Some magnetic and mechanical properties of fibre-reinforced nickel and nickel-iron alloys

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Composites containing tungsten wires reinforcing nickel and nickel–iron alloy matrices have been fabricated by a filament winding–electroplating technique and a considerable improvement in the tensile strength was achieved relative to the unreinforced matrix. The presence of the fibres was found to have a significant influence on the magnetic properties of the composites measured in the direction of the fibre axes. In composites having a matrix with a negative magnetostriction, the maximum permeability decreased with increasing volume fraction, V_f, and was also dependent on the fibre diameter and the magnitude of the magnetostriction. In cases where the matrix had a positive magnetostriction the maximum permeability was observed to increase with increasing V_f, reaching a peak value at $V_f \simeq 0.1$. It was suggested that the presence of stresses induced in the matrix during cooling from the heat-treatment temperature, due to the difference in the thermal expansion between the fibre and matrix, could explain this magnetic behaviour. By theoretical considerations, the peak was shown to coincide approximately with the volume fraction at which the maximum, uniaxial elastic stress was expected to form in the matrix. Above this volume fraction the uniaxial and transverse stresses became sufficiently high to cause plastic deformation in the entire matrix leading to the observed fall in the maximum permeability, although in all cases the value remained above that shown by the unreinforced matrix.

1. Introduction

The most practical high strength, soft magnetic alloys developed to date are the cobalt-based alloy, "Nivco" ($Bs = 12\,000$ G; $H_c = 11.5$ Oe) [1] and the NASA superalloy (Co–25W–1Ti–$\frac{1}{2}$Zr–$\frac{1}{2}$C; $B_s = 18\,500$ G; $H_c = 27.3$ Oe) [2]. Although these alloys exhibit a high tensile strength in the region of 1200 MN m^{-2}, they nevertheless have a relatively high coercive force, H_c, compared with the more conventional soft magnetic alloys.

Reinforcement of soft magnetic materials with high strength fibres has recently attracted some interest for potential use at elevated temperatures [3]. Such a means of strengthening may lead to significant increases in mechanical strength without impairing the magnetic properties to the degree associated with the more conventional strengthening mechanisms such as precipitation or dispersion hardening. Although the presence of fibres in a ferromagnetic matrix may impede domain wall motion to some extent, this may not be as detrimental to the magnetic properties as second phases in precipitation or dispersion-hardened systems, as the comparitively large fibres, used in most practical composite materials, have associated large inter-fibre spacings where the domain walls can move with relative ease.

A more important factor in fibre-reinforced materials may be the complex state of stress which can arise during the fabrication of a composite. The magnetic properties of most ferromagnetic materials change significantly with the application of stress, which may be ranked with the magnetic field strength and temperature as one of the most important factors affecting the magnetization.

Colling [3], has investigated some magnetic properties of a 50% nickel–iron alloy reinforced with tungsten wires. Various other compositions of this alloy show interesting magnetic properties such as high permeability at 79%Ni; positive magnetostriction at less than 82%Ni and negative magnetostriction at greater than 82%Ni. For this reason this material was considered suitable for further investigation. In this paper the fabrication of nickel and nickel–iron alloy/tungsten wire composites is described and the influence the fibres have on the magnetic properties is examined.

2. Experimental procedure

Various methods were developed in an attempt to fabricate a fibre-reinforced soft magnetic material which were based on sintering, hot-rolling or casting. Only partial success was achieved by these techniques as the high temperatures or pressures required caused fibre matrix inter-diffusion and fibre damage both of which resulted in a degradation of the mechanical properties of the composites. Electroplating, followed by a relatively low temperature anneal was finally selected as the most suitable method to produce this material.

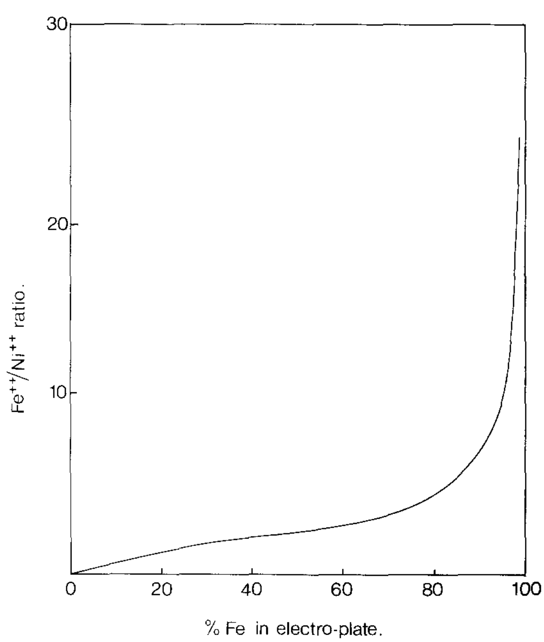

Figure 1 Relationship between the percentage of iron in iron–nickel electroplate and the weight ratio of Fe^{2+}/Ni^{2+} in the plating solution.

