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The correlation between the coercivity and the mechanical properties of sintered rare-earth–iron–boron magnets

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Abstract. Experimental proof of the correlation between the intrinsic coercivity, and the elastic and strength properties of sintered Pr–Fe–Co–B, Nd–Fe–Ti–B and Nd–RE–Fe–Co–M–B (RE ≡ Dy, Pr or Tb; M ≡ Al, Cr, Nb, (W, Re) or Zr) magnets has been examined. It is found that there exists strong correlation between the intrinsic coercivity ${}_iH_c$ and the mechanical properties; the increase in ${}_iH_c$ results in increases in the impact strength, ultimate tensile stress, bending strength and fracture toughness and in decreases in the elastic constant, Vickers hardness and tensile strength.

1. Introduction

Sintered RE–Fe–B-type magnets are of great technical importance because of their outstanding magnetic properties at room temperature and are widely used in areas requiring high performance and/or volume magnetization. However, further applications of RE–Fe–B magnets at elevated temperatures and in various environments are hampered by their major disadvantages such as the relatively low Curie temperature, large temperature coefficients of magnetic quantities and poor corrosion resistance due to the presence of RE-rich phases. The problems mentioned above can be solved particularly by firstly improving the intrinsic properties of the magnetic phase and secondly optimizing the microstructure. The typical microstructure of a sintered $Nd_{16}Fe_{76}B_8$ magnet (three-phase magnet), as shown in figure 1, consists generally of the following three phases:

(i) a hard magnetic $Nd_2Fe_{14}B$ phase (ϕ);
(ii) a non-magnetic $Nd_{1+\epsilon}Fe_4B_4$ phase (η);
(iii) a non-magnetic Nd-rich phase (n).

Great efforts have been made to improve the intrinsic properties of the ϕ-phase by the addition of further elements; the replacement of Nd by Dy and Tb causes an enormous increase in the coercivity [1–3], and the replacement of Fe by Co increases the Curie temperature [4] and thus improves the thermal stability of these magnets.

Another group of additives influences the phase relations and microstructure of the magnet. Recently, Fidler and Bernardi [5] on the basis of different effects on the microstructures have distinguished two types of alloying element:

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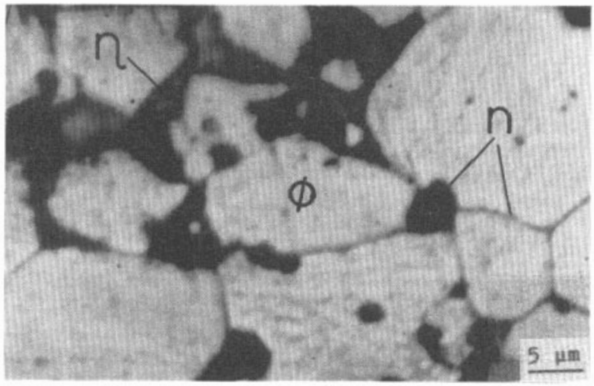

Figure 1. Optical micrograph of a sintered $Nd_{16}Fe_{76}B_8$ magnet

(i) type I: M1 ≡ Al, Ga, Cu, Sn, Zn, Ge and, as shown in our studies [6], Cr form in the intergranular region additional phases such as M1Nd, $M1_2Nd$ and $M1Fe_{14}Nd_6$;

(ii) type II: M2 ≡ Nb, V, Mo and Ti form precipitates in the intergranular region and in the ϕ grains such as $M2B_2$(Ti), $M2B_2Fe$(Nb, W) and $M2FeB_2$(V, Mo).

Both types of alloying elements increase the coercivity. The solubility of these elements in the ϕ-phase is limited; hence the intrinsic properties of the ϕ-phase change slightly and are mainly found in precipitates within ϕ grains, separating the ϕ grains which are believed to cause the coercivity increase.

The changes in microstructure due to the doped elements, in general, improve the corrosion resistance. Recently, we have summarized [7, 8] a series of our publications concerning this problem.

