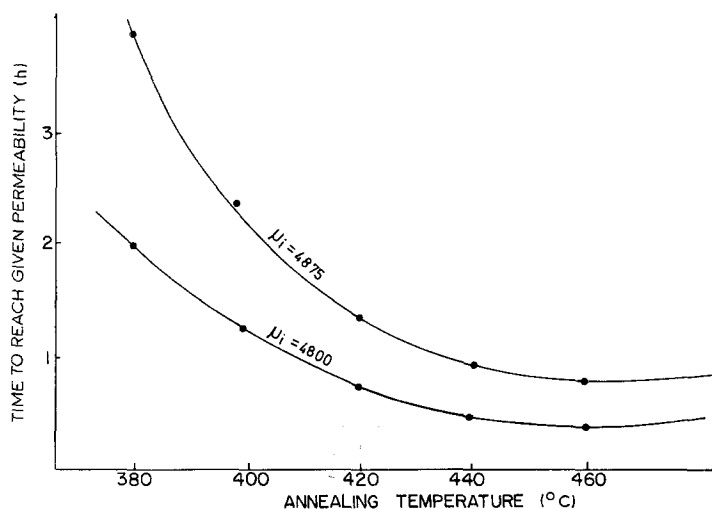


Figure 7 Time to reach a given initial permeability, μ_i , plotted against ordering temperature, T .

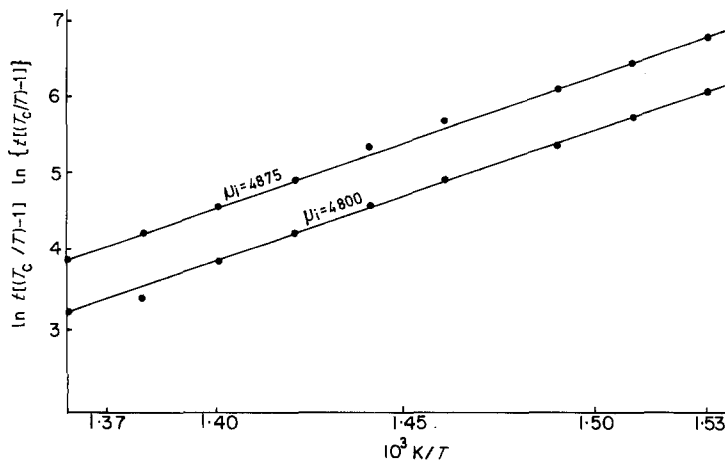


Figure 8 Graph of $\ln \{t[(T_c/T) - 1]\}$ against $1/T$ for calculating the activation energy, Q .

due to annihilation at vacancy sinks and the permeability may be reduced due to quenching strain which increases as quenching temperature increases.

In order to investigate the effect of molybdenum concentration on the diffusion-controlled ordering rate, the activation energies of s.r.o. formation were estimated. This is because from the measurements of activation energy we can understand the ordering rate better. The time, t , required to produce a given state of order is given by Taylor and Johnson [17] as

$$t \propto \left(\frac{T_c}{T} - 1 \right)^{-1} \exp(Q/kT)$$

or

$$t \left(\frac{T_c}{T} - 1 \right) \propto \exp(Q/kT) \quad (4)$$

where Q is the activation energy, T_c is the order-disorder temperature, and T is the annealing (or ordering) temperature.

Fig. 7 is derived from Fig. 5 and shows the plots of the time required to reach a given permeability (4875 and 4800) against the ordering temperature. The time decreases as ordering temperature increases. From Fig. 7 a graph of $\ln \{t[(T_c/T) - 1]\}$ against $1/T$ was plotted and is shown in Fig. 8. The two curves in the figure obey a linear relationship and by using Equation 4 the activation energy was calculated. The values of activation energies for various materials are shown in Table III.

The difference in activation energy between materials with molybdenum contents less than 4 wt % and those with higher molybdenum contents (> 4 wt % Mo), gives an indication that the materials with molybdenum contents greater than 4 wt % Mo order more rapidly than those with lower molybdenum concentrations and may have a different ordered structure. These findings led to a further investigation of the influence

of molybdenum content on permeability after furnace cooling or a quench at 600°C . Fig. 9 shows the results of the investigation. It is seen from these results that permeability drops as molybdenum increases and at about 4 wt % Mo permeability rises again. The maximum permeabilities obtained in the isothermal anneals for various materials are also summarized in Fig. 9. The plot shows a sharp change at 4 wt % Mo. This break at about 4 wt % Mo and the different activation energies above and below this concentration are thought to be due to different ordering mechanisms above and below molybdenum concentrations of about 4 wt %. Yeganyan and Selitsky [25] with work on Ni_3Fe -base alloys containing small additions of Group IV elements, found that there existed a critical concentration of alloying element (6 at.% Cr, 3.8 at.% Mo) which divided alloys with different capacities for ordering. Taylor and Johnson [17] observed a similar behaviour near to molybdenum concentrations of 4 wt %. Also changes in permeability when the molybdenum content exceeds 4% Mo could be due to