The composites in the present work were fabricated by a filament winding–electroplating process. Tungsten wire ranging in nominal diameter from 20 to 50 μm was wound onto a stainless steel mandrel at predetermined spacings by the use of a lathe. Nickel or a nickel–iron alloy was plated onto the mandrel for an appropriate time to give the required volume fraction. During plating, the mandrel was slowly rotated in the bath and a smooth, fine grained deposit between 50 and 100 μm thick was obtained. The electroplating bath used in this work was based on nickel and iron chlorides. A typical composition and operating conditions used to produce an 88% nickel–iron alloy were as follows:

$NiCl_2 \cdot 6H_2O$	200 g l^{-1}
$FeCl_2 \cdot 4H_2O$	200 g l^{-1}
boric acid	38 g l^{-1}
sodium lauryl sulphate	0.42 g l^{-1}
HCl to control bath pH at 1.5	
current density	1 A dm^{-1}
temperature	90–95° C
anodes	depolarized nickel and ferrovac iron

The nickel–iron ratio in the electroplate was varied by altering the Fe^{2+}/Ni^{2+} ratio in the plating bath, and the relationship between the solution and alloy composition is shown in Fig. 1.

After stripping the material from the mandrel it was heat-treated at 700° C for 1 h in dry argon–1% hydrogen and then air cooled. This treatment gave a ductile material with a mean grain diameter of approximately 5 μm.

The magnetic properties of the composites and the un-reinforced matrix were determined in the direction of the fibres using a null astatic magnetometer of a type similar to that described by Tobusch [4] using a maximum applied field of approximately 5 Oe. Initial magnetization and hysteresis curves were determined for specimens 30 mm long and 3 mm wide containing a monolayer of tungsten wires. Subsequent to the measurement of the magnetic properties the fibre volume fraction, V_f, of the composites was accurately determined by dissolution of the matrix in dilute nitric acid and weighing of the fibres. The nickel content of the alloys was determined gravimetrically by precipitation as nickel dimethylglyoxime. In addition a number of specimens were selected for tensile testing which was performed

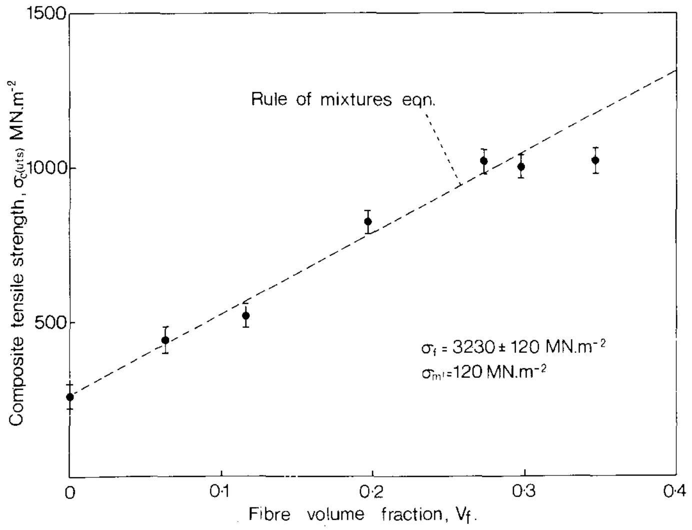

Figure 2 Dependence of the tensile strength on the fibre volume fraction for composites with 20 μm diameter tungsten wires reinforcing an 88% Ni–Fe alloy matrix.

on an Instron tensile machine at a strain-rate of 0.5 mm min^{-1}.

3. Results

The composite tensile strength σ_c (UTS) is given by the rule of mixture equation:

$$\sigma_c\,(\mathrm{UTS}) = \sigma_f V_f + \sigma'_m(1 - V_f) \qquad (1)$$

where σ_f is the UTS of the fibres and σ'_m is the stress carried by the matrix at the point of fracture.

