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The effects of carbon on the magnetic properties of nanocrystalline Fe-based alloys

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We investigate the magnetic properties of nanocrystalline $Fe_{73.5}Cu_1M_3Si_{13.5}B_9$ (M = Nb or Mo) alloys when C is substituted for B up to 2 at%. It is found that the permeability and coercivity deteriorate with the content of C in the case of both M = Nb and Mo. The saturation magnetization also deteriorates as C is substituted for B in the case of M = Mo but it improves linearly with the C content in the case of M = Nb. This increase in the saturation magnetization of the Fe–Cu–Nb–Si–B alloy with C addition can provide an opportunity to overcome one of the main disadvantages, low magnetic flux density, of the alloy. In the latter part of the work we also investigate the magnetic properties of $Fe_{76.5-y}Cu_1Nb_y(Si_{0.5}B_{0.4}C_{0.1})_{22.5}$ $(0 \leq y \leq 3)$ alloys, particular emphasis being given to the role of Nb in the presence of C. It is found that C may help Nb to suppress the growth of α-Fe grains in the alloy.

1. Introduction

The magnetic properties of the ternary Fe–B–C amorphous alloys have previously been investigated by many workers and they have often been compared with those of the binary Fe–B amorphous alloys in order to clarify the role of C and its effect on the magnetic properties [1–5].

A systematic investigation has been carried out by Hatta *et al.* [1, 3, 4] for the alloy systems $Fe_{84}B_{16-x}C_x$ and $Fe_{86}B_{14-x}C_x$. The values of M_s measured at room temperature have been found to be above 16.5 kG, which is slightly higher than the peak value of M_s (15.9–16.2 kG) in the binary Fe–B amorphous alloys [6]. After annealing the alloys under suitable conditions, even higher values of M_s have been obtained, for example, values of M_s over 17.5 kG for an $Fe_{86}B_7C_7$ alloy [3]. The results obtained by Hatta *et al.* are not in accord with those predicted by the charge transfer model which is based on simple band theory [7]. The charge transfer model predicts that the value of M_s in the ground state should decrease as C is substituted for B in the binary Fe–B amorphous alloys, since C has more electrons available than B to fill the Fe d-band. Hatta *et al.* have offered some explanations, without solid evidence, for the increase in M_s: an increase in density with the substitution of C for B, a decrease in the slope of the magnetization versus temperature curves, and a structural change on annealing.

Later Luborsky *et al.* [5] have again investigated the same alloy systems of $Fe_{84}B_{16-x}C_x$ and $Fe_{86}B_{14-x}C_x$ studied by Hatta *et al.* and found that the values of M_s of as-quenched alloy ribbons are not greater than the peak value of M_s in the binary Fe–B alloys. Their results are not in accord with those of Hatta *et al.*, but in agreement with the charge transfer model.

In addition to M_s, the magnetic properties of permeability and coercivity have also been examined. Although in some cases there are controversies regarding the behaviour of the variation of M_s with C content, as already discussed above, no controversies are found so far, among many workers, on the variation of μ and H_c with the amount of C substitution in the Fe–B amorphous alloys [3, 5].

In this work we further investigate the effects of the substitution of C for B, but the alloys used are not the amorphous Fe–B alloys but the nanocrystalline Fe–Cu–M–Si–B alloys where M is either Nb or Mo [8]. The present work has been carried out in an effort to increase M_s of the nanocrystalline alloys, since one of the main disadvantages of the nanocrystalline Fe–Cu–M–Si–B alloys is the low magnetic flux density. It would also be interesting to see how C substitution affects the magnetic properties of the nanocrystalline alloys.

In the latter part of the work we also investigate the magnetic properties of $Fe_{76.5-y}Cu_1Nb_y(Si_{0.5}B_{0.4}C_{0.1})_{22.5}$ $(0 \leq y \leq 3)$ alloys, particular emphasis being given to the role of Nb in the presence of C. It is to be noted that Nb in the nanocrystalline Fe–Cu–Nb–Si–B alloy ribbons is known to suppress the growth of grains of α-Fe phase, which leads to the extremely small size of the grains (usually in the range of 10 to 15 nm) [9]. In the case of thin films of nanocrystalline Fe–C alloys [10], C is also considered to play a role similar to that of Nb in the nanocrystalline alloy ribbons. If C plays a role in suppressing the growth of α-Fe grains in the ribbons, then we are able to reduce or possibly to eliminate Nb. The presence of

Nb is known to decrease M_s due to the dilution effect and furthermore it renders the ribbon brittle, which may cause some problems in handling it.

