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The effect of cobalt substitution on the annealing time of Nd–Fe–C ingot magnets

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Abstract

The effect of variable cobalt concentration and variable annealing times on the hard magnetic properties of Nd–Fe–C-type ingot magnets was studied. It was found that cobalt substitution can lead to a significant decrease in the annealing time needed to harden the ingots magnetically. A simple production route is described in which an isotropic ingot magnet is obtained after annealing for several hours with $(BH)_{max}=70$ kJ m^{-3}, $B_r=0.63$ T and ${}_JH_c=900$ kA m^{-1}.

1. Introduction

The tetragonal $Nd_2Fe_{14}C$ phase is formed at comparatively low temperatures contrary to the tetragonal $Nd_2Fe_{14}B$ phase which forms at a comparatively high temperature directly from the melt [1]. It was also found that the upper limit of the temperature stability range of the tetragonal phase in the systems $R_2Fe_{14}C_{1-x}B_x$ ($R \equiv$ Pr, Nd) increases very steeply with the boron content in the range $0<x<0.05$. This has been employed for finding annealing conditions for the generation of microstructures in ingot samples that give rise to substantial coercivities [2]. In general the coercivity proved to be dependent not only on the sample composition but also on the annealing temperature and the annealing time. Detailed investigations as to the effect of the annealing temperature on coercivity were made on ingots of the type $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$ [3]. When plotted as a function of the annealing temperature, the coercivity was found to give rise to a pronounced maximum around 975 °C. In the present investigation we have focused on the effect of annealing time, keeping the annealing temperature constant. We also investigated the effect of a variable cobalt concentration.

2. Experimental details

Various alloys of the composition $Nd_{16}Fe_{74-x}Co_xC_{9.5}B_{0.5}$ were prepared in an arc furnace from constituent elements which were of at least 99.9% purity. After arc melting, the alloys were wrapped in tantalum foil and sealed into evacuated quartz tubes. The annealing was performed for various annealing times t_A at 950 °C. After annealing, the quartz tubes containing the sample were cooled to room temperature in air. For annealing times below 1 h the samples were cooled to room temperature by breaking the quartz tube under water.

The samples were magnetically characterized by means of a vibrating-sample magnetometer. The phase composition was checked by powder X-ray diffraction.

3. Results

The coercivities measured on pieces of an ingot of the composition $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$ annealed for various annealing times at a temperature of 950 °C are shown in Fig. 1. The maximum applied flux density, without correction for demagnetization, was equal to 2 T in all cases. It can be seen from the figure that below $t_A=10$ h there is a very marked increase in the coercivity with increasing annealing time. By contrast, the coercivity remains almost constant for annealing times longer than about 10 h. As shown in more detail in the inset of Fig. 1 the coercivity is virtually zero in the as-cast samples, the main increase in H_c having already taken place within the first 30 mins. Ingots of somewhat different cobalt concentrations show basically

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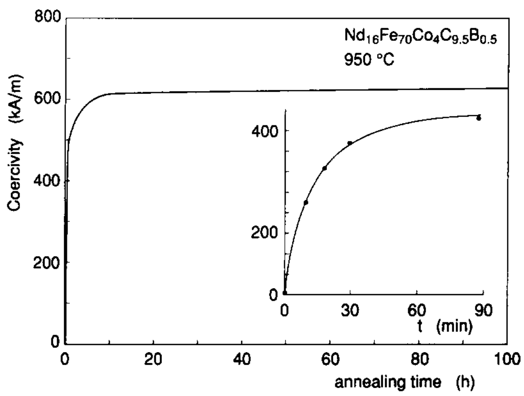

Fig. 1. Coercivity in ingot magnets as a function of the annealing time of an ingot of the composition $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$. The annealing temperature is 950 °C.