Fig. 2 illustrates that there is close agreement between the experimental values of the composite tensile strength and Equation 1 indicating that effective reinforcement is obtained in this material. The tensile strength of the composites with

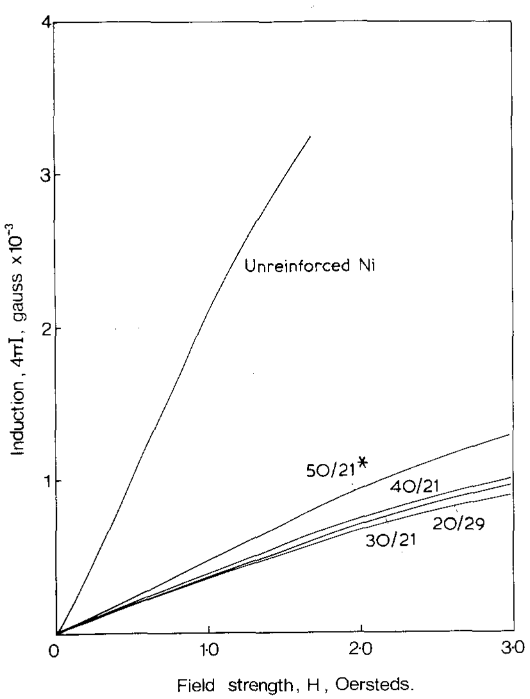

Figure 3 Magnetization curves of unreinforced nickel and nickel–tungsten composites containing various wire diameters (low V_f). *Nominal wire diameter (μm) and volume fraction, respectively.

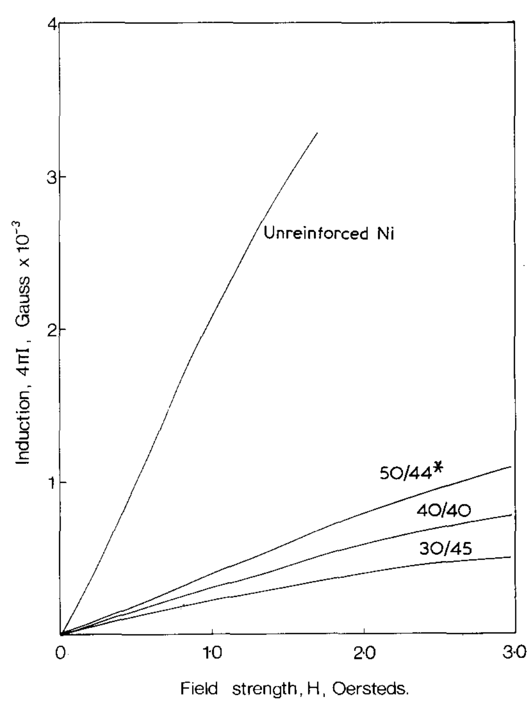

Figure 4 Magnetization curves of unreinforced nickel and nickel–tungsten composites containing various wire diameters (high V_f). *Nominal wire diameter (μm) and volume fraction, respectively.

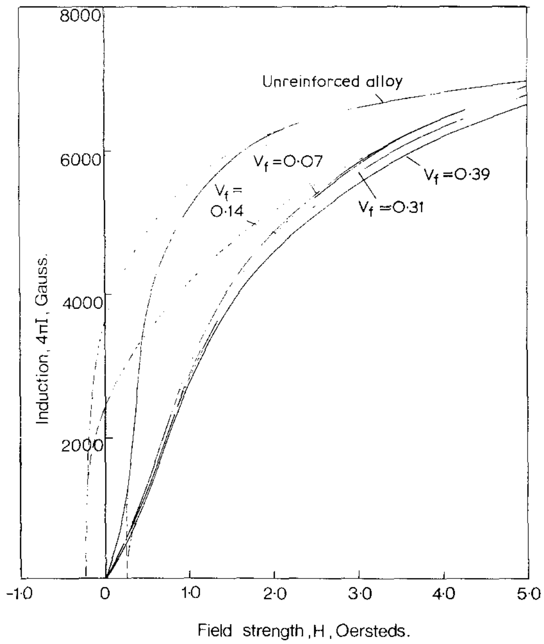

Figure 5 Magnetization curves of unreinforced 88% Ni–Fe alloys and composites containing various volume fractions of 20 μm diameter tungsten wire.