New applications of the magnets are connected not only with their magnetic and corrosion properties but also with their mechanical properties. One can easily believe that changes in the microstructure caused by alloying additions influence the mechanical properties. Therefore it is interesting to study the correlation between the intrinsic coercivity and the mechanical properties (i.e. elastic and strength properties) of the sintered RE–Fe–B-type magnets with different alloying elements. In this paper we report the new experimental results of such studies.

2. Experimental details

2.1. Sample preparation

The Pr–Fe–Co–B, Nd–Fe–Ti–B and Nd–RE–Fe–Co–M–B (RE ≡ Dy, Pr or Tb; M ≡ Al, Cr, Nb, (W, Re) or Zr) magnets used in this study were prepared by a conventional powder metallurgy technique. The material was produced by induction furnace melting of pure (99.9%) metals and Fe–20 wt% B master ally. The cast ingots were crushed to a coarse powder and then ground in the vibration ball mill into a fine powder (5 μm or less). The milled powders were compacted with a pressure of 500 MPa in a magnetic field of 1.6 MPa perpendicular to the pressing direction. Then the materials were sintered in a vacuum furnace at 1390 K for 2 h. Eventually, the samples were annealed at 1170 K for 1 h and at 870 K for 1 h in an argon atmosphere.

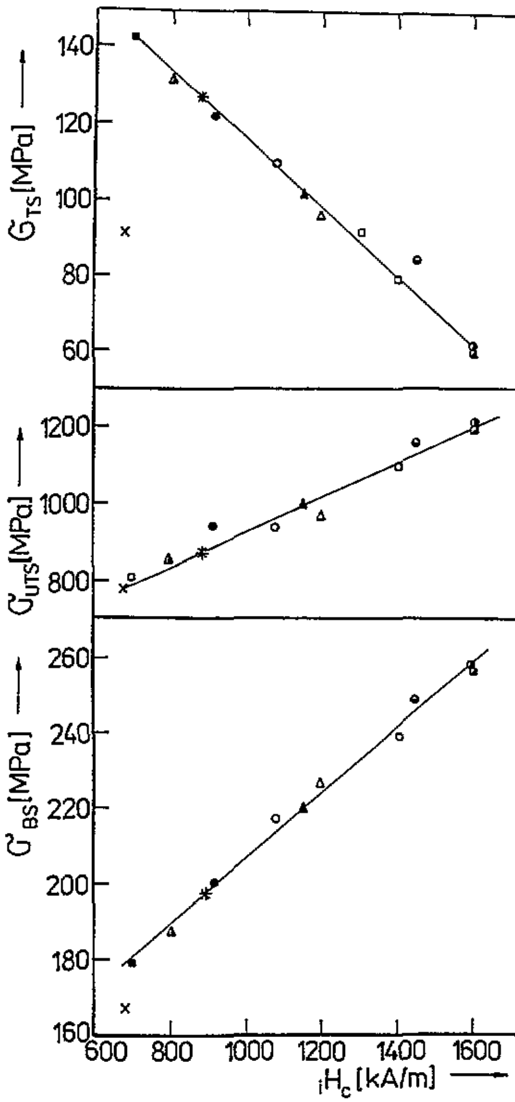

Figure 2. Relation between the intrinsic coercivity $_iH_c$ and the strength properties: σ_{TS} (tensile strength), σ_{UTS} (ultimate tensile stress) and σ_{BS} (bending strength).

2.2. *Experimental technique*

The magnetic measurements were performed at room temperature with a vibrating-sample magnetometer. The microstructure was analysed by means of optical microscopy and scanning electron microscopy. The magnetic domain structures were observed on the polished surface parallel to the easy-magnetization direction using the powder pattern method.

The densities of the tested magnets determined by comparing their weights in air and in toluene were the same in the limits of the error of the experimental method and equal to 7.45 ± 0.05 g cm^{-3}.

The mechanical properties of the magnets were characterized by measurements of

(i) the elastic properties, namely the elastic constant E and the impact strength α_K, and

(ii) the strength properties, namely the Vickers hardness H_V, the tensile strength σ_{TS}, the ultimate tensile strength σ_{UTS}, the bending strength σ_{BS}, and the fracture toughness K_{Ic}.

