Magnetic Properties of R–Nd–Fe–B (R = Tb, Dy) and Nd–Fe–Co–B Alloys in the Range –80 to 250°C

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Abstract—The remanence, coercivity, and maximum energy product of R–Nd–Fe–B (R = Tb, Dy) and Nd–Fe–Co–B alloys were studied in the range –80 to 250°C. Doping of Nd–Fe–B alloys with Tb and Dy was shown to increase coercivity and, accordingly, the temperature coefficient of magnetic induction. The introduction of 7–12 wt % Co raises the Curie temperature of the alloys, thereby improving their thermal stability.

INTRODUCTION

One important requirement for hard-magnetic materials and magnets fabricated from such materials is thermal stability—insensitivity of the magnetic flux to thermal influences.

As is well known, in all types of hard-magnetic materials, changes in magnetic flux and other magnetic characteristics are due to irreversible and reversible losses. The origins and mechanisms of these changes have been studied extensively [1]. After magnetization, permanent magnets are in a thermodynamically non-equilibrium state. In view of this, any changes in external conditions, in particular temperature, lead to a transition to another, more stable state, typically accompanied by changes in magnetic properties [2-4].

EXPERIMENTAL

The temperature coefficients (TCs) of reversible changes in magnetic properties were determined on \approx 3-mm-diameter spherical samples (\approx 100 mg) cut from sintered magnets and aged at 120°C for 2 h. In addition, we used rectangular magnets measuring 55 × 19.6 × 13.8 mm, also aged at 120°C.

Measurements on the spherical samples were made with a vibrating-sample magnetometer intended for testing small amounts of hard-magnetic materials and equipped with a thermal chamber for temperatures from -170 to 350° C.¹ Cooling to below 0°C was performed with liquid nitrogen. To prevent sample oxidation, measurements at elevated temperatures were made in an inert atmosphere. The demagnetization curves of the samples studied were measured at different temperatures, and the results were used to determine specific magnetization.

Next, the specific magnetization was converted, with consideration for the demagnetizing factor, into

magnetization and induction. In this way, we obtained the demagnetization curve for a particular alloy.

The coefficient of reversible changes in induction was determined from the demagnetization curves recorded at different temperatures. The demagnetization curves obtained at room and elevated temperatures can be used to evaluate the TCs of

(1) remanence,

$$\Gamma C_{B_{\rm r}} = \frac{B_{\rm r0} - B_{\rm rt}}{B_{\rm r0}(t_0 - t)} 100\%/^{\circ}{\rm C};$$

(2) coercivity,

$$TC_{H_c} = \frac{H_{c0} - H_{ct}}{H_{c0}(t_0 - t)} 100\%/^{\circ}C;$$

and energy product,

$$TC_{(BH)^{max}} = \frac{(BH)_0^{max} - (BH)_t^{max}}{(BH)_0^{max}(t_0 - t)} 100\%/^{\circ}C,$$

where subscripts 0 and t label room- and elevated-temperature values, respectively.

The TCs can also be determined at other points on the demagnetization curve, e.g., at the working point of a particular magnet.

The purpose of this work was to fabricate thermally stable magnets with irreversible changes in magnetic induction within 10% at $(BH)^{\text{max}} = 220-270 \text{ kJ/m}^3$ and within 15% at $(BH)^{\text{max}} \ge 270 \text{ kJ/m}^3$. The stabilization temperature was 120°C.

RESULTS AND DISCUSSION

For the magnets with $(BH)^{max} \le 270 \text{ kJ/m}^3$, the irreversible decrease in induction after stabilization at 120°C for 2 h was within 10% of the room-temperature induction. The results for eight Tb₃Nd₄Fe₇₅B₈ samples are presented in Table 1. The insignificant decrease in induction is attributable to the presence of Tb [5–7].

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However, we failed to fabricate magnets with $(BH)^{\text{max}} \ge 270 \text{ kJ/m}^3$ from Tb-doped alloys. Magnets with high energy products were prepared from Nd-Fe-B alloys, which had low Curie temperatures, $T_C \le 310^{\circ}$ C. In view of this, stabilization of these magnets at 120°C notably reduced the magnetic induction, down to 40% of the initial value.

The spherical samples of different alloys, cut from sectoral magnets, were studied in the temperature range -60 to 120° C. TC_B was found to significantly depend on temperature: in the range -60 to 20° C, TC_B was always substantially lower than between 20 and 120° C.