2. Experimental procedure

Ribbons of $Fe_{73.5}Cu_1M_3Si_{13.5}B_{9-x}C_x$ (M = Nb or Mo, $0 \leq x \leq 2.0$) alloys were prepared by the single-roll melt-spinning technique. The ribbons were confirmed to be amorphous by X-ray diffraction. The thickness of ribbons was about 20 μm and the ribbon width was about 2 mm. The ribbons were wound on to toroidal cores with an inner diameter of 21 mm and they were annealed in a vacuum at temperatures ranging from 500 to 600 °C for 1 h.

The relative permeability (μ) of the annealed samples was measured by using an impedance analyser at a frequency of 1 kHz and an applied field of 5 mOe (multiply by 7.958×10^{-2} to convert to the SI unit of $\mathrm{A\,m^{-1}}$) and the d.c. magnetic properties, coercivity (H_c) and magnetic induction at an applied field of 10 Oe (B_{10}) determined using a hysteresis loop tracer. The saturation magnetostriction (λ_s) was measured by the three-terminal capacitance method. The microstructure was observed by transmission electron microscopy (TEM).

3. Results and discussion

In Fig. 1a–d the results for μ are shown as a function of annealing temperature for the compositions $x = 0.5$, 1.0, 1.5 and 2.0, respectively. From Fig. 1 it is observed that, at a given C content, the permeabilities of the alloys with M = Nb are significantly higher than those of the alloys containing Mo. The optimum annealing temperature is nearly the same, being independent of the C content and it is 550–570 °C in the case of M = Nb and 530–540 °C in the case of M = Mo. In order to see how the peak permeability varies with the C content more clearly, we have plotted curves of the peak values of μ versus x and they are shown in Fig. 2, where it is seen that μ decreases

