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Effect of Copper on the Temperature Dependence of the Magnetization of Sintered (Pr, Dy)–(Fe, Co)–B Materials

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Abstract—The effect of copper on the temperature dependence of the magnetization of sintered $(Pr_{0.52}Dy_{0.48})_{13}(Fe_{0.65}Co_{0.35})_{bal}Cu_xB_7$ materials with $x = 0$–6.3 at % has been studied in the temperature range from –60 to +80°C. Copper is made a part of composition at the stage of powder processing. It was found that the temperature dependences of magnetization for compositions with $x = 0$, 1.3, 2.1, and 6.3 at % are characterized by a maximum. The maximum deviation of the maximum magnetization from the magnetization at –60°C does not exceed 5.5%.

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INTRODUCTION

The modern magnetic characteristics, in particular, the temperature coefficient of induction (TCI) cannot be the basis of technical requirements for hard magnetic materials for navigation instruments. TCI characterizes a hard magnetic material only in the case of a linear temperature dependence of magnetization J. However, even for $R_2Fe_{14}B$ intermetallics (R = rare-earth metal), the magnetization can have a maximum in the temperature range from –60 to +80°C [1]; i.e., the magnetization varies nonlinearly. Therefore, the temperature dependence of magnetization J should be studied, and a hard magnetic material can be characterized on the basis of this analysis. It was shown in our previous studies [2, 3] that the alloying of sintered hard magnetic materials of the R–Fe–B system with cobalt leads to the fact that an R–$(Fe_{1-y}Co_y)$–B material with $y > 0.2$ atomic fractions corresponds to the R–Co–B phase diagram.

In the present study, we estimate the effect of copper on the temperature dependence of the magnetization of R–$(Fe_{1-y}Co_y)$–B materials with R = Pr and Dy and $y > 0.2$ atomic fractions.

EXPERIMENTAL

We studied $(Pr_{0.52}Dy_{0.48})_{13}(Fe_{0.65}Co_{0.35})_{bal}Cu_xB_{6-7}$ materials (composition is given in at %). The preparation technology of sintered magnets is described in [2, 4]. Copper in the form of a PMS-1 powder (State Standard GOST 4960–75) was made a part of the composition at the stage of fine milling. The magnetic characteristics were measured in the temperature range from –60 to +80°C using a PPMS-9 instrument and spherical samples. A magnetic field was applied parallel to the easy magnetization axis of samples.

RESULTS

Figures 1–4 show the temperature dependences of the magnetization of the hard magnetic materials whose compositions are given in the table. The temperature dependences of all samples are characterized by a maximum observed in the temperature range from –60 to +80°C. The temperature dependence is char-

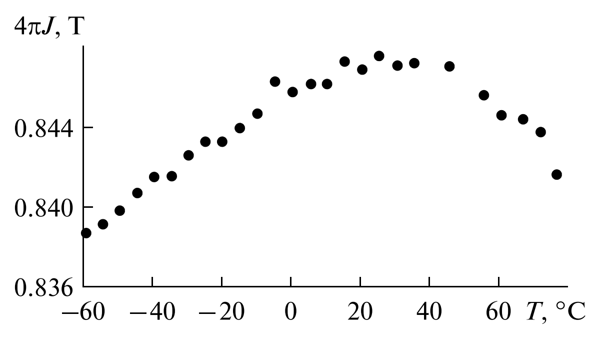

Fig. 1. Temperature dependence of the magnetization of $(Pr_{0.52}Dy_{0.48})_{13.6}(Fe_{0.65}Co_{0.35})_{79.5}B_{6.9}$ samples measured in a field $H = 320$ kA/m; $\Delta_{-60°C} = 1.1\%$, $\Delta_{+80°C} = 0.71\%$, $T_{max} = 30°C$.

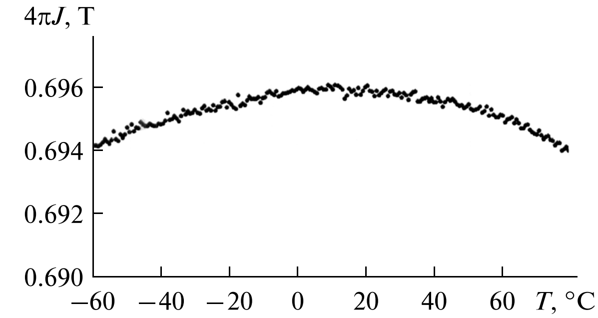

Fig. 2. Temperature dependence of the magnetization of $(Pr_{0.52}Dy_{0.48})_{13.4}(Fe_{0.65}Co_{0.35})_{bal}Cu_{1.3}B_{6.8}$ samples measured in a field $H = 0$ kA/m; $\Delta_{-60°C} = \Delta_{+80°C} = 0.29\%$, $T_{max} = 10°C$.

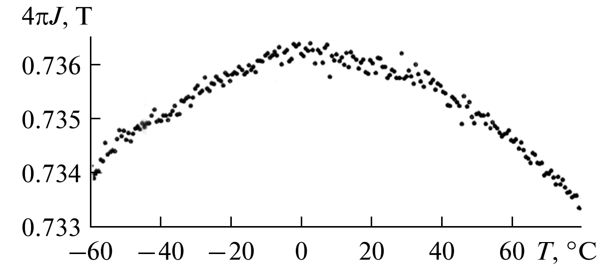

Fig. 3. Temperature dependence of the magnetization of $(Pr_{0.52}Dy_{0.48})_{13.3}(Fe_{0.65}Co_{0.35})_{bal}Cu_{2.1}B_{6.8}$ samples measured in a field $H = 0$ kA/m; $\Delta_{-60°C} = 0.29\%$, $\Delta_{+80°C} = 0.40\%$, $T_{max} = 0°C$.