The samples for the measurements of E, α_K, σ_{BS} and K_{Ic} were cuboids of 5 mm × 5 mm × 35 mm in size and for measuring σ_{TS} and σ_{UTS} were in the shape of cylinders of diameter 10 mm and 20 mm long. The sample surfaces were finely ground and polished before investigations in order to remove the surface cracks and the residual stress generated during preparation. Young's modulus E was obtained by acoustic velocity measurements of the magnets. The impact strength α_K was tested according to *USSR Standard GOST-4454-60* [9]. The hardness was measured using a Vickers hardness tester. The test load was kept constant at 294 N (HV 30 scale). The measurements were carried out using three samples of each kind of magnet and at least ten hardness tests were made on each sample. Then the results were averaged. The strength properties such as σ_{TS}, σ_{UTS} and σ_{BS} were determined by testing ten specimens under identical conditions by means of an Instron machine. The tensile strength σ_{TS} was determined using the method of compression along the diameter of cylindrical samples [10, 11]. This method minimizes the influence of surface defects on the results of the measurements. The fracture toughness K_{Ic} was applied to evaluate the fracture strength of the magnets. K_{Ic} was estimated by the Vickers indentation method. The detailed procedure for this method was recently described by Otsuki *et al* [12]. Fractographic studies of selected samples were also conducted on a scanning electron microscope to characterize and compare the fracture surface.

3. Experimental results and discussion

Typical values of the magnetic properties for the magnets studied are listed in table 1.

Table 1. Magnetic properties of the sintered magnets Pr–Fe–Co–B, Nd–Fe–Ti–M–B and Nd–*RE–Fe–Co–M–B* (*RE* ≡ *Dy, Pr or Tb; M* ≡ *Al, Cr, Nb,* (*W, Re*) *or Zr*).

No.	Composition of magnets (at.%)	Magnetic properties B_r (T)	${}_iH_c$ (kA m^{-1})
1	$Pr_{16}Fe_{71}Co_5B_8$	1.16	680
2	$Nd_{16}Fe_{76}B_8$	1.23	800
3	$Nd_{16}Fe_{71}Ti_5B_8$	1.16	900
4	$Nd_{16}Fe_{71}Co_5B_8$	1.15	700
5	$Nd_{15}DyFe_{71}Co_5B_8$	1.18	915
6	$Nd_{14}DyFe_{68.5}Co_5AlCr_2B_{7.5}$	1.05	1080
7	$Nd_{14}Dy_{2.5}Fe_{61}Co_{12}Al_3B_{7.5}$	1.05	1200
8	$Nd_{14}Pr_2Fe_{67}Co_5Al_4B_8$	1.05	1150
9	$Nd_{14}Dy_{2.5}Fe_{65.5}Co_5Al_2Nb_3B_8$	1.06	1400
10	$Nd_{15}DyFe_{69.5}Co_5(W,Re)_{1.5}Zr_{0.5}B_8$	1.19	1450
11	$Nd_{14}Dy_2Fe_{67}Co_5AlCr_{1.5}Nb_2B_{7.5}$	1.05	1600
12	$Nd_{12}Tb_2Dy_2Fe_{71.5}CoB_{7.5}$	1.05	1600

The studies show that there is correlation between the mechanical properties and the intrinsic coercivity ${}_iH_c$ (figures 2–6). The significant observations are as follows.

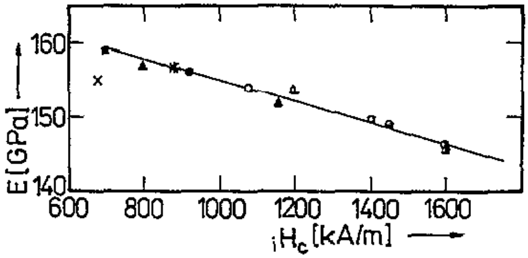

Figure 3. Elastic constant E versus intrinsic coercivity $_{i}H_{c}$.