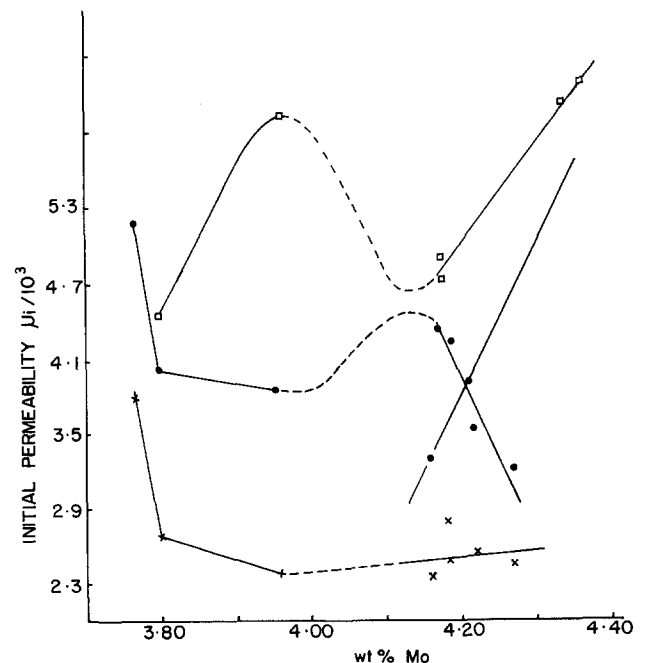


Figure 9 Variation of the initial permeability, μ_i with molybdenum concentrations. (\square) Maximum μ_i in isothermal anneals for various compositions. (\bullet) Furnace cooled. (\times) Quenched from 600°C to room temperature.

TABLE III

Material	Activation energy, Q (kJ mol^{-1})
3.80 wt % Mo	210
3.96 wt % Mo	196
4.18 wt % Mo (A)	142
4.18 wt % Mo (B)	137
4.34 wt % Mo	130

differences in the already small value of both the anisotropy constant and magnetostriction.

4. Conclusion

There was a decrease in lattice parameter between 0.15 to 0.22 pm (0.04 to 0.06%) which confirms the occurrence of atomic ordering in these materials. It was also found that ordering increases the Curie temperature of the ordered alloys by about 1.4 to 6.6% due to the formation or production of stronger and shorter bonds between the nickel and iron atoms which increases the overall exchange energy. Maximum permeability was obtained at a critical degree of s.r.o. which is thought to correspond to a state when both the anisotropy and magnetostriction constants are close to zero. The maximum permeability was independent of ordering temperature below 460°C, but the time taken to reach this maximum permeability decreased with increasing annealing temperature. On annealing in the temperature range between 460 and 500°C, the maximum permeabilities decreased and this was associated with the nucleation and growth of microdomains of l.r.o. in short-ordered matrix. These microdomains pin domain walls and reduce the permeability. Annealing between 500 and 600°C produced equilibrium amounts of s.r.o. which were less than the critical amount required for maximum permeability and permeability increased initially with annealing time and then became constant.

Quenching experiments show that between 600 and 1100°C there was a slight increase in permeability which was due to enhanced ordering by quenched-in vacancies. The maximum permeability attained in this region was smaller than the lower temperature region because the amount of s.r.o. produced by quenched-in vacancies is about 60% of that produced by diffusion-enhanced ordering. The activation energy for order formation was found to be smaller in alloys with molybdenum concentrations above 4 wt % Mo. This is possibly due to the formation of different ordered structures above 4 wt % Mo.

References

1. R. M. BOZORTH and D. L. BOOTHBY, *J. Appl. Phys.* **18** (1947) 173.
2. R. M. BOZORTH, "Ferromagnetism", (Van Nostrand, New York, 1951).
3. *Idem*, *Rev. Mod. Phys.* **25** (1953) 42.
4. R. C. JACKSON and E. W. LEE, *J. Mater. Sci.* **1** (1966) 362.
5. H. H. SCHOLEFIELD, R. V. MAJOR and B. GIBSON, *Br. J. Appl. Phys.* **18** (1967) 41.
6. R. J. WAKELIN and E. L. YATES, *Proc. Phys. Soc.* **B66** (1953) 221.
7. G. W. ELMEN and H. D. ARNOD, *J. Frank. Inst.* **195** (1923) 651.
8. G. W. ELMEN, *ibid.* **206** (1928) 317.
9. *Idem*, *ibid.* **207** (1929) 583.
10. M. LEWIS, *Br. J. Appl. Phys.* **15** (1964) 407.
11. P. G. COLLAR, PhD thesis, Sheffield University, UK (1966).
12. A. SMOLUCHOWSKI, *J. Phys. Rad.* **12** (1951) 389.
13. T. AKOMOLAFE, PhD thesis, University of Leeds, UK (1983).
14. P. LEECH and C. SYKES, *Phil. Mag.* **27** (1938) 742.
15. C. G. SHULL and M. K. WILKINSON, *Br. J. Appl. Phys.* **97** (1955) 304.
16. V. I. GOMAN'KOV, I. M. PUZEI and A. A. LOSHIMANOV, *Fiz. Met. Metall.* **21** (1966) 134.
17. E. G. TAYLOR and G. W. JOHNSON, *J. Mater. Sci.* **12** (1977) 51.
18. A. LUTTS and P. M. GULLEN, *Phys. Status Solidi* **41** (1970) 481.
19. W. J. CARR Jr, *J. Appl. Phys.* **29** (1958) 436.
20. C. E. RICHARDS, A. C. LYNCH and E. V. WALKER, *Proc. IEEB* **104** (1957) 343.
21. R. D. ENOCH and A. D. FUDGE, *Br. J. Appl. Phys.* **17** (1966) 623.
22. R. D. ENOCH and A. WINTERBORN, *IEE Conf. Publ.* **33** (1967) 123.
23. R. D. ENOCH and D. L. MURRELL, *J. Appl. Phys.* **42** (1971) 133.
24. A. FERRO and G. GRIFFA, *J. Phys. Chem. Sol.* (1970) 2789.
25. I. L. YEGANYAN and Y. P. SELISSKY, *Fiz. Met. Metall.* **27** (1967) 199.

Received 14 January
and accepted 3 June 1987