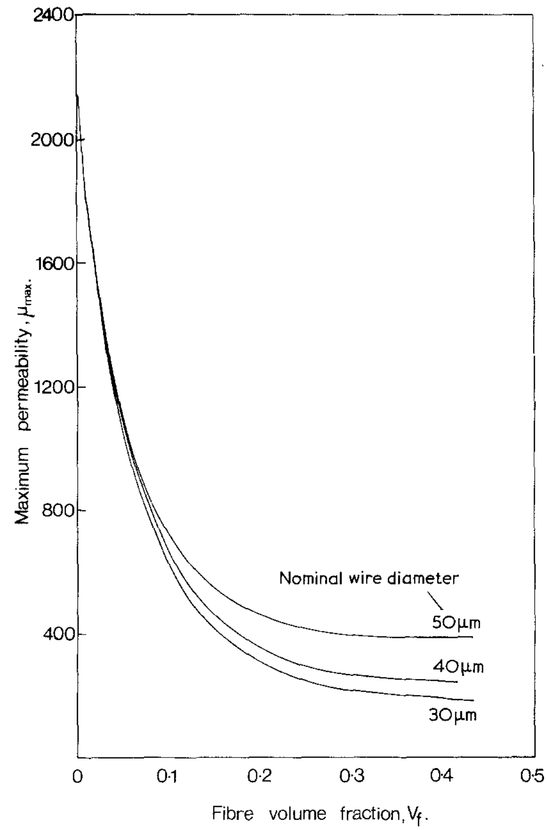

Figure 6 Relationship between the maximum permeability and the fibre volume fraction of Ni–W composites containing various wire diameters.

$V_f = 0.35$ is somewhat low but the values shown by the higher volume fraction composites was above 1000 MN m^{-2}, approaching the strengths shown by the soft magnetic alloys previously mentioned [1, 2].

The magnetic properties of composites with a matrix having a negative magnetostriction are shown in Fig. 3 to 5. These curves illustrate that the maximum permeability, μ_{max}, of all the composites was reduced relative to the unreinforced matrix, and appeared to decrease with increasing fibre content. In addition the nickel–tungsten composites showed that the reduction of μ_{max} was dependent on the fibre diameter as Fig. 6 illustrates. An increase in the coercive force was also noted in the nickel–tungsten composites and appeared to be dependent on both the fibre diameter and the volume fraction (Fig. 7).

The 88% Ni–Fe composites showed no significant variation in coercive force and, over-all, this value was much lower than that observed in the Ni–W composites. However, a decrease in the residual induction was noted as shown by the demagnetization curves, indicated by the broken lines, in Fig. 5.

Zero magnetostriction in Ni–Fe alloys occurs at a composition of approximately 82% Ni. Fig. 8 shows the magnetization curves for specimens having this composition. A fall in the maximum permeability, compared with the unreinforced matrix is again noted in these composites, although not to the degree observed in alloys with a higher nickel content. In particular the composite with $V_f = 0.11$ showed a relatively high value of μ_{max}.

Composites having a matrix composition of 50% and 60% Ni–Fe (positive magnetostriction) have magnetization curves shown in Figs. 9 and 10 respectively. It can be seen that at these compositions the value of μ_{max} increased initially with fibre content but tended to fall off at higher volume fractions although it still stayed above the values obtained for the unreinforced matrix. In addition the demagnetization curves in Figs. 9 and 10 illustrate that the higher permeability composites show a lower coercive force and a significantly higher residual magnetization.

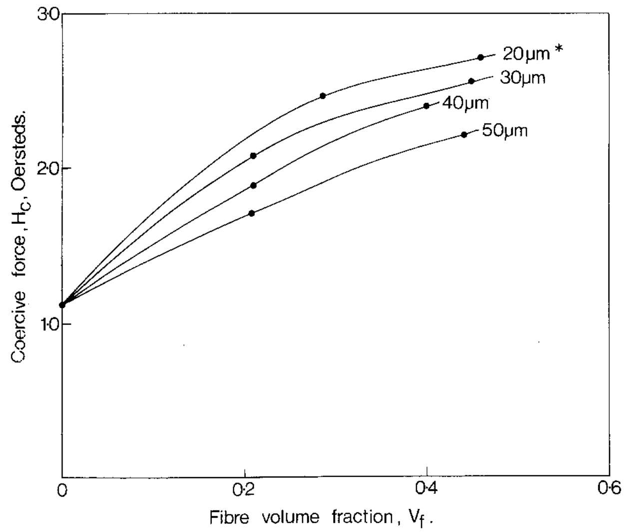

Figure 7 Relationship between the coercive force and the fibre volume fraction of Ni–W composites containing various wire diameters. *Nominal wire diameters.

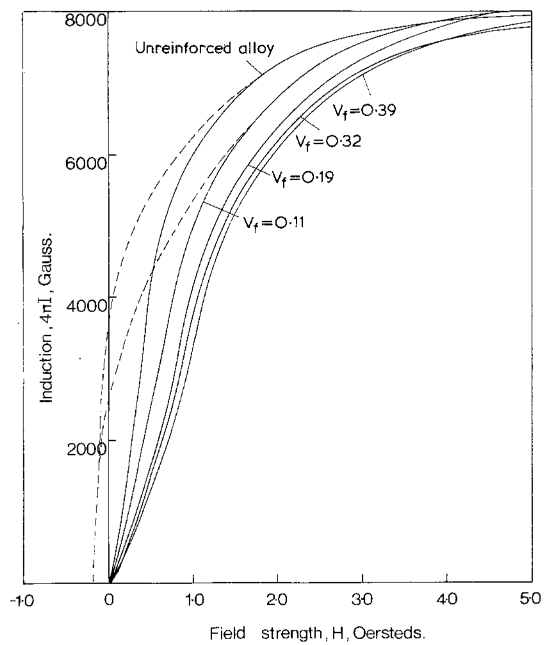

Figure 8 Magnetization curves of unreinforced 82% Ni–Fe alloy and composites containing various volume fraction of 20 μm diameter tungsten wire.