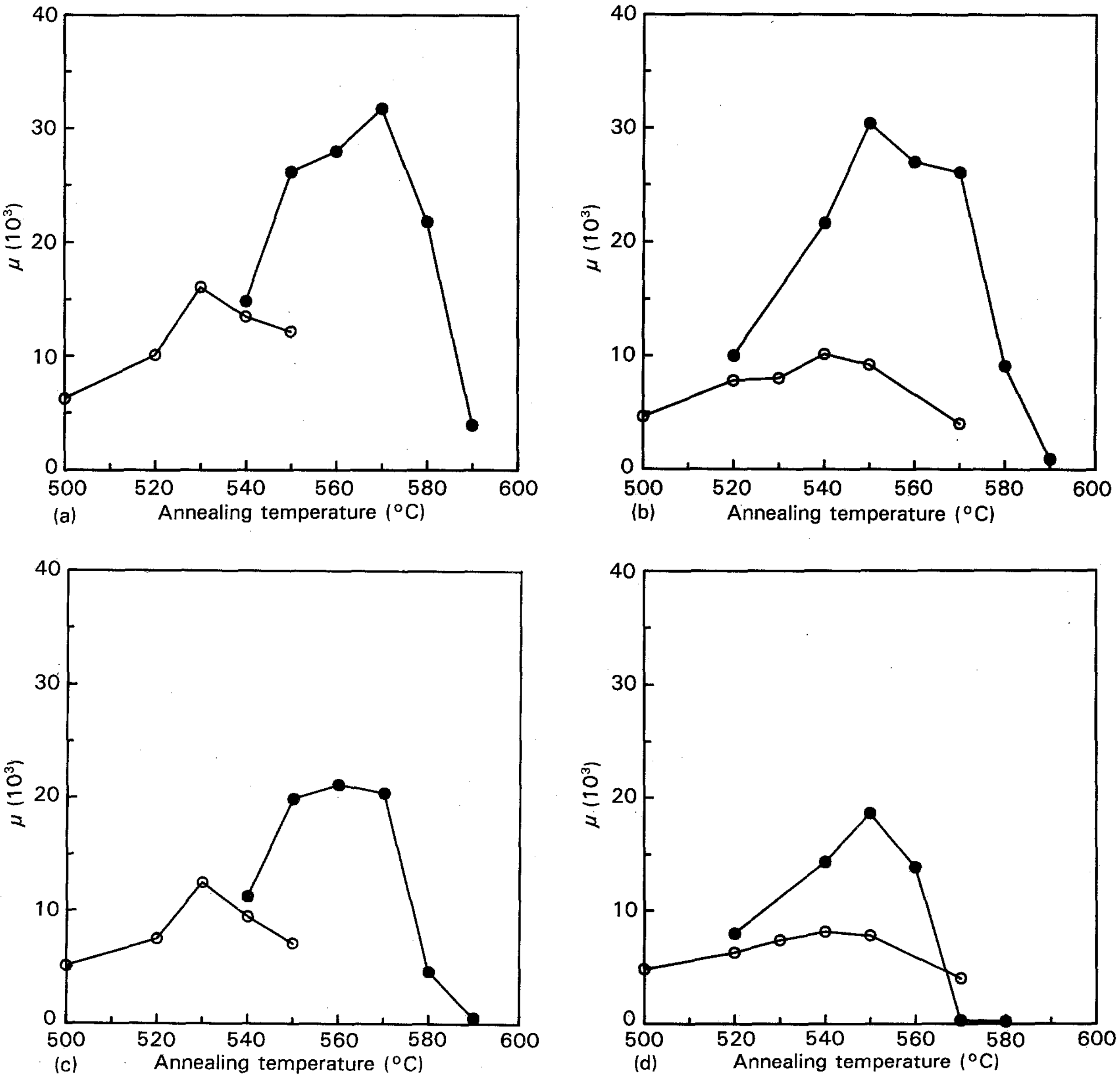

Figure 1 The value of μ as a function of annealing temperature for the alloys $Fe_{73.5}Cu_1M_3Si_{13.5}B_{9-x}C_x$: (●) M = Nb, (○) M = Mo. (a) $x = 0.5$, (b) $x = 1.0$, (c) $x = 1.5$, (d) $x = 2.0$.

linearly with x for all compositions investigated in the present work.

Figs 3 and 4 show the results for H_c and B_{10}, respectively, as a function of x. It can be seen from Fig. 3 that H_c increases linearly with x in the cases of both M = Nb and Mo. The value of H_c in the case of M = Nb is lower than that of H_c in the case of M = Mo by about 5 mOe.

Although μ and H_c vary quite similarly with x in the cases of both Nb and Mo as can be seen from Figs 2 and 3, the dependences of B_{10} on x are quite different, as can be observed from Fig. 4. In the case of M = Nb, B_{10} increases with x for the whole composition range, whilst in the case of M = Mo, B_{10} drops from ~ 12.5 kG to ~ 12 kG as C is substituted for B by 0.5 at % but, as the amount of C increases further, B_{10} increases with x. It is to be noted here that, since the initial permeability of the alloys is very large, the value of B_{10} should be close to that of M_s.

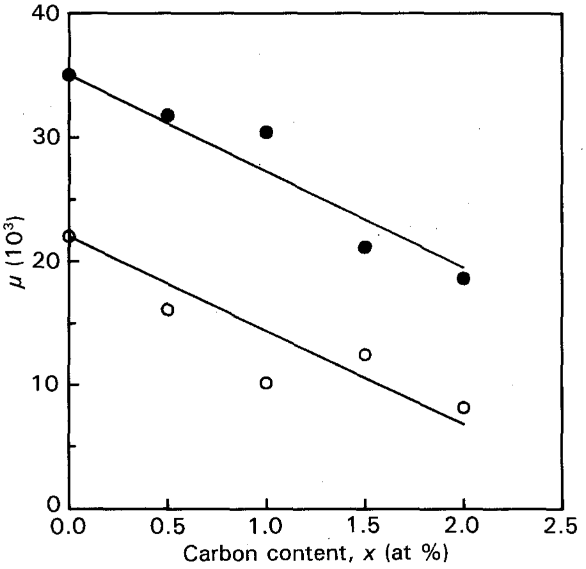

Figure 2 The value of μ as a function of x for $Fe_{73.5}Cu_1M_3Si_{13.5}B_{9.0-x}C_x$ alloys annealed at the optimum temperature: (●) M = Nb, (○) M = Mo.