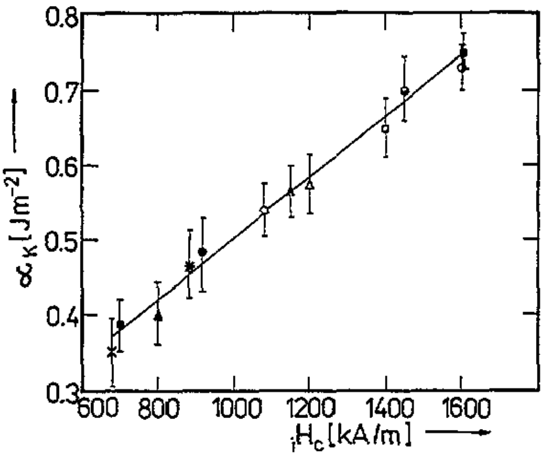

Figure 4. Impact strength α_K versus intrinsic coercivity $_{i}H_{c}$.

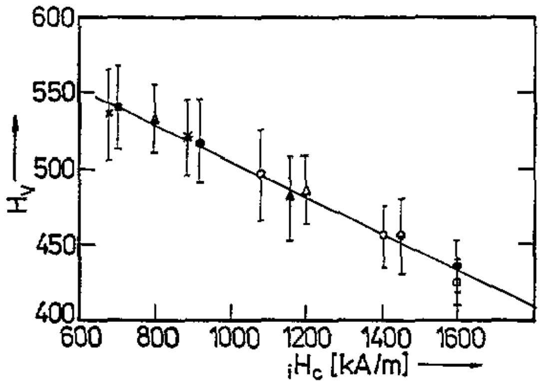

Figure 5. Vickers hardness H_V versus intrinsic coercivity $_{i}H_{c}$.

(i) With the increase in the intrinsic coercivity $_{i}H_{c}$ of sintered magnets, elastic properties such as the elastic constant E and the Vickers hardness H_V decrease and the impact strength

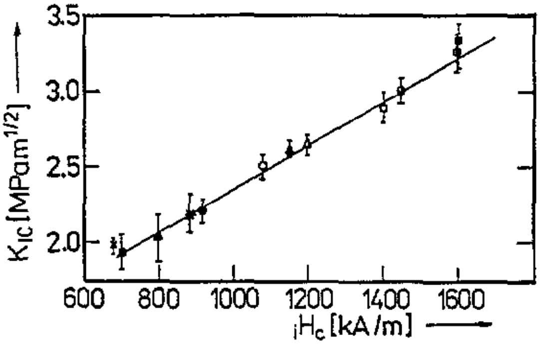

Figure 6. Fracture toughness K_{Ic} versus intrinsic coercivity $_iH_c$.

α_K increases.

(ii) An increase in the intrinsic coercivity $_iH_c$ of the sintered magnets results in an increase in the strength properties, except for the tensile strength σ_{TS} which decreases with the increase in $_iH_c$.

(iii) The fracture toughness K_{Ic} of the sintered magnets was equal to 1.85–3.25 MPa m$^{1/2}$ and increased with increase in the intrinsic coercivity. Further, K_{Ic} linearly decreases with increasing Vickers hardness (figure 7), which is in good agreement with the results of [12, 13].

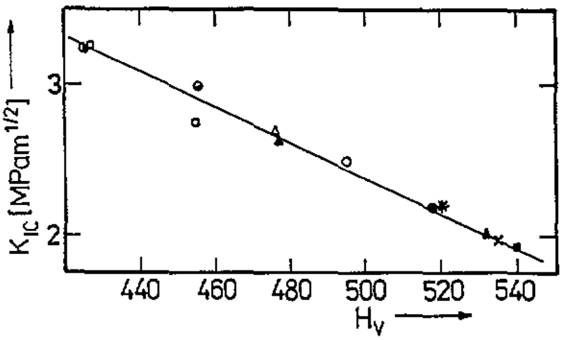

Figure 7. Dependence of the fracture toughness K_{Ic} on the Vickers hardness H_V.