acterized by the position of the maximum and the ratios

$$\Delta_{-60°C} = [J(T_{max}) - J(-60°C)]100/J(-60°C)$$

$$\text{and } \Delta_{+80°C} = [J(T_{max}) - J(+80°C)]100/J(+80°C),$$

where $J(T_{max})$, $J(-60°C)$, and $J(+80°C)$ are the magnetizations at temperatures T_{max}, −60, and +80°C, respectively. It follows from the data given in the table that no regularities in the temperature of maximum T_{max} are observed. Moreover, no maximum (and T_{max}) in the temperature dependence of the magnetization of the $(Pr_{0.52}Dy_{0.48})_{13.1}(Fe_{0.65}Co_{0.35})Cu_{3.3}B_{6,7}$ sample is observed. It is also seen from the table that, as the copper content increases from 1.3 to 6.3 at %, the magnetization corresponding to temperature T_{max} increases monotonically from 0.696 to 0.805 T. The magnetization of the $(Pr_{0.52}Dy_{0.48})_{13.6}(Fe_{0.65}Co_{0.35})_{79.5}B_{6.9}$ sample cannot be used for a comparison, since the measurements were performed in a field of 320 kA/m.

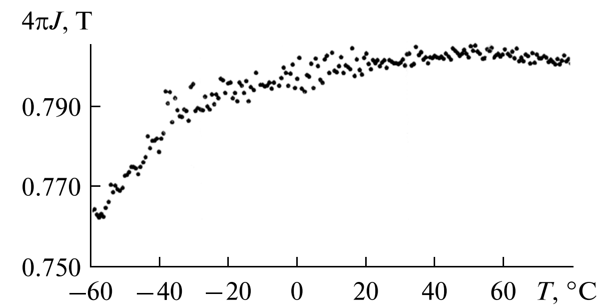

Fig. 4. Temperature dependence of the magnetization of $(Pr_{0.52}Dy_{0.48})_{12.7}(Fe_{0.65}Co_{0.35})_{bal}Cu_{6.3}B_{6.5}$ samples measured in a field $H = 0$ kA/m; $\Delta_{-60°C} = 5.48\%$, $T_{max} = 60°C$.

DISCUSSION OF RESULTS

Note again that there is no sense to compare the results given in Figs. 1–4 using the values of TCI. It is obvious that, for the data in Figs. 1–4, the values of TCI for the temperature range from −60 to +80°C can be used but it cannot characterize the behavior of the $J(T)$ dependence. Because of this, we use the T_{max}, $\Delta_{-60°C}$, and $\Delta_{+80°C}$ parameters to characterize the temperature dependence of the magnetization. We showed in our previous study [4] that the effect of copper on TCI can be explained by the substitution of copper atoms for iron atoms at lattice sites $16k_2$ and $8j_1$. In this case, nonmagnetic copper atoms that substitute for iron atoms at the $3d$ sites suppress some antiferromagnetic

Parameters* characterizing the temperature dependence of magnetization $J(T)$ measured at temperatures from −60 to +80°C in applied magnetic field H

Sample material	$\Delta_{-60°C}$	$\Delta_{+80°C}$	T_{max}, °C	H, kA/m	4πJ**, T
	%				
$(Pr_{0.52}Dy_{0.48})_{13.6}(Fe_{0.65}Co_{0.35})_{79.5}B_{6.9}$	1.10	0.71	30	320	0.847
$(Pr_{0.52}Dy_{0.48})_{13.4}(Fe_{0.65}Co_{0.35})_{bal}Cu_{1.3}B_{6.8}$	0.29	0.29	10	0	0.696
$(Pr_{0.52}Dy_{0.48})_{13.3}(Fe_{0.65}Co_{0.35})_{bal}Cu_{2.1}B_{6.8}$	0.29	0.40	0	0	0.736
$(Pr_{0.52}Dy_{0.48})_{12.7}(Fe_{0.65}Co_{0.35})_{bal}Cu_{6.3}B_{6.5}$	5.48	–	60	0	0.806
$(Pr_{0.52}Dy_{0.48})_{13.1}(Fe_{0.65}Co_{0.35})_{bal}Cu_{3.3}B_{6.7}$	–	–	–	0	0.686

* Δ is the relative maximum in the magnetization curve.

** The values of 4πJ are given for temperature T_{max}; the values of 4πJ for the sample with 3.3 at % Cu is given at $T = +80°C$.

exchange interactions of the nearest magnetic moments coupled with the magnetic moment of a selected site. This inevitably should affect the temperature dependence of magnetization J and Figs. 1–4 demonstrate this effect. The cause for the absence of maximum in the dependence $J(T)$ for the $(Pr_{0.52}Dy_{0.48})_{13.1}(Fe_{0.65}Co_{0.35})_{bal}Cu_{3.3}B_{6.7}$ sample is still unclear and call for a further investigation.

Let us consider the measurement conditions in detail. The demagnetizing factor magnitude for a spherical sample is $N = 4\pi3$ [5]. Hence, we can easily estimate the internal magnetizing field in which measurements were performed in the absence of external magnetic field (200 kA/m), and this field is opposite to $J(T)$. The measurements for the sample free from copper were performed in a field of 100 kA/m that is parallel to $J(T)$. It is likely that, because of this fact, $4\pi J$ corresponding to T_{max} for this sample is substantially higher than that for the copper-alloyed samples (see table).

CONCLUSIONS

It was shown that the temperature dependences of the magnetization of sintered $(Pr_{0.52}Dy_{0.48})_{13}(Fe_{0.65}Co_{0.35})_{bal}Cu_xB_{6.7}$ materials with $x = 0$, 1.3, and 2.1 at % has a maximum in the temperature range from −60 to +80°C. The relative maximum height decreases with increasing copper content.

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