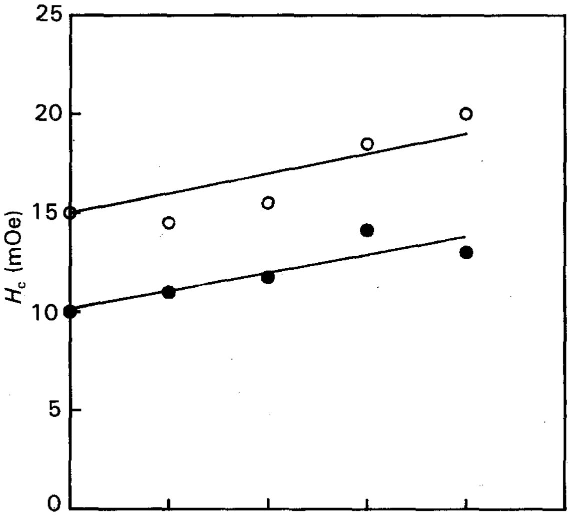

Figure 3 The value of H_c as a function of x for $Fe_{73.5}Cu_1M_3Si_{13.5}B_{9.0-x}C_x$ alloys annealed at the optimum temperature: (●) M = Nb, (○) M = Mo.

It is clear from both the present results for the nanocrystalline alloys and the previous ones for the amorphous alloys that the soft magnetic properties of μ and H_c deteriorate, as the amount of C substituted for B increases for the Fe-based alloys containing B. The behaviour for the variation of M_s with C content, however, is not simple, as is clearly evidenced from the results shown in Fig. 4. The increase in the saturation magnetization of the Fe–Cu–Nb–Si–B alloy with C addition can provide an opportunity to overcome one of the main disadvantages of the alloy, low magnetic flux density.

In order to seek the reason why the soft magnetic properties of μ and H_c deteriorate with increasing C content, we have observed the grains of α-Fe by TEM, since it is known that their size significantly affects the soft magnetic properties of nanocrystalline alloys [11, 12]. Some of the TEM results are shown in Fig. 5a and b for the samples with $x = 0$ and 2.0, respectively. As can be seen from Fig. 5, the grain size of the sample with the high C content appears to be larger than that of the sample with the low C content. Since in nanocrystalline alloys μ and H_c deteriorate as the grain size increases [11, 12], the present results for μ and H_c may be explained by the TEM results.

We have also measured λ_s of the nanocrystalline alloys in order to further explain the variation of the

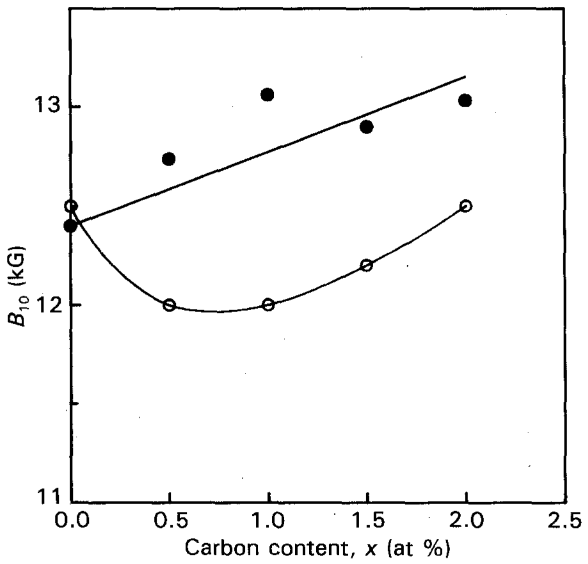

Figure 4 The value of B_{10} as a function of x for $Fe_{73.5}Cu_1M_3Si_{13.5}B_{9.0-x}C_x$ alloys annealed at the optimum temperature: (●) M = Nb, (○) M = Mo.

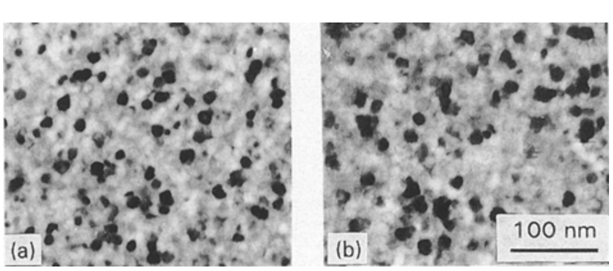

Figure 5 Microstructure of the nanocrystalline alloys observed by TEM: (a) $x = 0$, (b) $x = 2.0$.

soft magnetic properties with C content. It has been found, however, that the value of λ_s does not vary with the C content and it is within the range of $2–3 \times 10^{-6}$.