The above observations are confirmed by the scanning electron fractographs which show that as-received samples exhibit a totally transgranular cleavage mode (figure 8). It can be seen that grains of the ϕ-phase are cleavage brittle and the Nd-rich phase is fractured ductile. Thus, as our observations show, the coarse grains of the ϕ-phase near the voids are the fracture-initiating defects. The inclusions play a less important role compared with the previous factors.

In nucleation-type magnets such as sintered RE–Fe–B the intrinsic coercivity $_iH_c$ can be described [14] by

$$_iH_c = 2K_1/\mu_0 M_s \alpha_K \alpha_\psi - N_{eff} M_s \quad (1)$$

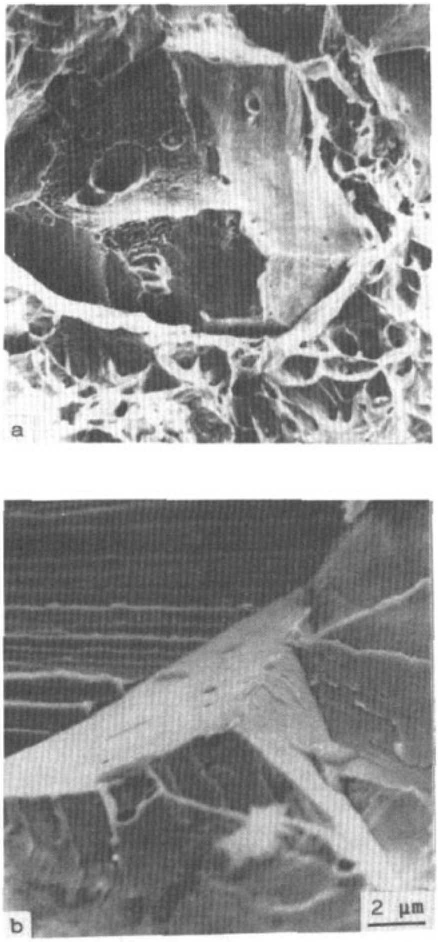

Figure 8. Scanning electron micrographs of fracture on the two different types of grain boundary occurring in a sintered $Nd_{14}DyFe_{69.5}Co_5AlCr_2B_{7.5}$ magnet: (a) Nd-rich layer; (b) no Nd-rich layer phase between the grains.

where $2K_1/\mu_0 M_s$ is the maximum nucleation field, M_s is the saturation magnetization, α_K describes the effect of the inhomogeneity of the magnetocrystalline anisotropy at the grain surface on the nucleation field, α_ψ is the magnetic decoupling and misalignment of grains and N_{eff} is the internal stray field.

Typical domain structures of the thermally demagnetized $Nd_{15}DyFe_{71}Co_5B_8$ sintered magnet on the surface parallel to the easy direction (figure 9) also show the morphology, grain size and multiple-domain structure in ϕ grains because the grains of this phase exceed the theoretical single-domain particle diameter of 0.3 μm [15]. In such oversized grains the total free energy of a multiple-domain structure is lower than that of a single-domain structure. Therefore the average grain size of the sintered $Nd_{16}Fe_{76}B_8$ magnet is larger and the grains are less magnetically isolated than the magnets containing additives (figure 10). A small grain size is desirable to attain the high coercivity [16, 17] because firstly the local demagnetizing field at the edges increases with increase in the grain size

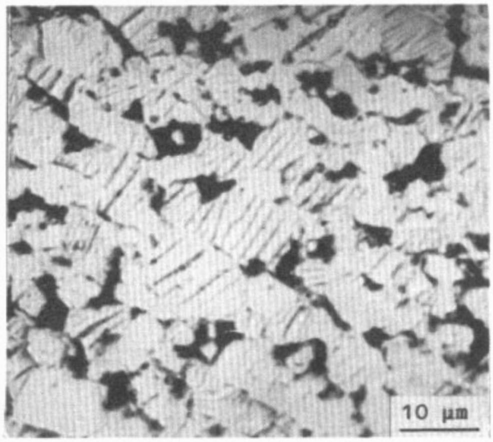

Figure 9. Typical magnetic domain structures of the thermally demagnetized $Nd_{16}Fe_{76}B_8$ magnet.