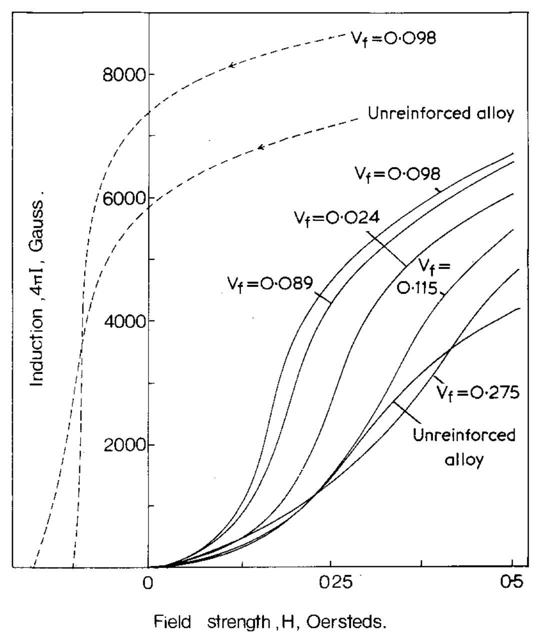

Figure 9 Magnetization curves of unreinforced 50% Ni–Fe alloy and composites containing various volume fractions of 20 μm diameter tungsten wire.

4. Theory

The presence of a stress in a ferromagnetic material which is below the elastic limit will generally have a negligible effect on the saturation magnetization but it may result in a significant increase or decrease in the magnetic permeability. In a plastically deformed material, the internal strains invariably result in a decrease in the permeability and, in addition to becoming magnetically harder, the material will usually become mechanically harder.

For this paper, it is convenient to classify ferromagnetic materials as having positive or negative magnetostriction. In materials having positive magnetostriction (e.g. Ni–Fe alloys < 82% Ni) the maximum permeability in the direction of the applied stress is increased by a tensile stress below the elastic limit and the material expands when magnetized. Conversely, in a material with a negative magnetostriction (e.g. Ni–Fe alloys > 82% Ni) the maximum permeability is decreased by tension (both above and below the elastic

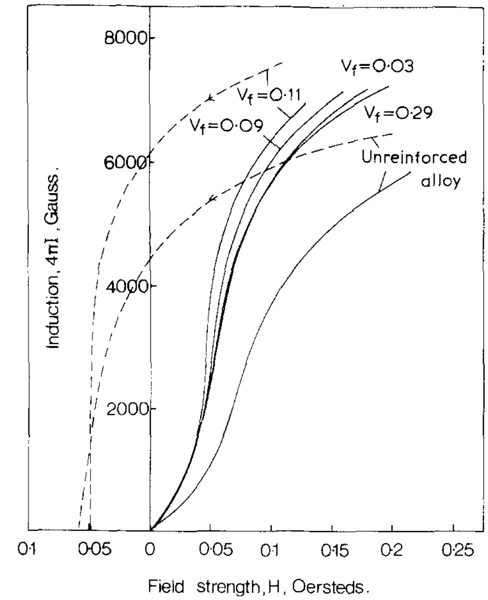

Figure 10 Magnetization curves of unreinforced 60% Ni–Fe alloy and composites containing various volume fractions of 20 μm diameter tungsten wire.

limit) and the material contracts when magnetized. The magnitude of the effect of stress on the magnetic properties is shown by the hysteresis loops in Fig. 11 for materials having high positive and negative values of magnetostriction. A compress-