Recently a new nanocrystalline alloy system has been developed by Fujii *et al.* [13]. In the new alloy system whose compositions are $Fe_{78}P_{18-x}C_xCu_{0.5}Ge_3Si_{0.5}$ ($2 \leq x \leq 8$), it has been found that the soft magnetic parameters μ and H_c deteriorate as x (the amount of C substituted for P) increases. The value of M_s, however, has been found to improve with x. These results further demonstrate that the substitution of C is detrimental to the soft magnetic properties. They also provide a further example of increasing M_s by the substitution of C.

Let us now consider the role of Nb, in the presence of C, in forming the nanocrystalline grains. In Fig. 6 are shown the results for μ as a function of y (the Nb content) for the alloys $Fe_{76.5-y}Cu_1Nb_y(Si_{0.5}B_{0.4}C_{0.1})_{22.5}$. The value of μ is very small when no Nb is present in the alloy and increases slightly to about 6000 at $y = 1.0$. The value of μ then jumps to about 30 000 as the Nb content increases from 1 to 2 at %. The increase in μ, however, is very small as the Nb content increases further to 3 at %. This indicates that 2 at % Nb is enough to effectively suppress the growth of the α-Fe grains, which is to be compared with the minimum Nb content of 3 at % for the same purpose for the alloys the composition of which is similar to that used in the present work but with no added C [14]. The present result indicates that C may play a role in helping Nb to suppress the growth of the α-Fe grains.

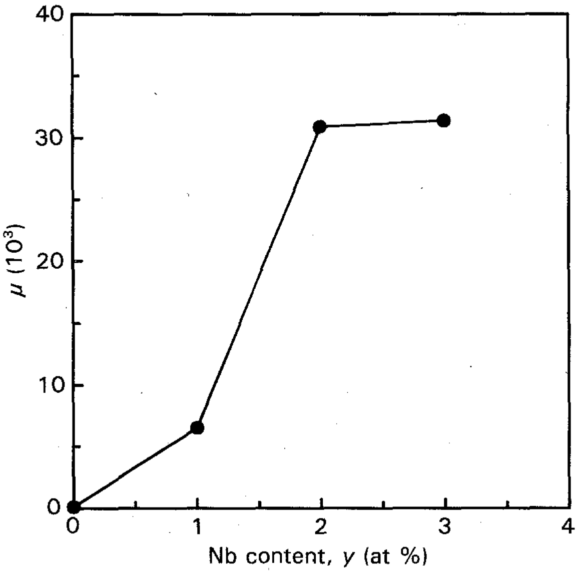

Figure 6 The value of μ as a function of Nb content y for the alloys $Fe_{76.5-y}Cu_1Nb_y(Si_{0.5}B_{0.4}C_{0.1})_{22.5}$.

4. Conclusions

We have investigated the magnetic properties of nanocrystalline $Fe_{73.5}Cu_1M_3Si_{13.5}B_9$ (M = Nb or Mo) alloys when C is substituted for B up to 2 at %. In the cases of both M = Nb and Mo, the soft magnetic properties of permeability and coercivity have been found to deteriorate with the amount of C substituted. The saturation magnetization also deteriorates as C is substituted for B in the case of M = Mo but it improves linearly with C content in the case of M = Nb. This improvement in the magnetization of the Fe–Cu–Nb–Si–B alloy with C addition is considered to be important, since one of the main disadvantages of the nanocrystalline alloy is low magnetic flux density.

We have also investigated the magnetic properties of $Fe_{76.5-y}Cu_1Nb_y(Si_{0.5}B_{0.4}C_{0.1})_{22.5}$ ($0 \leq y \leq 3$) alloys in order to examine the role of Nb in the presence of C. It has been found that 2 at % of Nb is enough to effectively suppress the growth of the α-Fe grains. This result may demonstrate that C plays a role of helping Nb to suppress the growth of the α-Fe grains.

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