For the Tb-doped alloys with the irreversible decrease in magnetic induction within 10%, TC_B ranges from -0.078 to -0.100%/°C (Table 2). One way of preparing materials with a near-zero TC_B is by doping with rare earths (light or heavy).

In addition, we examined the effect of alloy composition on the decrease in magnetization as a function of the applied demagnetizing field. In Nd–Fe–B alloys, the reduction in magnetization in a demagnetizing field of 640 kA/m attains 40–50% of the initial value, because the intrinsic coercivity of these alloys, $_iH_c$, is comparable with the demagnetizing field. In the magnets with higher energy products, $_iH_c$ ranges from 640 to 870 kA/m. Doping with heavy rare earths, such as Tb and Dy, is known to raise $_iH_c$, occasionally to above 2000 kA/m [8–10], simultaneously reducing the magnetization of the magnets fabricated from such alloys.

The magnets with lower energy products have $_iH_c > 1000$ kA/m and are, accordingly, less subject to the effect of demagnetizing fields. In particular, the decrease of magnetization in a demagnetizing field of 640 kA/m is within 15% of the initial value.

Thus, our results demonstrate that $Tb_3Nd_{14}Fe_{75}B_8$ magnets feature a higher thermal stability and are less subject to the effect of demagnetizing fields.

We also studied the magnetic properties of $Tb_3Nd_{14}Fe_{75}B_8$ magnets as a function of temperature. Measurements were made on plates of dimensions $12 \times 12 \times 5$ mm.

As illustrated in the figure, the irreversible decrease in magnetic induction becomes more pronounced with increasing temperature. Our results demonstrate that higher coercivity materials are less susceptible to thermal influences.

Moreover, the magnetic induction depends strongly on the heating duration, decreasing sharply during the first hour and then leveling off. Additional studies of $Tb_5Dy_3Nd_9Fe_{75}B_8$ show that heating sharply reduces TC_B but $(BH)_{max}$ remains no higher than 100 kJ/m³.



Specific magnetization as a function of temperature for $(1) Nd_{17}Fe_{75}B_8$ and $(2) Tb_3Nd_{14}Fe_{75}B_8$.

In view of this, it is believed that doping of Nd–Fe– Co–B alloys with Tb or Dy increases coercivity and, accordingly, TC.

Table 1. Effect of thermal stabilization on the characteristics of $Tb_3Nd_{14}Fe_{75}B_8$ magnets

	B _r , T		$(BH)^{\max}$, kJ/m ³		л, %
Sam- ple no.	before sta- bilization	after stabi- lization	before sta- bilization	after stabi- lization	Decrease in magnetization
1	1.06	0.98	225	192	7.5
2	1.05	0.94	221	176	9.6
3	1.07	0.99	229	196	7.5
4	1.06	0.96	225	184	9.4
5	1.00	0.99	200	196	1.0
6	1.08	1.05	234	220	2.8
7	1.08	1.01	234	204	6.5
8	1.07	1.01	229	204	5.6

Table 2. Values of TC_B in different temperature ranges

Allow	TC _B , %/°C			
Anoy	60+120°C	60+20°C	–20…+120°C	
Tb ₃ Nd ₁₄ Fe ₇₅ B ₈	-0.071	-0.060	-0.084	
Nd ₁₅ Fe ₆₇ Co ₁₀ B ₈	-0.089	-0.082	-0.100	
Nd ₁₅ Fe ₆₅ Co ₁₂ B ₈	-0.078	-0.074	-0.086	
Tb _{3.5} Nd _{13.5} Fe ₇₅ B ₈	-0.069	-0.052	-0.087	
Nd ₁₇ Fe ₇₅ B ₈	-0.070	-0.065	-0.078	

CONCLUSION

Our results for the Tb₃Nd₁₄Fe₇₅B₈ alloy demonstrate that the irreversible decrease in the magnetic induction of magnets with $_iH_c \le 800$ kA/m is $\approx 55\%$ in the range 20–40°C and $\approx 80\%$ in the range 20–180°C. The irreversible decrease in the magnetic induction of magnets with $_iH_c = 1100-2000$ kA/m, fabricated from Tb- and Dy-doped Nd–Fe–Co–B alloys, is within 5% in the range 20 to 250°C and within 7% between 290 and 457°C.

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