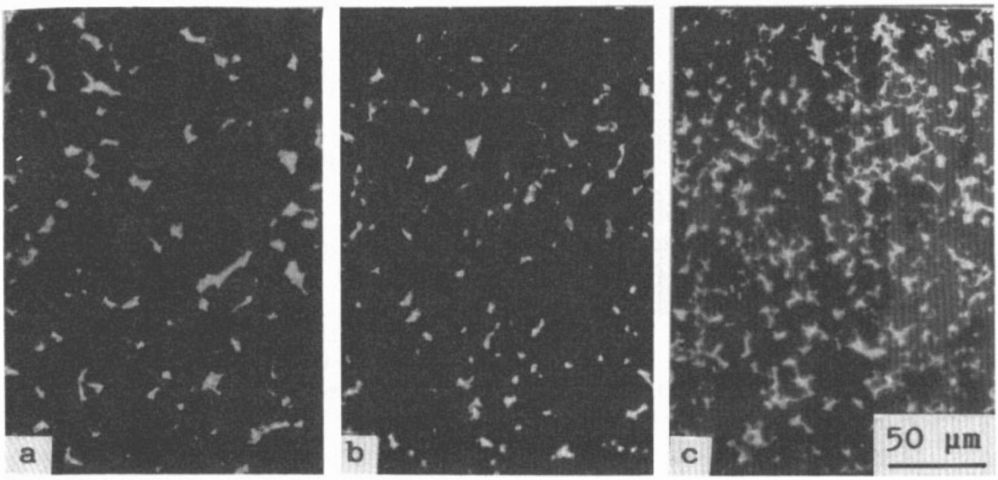

Figure 10. Backscattered electron image of sintered magnets: (a) $Nd_{16}Fe_{76}B_8$; (b) $Nd_{14}DyFe_{69.5}Co_5AlCr_2B_{7.5}$; (c) $Nd_{14}Dy_{2.5}Al_2Nb_3B_8$.

and secondly in larger grains the probability of inhomogeneities at which demagnetization can be nucleated is higher [18]. In turn, if some neighbouring grains are not completely magnetically isolated by the non-magnetic phase, reversal of the magnetization in one of these grains will lead to a cascade of demagnetization process in neigbouring grains [18, 19]. This type of inhomogeneity leads to deterioration of the coercivity; as the number of grains exchange coupled to one another increases, so the coercivity decreases [19]. Moreover, the domain structure reveals that the misoriented grains produce a local demagnetizing field, which also causes ${}_iH_c$ to decrease. Recently Schrefl and Fidler [20] and Schrefl *et al* [21] have shown that non-magnetic intergranular phases reduce the effect of misoriented grains.

A more important effect on the intrinsic coercivity is that the non-magnetic phases in a matrix of ϕ-phase and pores produce demagnetization field. The main benefit of all the additives (Dy, Tb, Pr and Co too) is their strong modification of the intergranular phase that can magnetically isolate grains of the ϕ-phase. Another important effect is the growth inhibition of the ϕ grains during sintering. These effects are distinctly shown in figure 10 in which as an example the scanning electron micrographs for the magnets with different amounts of alloying additions are depicted. One can assume that the same effects (i.e. small homogeneous grains of the ϕ-phase isolated from one another and elimination of the porosity) cause an improvement in the mechanical properties of the magnets investigated. It may be considered to be the reason for the observed correlation between the intrinsic coercivity and the mechanical properties of sintered Nd–Fe–B-type magnets.

So we believe that, to attain better understanding of the mechanism leading to the strong correlation between the intrinsic coercivity and the mechanical properties in sintered rare-earth magnets, not only chemical analysis, x-ray analysis and studies of the magnetic and mechanical properties but also metallography investigations are necessary.

4. Summary

From the experimental results and the analysis of the intrinsic coercivity, microstrucure, domain structure and mechanical properties it may be concluded that strong correlation exists between the intrinsic coercivity and elastic and strength properties. Both the intrinsic coercivity and the mechanical properties are influenced by the same microstructure effects, i.e. fine homogeneous grains of the ϕ-phase isolated from one another by the non-magnetic phase and the elimination of porosity.

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