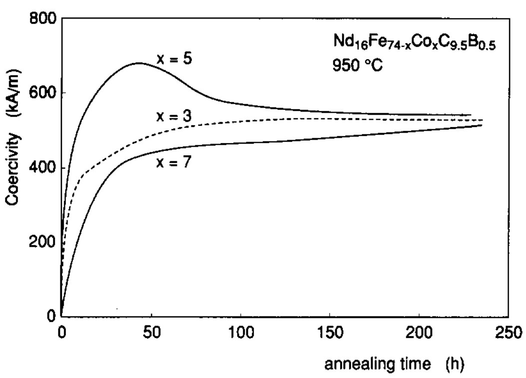

Fig. 2. Coercivity in ingot magnets as a function of the annealing time of ingots of the composition $Nd_{16}Fe_{74-x}Co_xC_{9.5}B_{0.5}$. The annealing temperature is 950 °C.

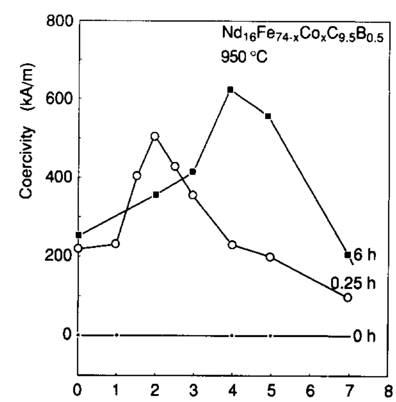

Fig. 3. Concentration dependence of the coercivity in ingot magnets of the type $Nd_{16}Fe_{74-x}Co_xC_{9.5}B_{0.5}$ after annealing at 950 °C for $t_A=6$, 0.25 and 0 h.

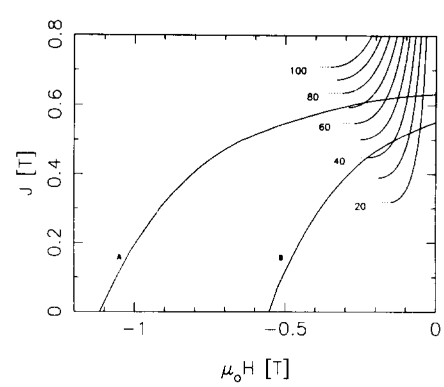

Fig. 4. Demagnetizing curves of an ingot magnet of the composition $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$ after annealing for 9 h at 950 °C: curve A, 20 °C; curve B, 100 °C.

the same behaviour, although the initial increase in H_c is less strong, as may be seen from Fig. 2. In the ingots with the composition $Nd_{16}Fe_{69}Co_5C_{9.5}B_{0.5}$ there is a shallow maximum for annealing times slightly below 50 h. All alloys shown in Fig. 2 tend towards about the same ultimate H_c value reached in fairly long annealing times, being slightly lower than that found in the alloy with $x=4$ (see Fig. 1).

X-ray diffraction performed on the various samples showed that they consist mainly of the 2:17 phase in the as-cast state. The 2:17 phase disappears upon annealing and can no longer be discerned in the X-ray diagrams of samples annealed longer than 5 h, say.

The concentration dependence of the coercivity is shown in Fig. 3 in which results obtained for the same annealing temperature and annealing time can be compared. The coercivity was found to be zero in all as-cast alloys, independent of the cobalt concentration. A fairly pronounced maximum was observed in the concentration dependence of H_c of alloys that had been annealed for a comparatively short time ($t_A=0.25$ h). The maximum is seen to shift towards a higher cobalt concentration in alloys annealed for much longer times ($t_A=6$ h).