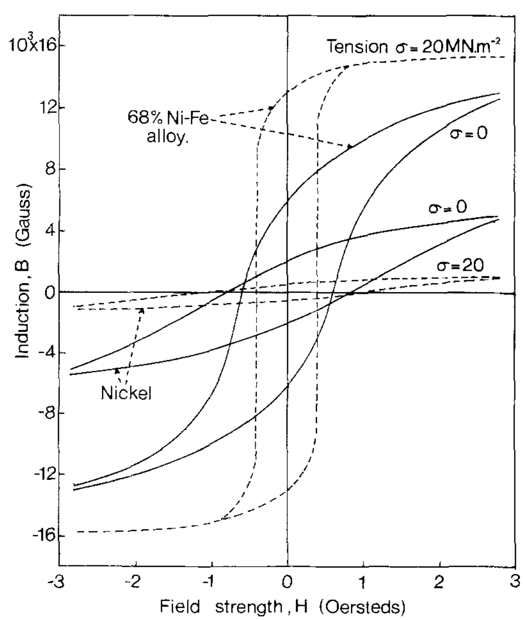

Figure 11 Magnetic hysteresis curves showing the effect of tension on materials having high positive and negative values of magnetostriction (after Bozorth [9]).

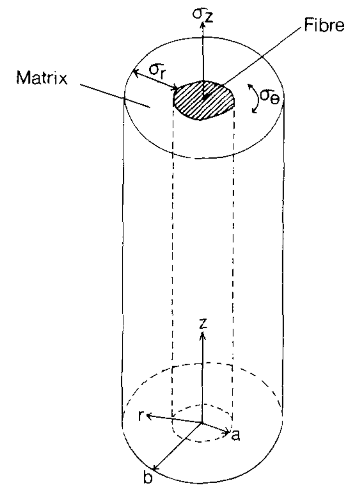

Figure 12 Two element model used to derive values for the axial and transverse elastic stresses formed in the matrix of a composite during cooling.

ive stress below the elastic limit will generally have the opposite effect to tension on the magnetic properties but the yield stress in compression will not necessarily be the same as in tension.

The magnitude of the effect produced by an elastic stress will depend on the value of magnetostriction which will vary with the alloy composition. The highest value of negative magnetostriction in the present materials occurs at 100% Ni, passes through zero at approximately 82% Ni and rises to high positive values near 50 and 20% Ni. The effect of a given elastic stress would, therefore, be expected to be greatest at the maximum values of magnetostriction and least for a composition near to 82% Ni.

5. Discussion

The fabrication of the majority of fibre-reinforced metals is carried out at an elevated temperature or, as in the present case, a heat-treatment is necessary to give optimum physical properties. As the fibres and matrix usually have different coefficients of expansion, stresses will be induced in both the fibres and the matrix on cooling from the fabrication or heat-treatment temperature. An approximate quantitative estimate of these stresses can be made by the use of equations originally developed by Poritsky [5] and more recently applied to metal matrix composites by Chawla and Metzger [6].

Consider the two element model shown in Fig. 12. Here, a is the radius of the fibre and b is the radius of the equivalent shell of matrix surrounding each fibre. A number of such elements make up the composite and each one is considered to act independently. On cooling the composites described in the present work, the high expansion coefficient of the matrix, K_m, means it will attempt to contract more than the fibres. However, it has been shown that during heat-treatment of similar composites a strong bond forms between the nickel and tungsten [7] and a similar strong bonding is to be expected in the Ni–Fe alloy matrix composites. Thus, the matrix will be prevented from contracting in the axial, z, direction and a tensile stress will arise. In addition to the axial stress, transverse stresses will also form in the matrix. The axial elastic stress σ_z, the transverse radial elastic stress σ_r and the tangential or hoop elastic stress σ_θ from [5] and [6] can be expressed as follows:

$$\sigma_z = -[E_m\delta/(1+\alpha+\alpha\beta H)]\left[2\nu\left(\frac{a}{b}\right)^2 + \frac{(1+\alpha+\alpha\beta H)}{1+\beta H}\right] \quad (2a)$$

$$\sigma_r = -[E_m\delta/(1+\alpha+\alpha\beta H)]\left[\left(\frac{a}{b}\right)^2 - \left(\frac{a}{r}\right)^2\right] \quad (2b)$$

$$\sigma_\theta = -[E_m\delta/(1+\alpha+\alpha\beta H)]\left[\left(\frac{a}{b}\right)^2 + \left(\frac{a}{r}\right)^2\right] \quad (2c)$$

where: $H = \dfrac{E_m}{E_f}$,

$\delta = (K_m - K_f)(T - T_o) < 0$,

$\alpha = \dfrac{a^2}{b^2}(1-2\nu)$,

$\beta = \left(\dfrac{b^2}{a^2} - 1\right)$,

E_m and E_f are the Young's moduli of the matrix and fibres respectively,

ν is Poisson's ratio and can be taken 0.3 for both the fibres and the matrix without serious error

T is the annealing temperature of the matrix which has been given as approximately 450° C for electrolytic nickel [8] and a similar value was indicated by the nickel–iron alloys in the present work.