The ingot of the composition $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$ annealed for 9 h at 950 °C was selected for a more detailed study of its hard magnetic properties. This alloy is located on the curve in Fig. 1 close to the point where the curve starts to become saturated and where annealing for longer times is not expected to lead to a further enhancement in the coercivity. The demagnetizing curve, obtained after magnetizing the ingot in a flux density of 8 T is shown in Fig. 4. It is seen that the coercivity equals about 900 kA m^{-1} (equivalent to a flux density of 1.12 T) and that the remanence is 0.63 T. In the plot in Fig. 4 we used a demagnetizing factor N of 1/3 and density of 7.3 g cm^{-3}. The maximum energy product of the ingot is $(BH)_{max}=70$ kJ m^{-3}. Included in Fig. 4 is the demagnetizing curve measured at 100 °C. There is a strong drop in the coercivity and a comparatively small reduction in remanence compared with the room-temperature data. The temperature dependence of H_c is shown in Fig. 5(a).

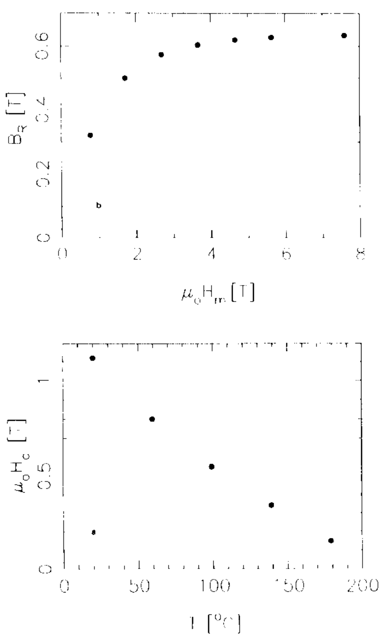

Fig. 5. (a) Temperature dependence of H_c of an $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$ ingot magnet obtained after annealing at 950 °C for 9 h. (b) Dependence of the room-temperature remanence B_R on the magnetizing field of an $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$ ingot after annealing for 9 h at 950 °C.

In order to obtain an impression of the field strength needed to magnetize the magnets at present obtained we employed magnetizing fields of different strengths. In general it appeared necessary to use flux densities stronger than 4 T to obtain a satisfactory magnetization of the ingots. This may be illustrated by means of Fig. 5(b) where the remanence is plotted *vs.* the magnetizing field.

4. Discussion

The results found in the course of the present investigation show that a microstructure with favourable hard magnetic properties has already developed in the initial stage of the heating treatment and that extended annealing does not lead to improvements in the coercivity. In fact, we showed that the magnetic properties of an ingot magnet of the composition $Nd_{16}Fe_{70}Co_4C_{9.5}B_{0.5}$ annealed for 9 h at 950 °C had virtually the same properties as when annealed for 72 h [3]. Almost equally good results were obtained for even shorter annealing times (6 h). This reduction in annealing time by almost an order of magnitude may become of considerable commerical importance when considering manufacture of these magnets by on-line production.

The shape of the curves describing the development of the coercivity as a function of the annealing time are quite interesting.

The initial dependence of H_c on annealing time t_A suggests that a relation of the type $H_c = \text{constant} \times t_A^{1/2}$ exists (t_A up to about 50 min in the inset in Fig. 1). Such a time dependence is frequently found for diffusion-controlled thickening of plate-shaped precipitates. In itself, such a growth mechanism for the particles of the tetragonal phase is not unlikely. If it were true, this would mean that H_c increases linearly with the thickening. Further analysis of H_c is unfortunately hampered by the fact that the hysteresis loops and H_c are due to the combined contributions of the magnetically hard tetragonal phase and the magnetically less hard rhombohedral phase ($Nd_2Fe_{17}C_x$ has an easy plane), while little is known about the magnetic coupling between small particles of these phases.

It follows from the results shown in Fig. 2 that the cobalt concentration is fairly important when comparatively short annealing times of the ingots are to be applied. The surprising result that may be derived from the present investigation is that a strong reduction in the annealing time needed to achieve magnetic hardening of the ingots is found in only a limited cobalt concentration range. For annealing times of several hours, H_c reaches an optimum value for x between 4 and 5 in $Nd_{16}Fe_{74-x}Co_xC_{9.5}B_{0.5}$.

References

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