Substituting appropriate values into Equations 2a to c indicates that transverse stresses fall off rapidly with increasing distance from the fibre–matrix interface, but near the interface high stresses do develop which are likely to be of sufficient magnitude to cause plastic flow, even at very low volume fractions. However, as the material surrounding the fibres represents only a small percentage of the total matrix at small volume fractions, the deformation arising there may be largely neglected.

The axial elastic stress is uniform throughout the matrix and increases with increasing volume fraction. As the volume fraction is increased, the situation becomes somewhat complicated as the high stresses near the fibre–matrix interface become increasingly significant, although in the present case, calculation shows that the axial stresses appear to be predominant midway between the fibres.

Fig. 13 illustrates the axial elastic stress, calculated from Equation 2a, expected to form in the matrix of the present composites during cooling taking:

$K_m = 12.8 \times 10^{-6}$ (for Ni and Ni–Fe alloys)

$K_m = 11.7 \times 10^{-6}$ (for Fe)

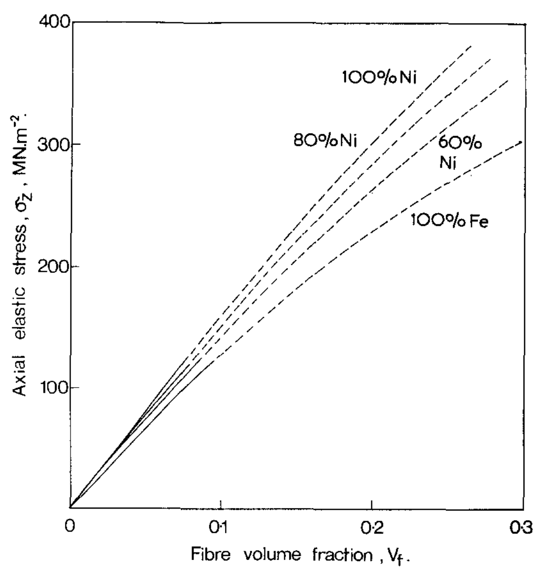

Figure 13 Relationship between the axial elastic stresses expected to form in the matrix and fibre volume fraction of composites containing matrices with various alloy compositions.

$$K_f = 4.9 \times 10^{-6} \text{ (for W wire)}$$

$$T - T_o = 430^\circ \text{C}$$

and using the measured values:

$$E_f = 346 \text{ GN m}^{-2}$$

$$E_{m(Ni)} = 210 \text{ GN m}^{-2}$$

$$E_{m(Fe)} = 120 \text{ GN m}^{-2}$$

In practice, however, elastic stresses above a certain level cannot form, as stress relaxation by plastic deformation will occur. Values for the yield stress of approximately 120 MN m^{-2} have been obtained for nickel fabricated by the present method [7] and alloys with a high nickel content appear to have a similar yield point [9].

In composites containing more than 82% Ni, the tensile elastic axial stress and plastic deformation, which become significant at high volume fractions, will both result in a reduction in the permeability as seen in Figs. 3 to 5.

Examination of Fig. 13 would suggest that stresses high enough to cause plastic deformation in these composites would form at volume fractions above approximately 10% assuming the matrix yields at the same stress as the unreinforced material. In practice some plastic deformation may have occurred below this volume fraction, near to the fibres, due to the high transverse stresses developed at the interface.

The dependence of the permeability on the fibre diameter shown in Fig. 6 may be explained in terms of the transverse stresses, as the uniform axial stress would be expected to be independent of a, the ratio a/b being constant for different fibre diameters at any given volume fraction. Although the actual magnitude of the transverse stresses near the fibre–matrix interface will not vary significantly with fibre diameter, as the fibres become smaller the volume of matrix affected by the transverse stresses will become larger and the matrix will be subjected to a higher mean stress. Thus, the more extensive elastic and plastic deformation occurring in the smaller diameter composites results in a lower value of μ_{max}.

Plots of μ_{max} against V_f for the 50% and 60% Ni–Fe matrix composites indicates a maximum value of permeability at $V_f \approx 0.1$ (Fig. 14), after which μ_{max} begins to fall but still stays above the values shown by the unreinforced matrices. It can be seen that the 60% Ni–Fe composites and unreinforced matrix show much higher values of

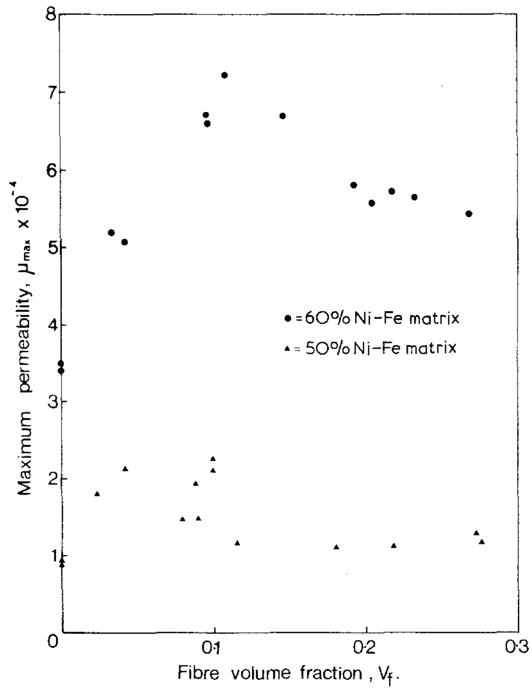

Figure 14 Relationship between the maximum permeability and fibre volume fraction for composites containing 20 μm diameter tungsten wires and matrices with a positive magnetostriction.

μ_{max} compared with the materials containing 50% Ni. This can be accounted for by the significant increase in the magnetic anisotropy constant on decreasing the nickel content from 60% to 50% [9] which results in the 50% Ni alloy being more difficult to magnetize. The initial increase in the magnetization with fibre content may be explained by considering the thermally induced stresses. Fig. 13 illustrates that the axial elastic stress increases with increasing volume fraction and since the stress is tensile an increase in the permeability will result. However, at $V_f \approx 0.1$, the yield stress of the matrix is reached and further increase in the fibre content will lead to plastic deformation of the matrix. Thus, the greatest observed values of maximum permeability are in the region of the calculated volume fraction at which the axial stress reaches the matrix yield point and can explain why a fall in μ_{max} is seen above this fibre content. In addition, the influence of the transverse stresses must not be neglected, as these become increasingly important as the volume fraction rises and the fibre separation becomes smaller. Thus, it is likely that a combination of axial and transverse stresses produce the plastic deformation leading to the observed reduction in the magnetic permeability above $V_f \approx 0.1$. Fig. 13 further illustrates that although the value of μ_{max} tends to decrease above $V_f \approx 0.1$ in both cases, higher values of μ_{max} are obtained at lower volume fractions and the fall-off is much more rapid in the 50% Ni–Fe composites. This is possibly due to the 50% Ni–Fe matrix having a greater strain sensitivity as the magnetostriction is higher than the 60% Ni–Fe matrix [9].

The reduction in the permeability in the 82% Ni matrix composites might not be expected for a matrix exhibiting zero magnetostriction. However, the actual composition is critical and the chemical analysis of this alloy gave a value of 82.4% Ni, which is a little above the point of zero magnetostriction. These composites appeared to be less stress sensitive than the 88% Ni–Fe alloys as the fall in permeability was not as marked. The composite with $V_f \approx 0.11$ especially showed a comparatively high value of permeability and Fig. 13 suggests that this is near the volume fraction at which the axial stresses are just beginning to cause plastic deformation. The higher volume fraction composites shown in Fig. 5 would be expected to be more heavily plastically deformed explaining their lower permeability.

6. Conclusions

It has been demonstrated that the filament winding–electroplating technique is a suitable means of fabricating nickel and nickel–iron alloy matrix–tungsten wire composites. With suitable modifications it is likely that such a technique can be extended to other systems in which the fibres have some electrical conductivity.

The results presented in this paper show that fibre-reinforcement is a viable means of increasing the tensile strength of soft magnetic materials. However, the thermal stresses arising during fabrication can have a considerable influence on the magnetic properties. In some cases this influence can be beneficial. Thus, a fibre with a low expansion coefficient reinforcing a matrix with a high expansion coefficient can induce a tensile axial stress in the matrix during cooling from the fabrication or heat-treatment temperature. If this stress is below the elastic limit of the ferromagnetic matrix an increase in permeability is observed in alloys with positive magnetostriction. However, at high volume fractions, higher stresses form in the matrix leading to plastic deformation and a reduction in the permeability.

By tailoring the properties of the matrix and the fibres it should be possible to produce both enhanced mechanical and magnetic properties in many soft ferromagnetic materials. In practice, however, most high strength fibres have relatively low expansion coefficients and will, therefore, be limited to use in alloys with a low or positive magnetostriction.

A closer match of the fibre and matrix expansion coefficients or a higher matrix yield stress would be preferable to those in the present materials with positive magnetostriction. This would allow a higher volume fraction of fibres to be incorporated into the matrix before extensive plastic deformation arises during the fabrication stage resulting in a fall-off in magnetic properties. High composite strength combined with enhanced magnetic properties would then be attainable.

Acknowledgement

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