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High-Temperature Magnetic Properties of Mechanically Alloyed SmCo$_5$ and YCo$_5$ Magnets

I. A. Al-Omari, R. Skomski, R. A. Thomas, D. Leslie-Pelecky, and D. J. Sellmyer

Abstract—The high-temperature coercivity of mechanically alloyed and subsequently annealed RCo$_5$ (R = Sm and Y) is studied. The annealed materials have the hexagonal CaCu$_5$ structure with 2:17 (or 1:7) regions as a minor phase. High-temperature magnetic measurements show that the coercivities of materials decrease with increasing temperature from room-temperature to 873 K, but that the temperature coefficient of the coercivity of YCo$_5$ is much smaller than that of SmCo$_5$. This behavior is explained in terms of the intrinsic temperature variation of the magnetocrystalline anisotropy.

Index Terms—Anisotropy, coercivity, finite-temperature magnetism, permanent magnets.

I. INTRODUCTION

RECENTLY, samarium–cobalt based permanent magnets [1], [2] have attracted renewed interest due to their superior high-temperature properties [3]–[5]. In rare-earth transition-metal intermetallics, such as SmCo$_5$ and Nd$_2$Fe$_{14}$, the rare-earth anisotropy is responsible for the high anisotropy (and coercivity), whereas the transition-metal sublattice produces a high saturation magnetization and Curie temperature [2], [6]. In almost all cases of interest, the magnitudes of the cobalt moments are somewhat lower than isotructural iron moments, but the strong interatomic exchange ensures a high Curie temperature and helps to realize a strong rare-earth anisotropy at and above room temperature. Typical Sm–Co based permanent magnets are produced by sintering and are used as bonded magnets (single-phase SmCo$_5$ based) or as Sm$_2$Co$_{17}$/SmCo$_5$ based two-phase magnets [1], [2], although appreciable room-temperature coercivities of 17 kOe [1.7 T] can also be obtained by mechanical alloying [7].

The pronounced temperature dependence of the rare-earth anisotropy makes the rare-earth sublattice comparatively unimportant at high temperatures [6], [8]. This effect is particularly important in rare-earth cobalt magnets with the hexagonal CaCu$_5$ structure, where the Co atoms yield an unusually strong transition-metal contribution to the anisotropy. In fact, the first 1:5 permanent magnets in the late 1960s were YCo$_5$ magnets, and only in the early 1970s, when people began to recognize the role of the rare-earth anisotropy, did emphasis shift toward samarium–cobalt magnets.

The practical idea behind the present work is to explore the feasibility of rare-earth free and therefore comparatively cheap high-temperature permanent magnets with moderate energy products (1 to 10 MGOe).

II. SAMPLE PREPARATION AND STRUCTURE

SmCo$_5$ and YCo$_5$ alloys were prepared by mechanical alloying from elemental powders. The starting Sm and Y powders are ϕ40 mesh and 99.9% purity, while the Co powder is ϕ325 mesh with a puritiesy of 99.8%. The powders were alloyed and handled in an argon-filled glove box to prevent oxidation. The milling was performed in a hermetically sealed tungsten-carbide-lined vial in a SPEX 800 mixer/mill inside the argon-filled glove box, using a 3:1 ball-to-powder mass ratio. The milling was interrupted every two hours to remove a small amount of powder for x-ray diffraction and to break up clumps of powder. The x-ray diffraction patterns of the milled and unmilled powders are very similar to those shown in [7]. After milling for 16 hours, the SmCo$_5$ material has an amorphous structure, but annealing at 800°C for 5 min yields sharp diffraction peaks corresponding to SmCo$_5$ with 2:17 (or 1:7) regions as a minor phase. The YCo$_5$ samples, milled for 18 hours and annealed at 900°C for 5 min, exhibit a similar behavior.

III. MAGNETIC PROPERTIES

The samples were prepared by mixing the powder with Omega high-temperature cement and magnetizing in a field of 18 kOe. Hysteresis loops were measured by a vibrating sample magnetometer (VSM) in fields up to 10 kOe and at temperatures from 20°C to 630°C.

Fig. 1 shows the coercivities of SmCo$_5$ and YCo$_5$ as functions of temperature. The coercivity decreases with increasing temperature for both materials. For SmCo$_5$, the coercivity decreases from 11 kOe at room temperature to 0.15 kOe at 500°C, whereas the respective values for YCo$_5$ are 3.6 kOe and 0.6 kOe. In contrast, the shape of the hysteresis loops (not shown here) did not exhibit any significant temperature or materials dependence.

IV. DISCUSSION

To explain the temperature behavior of the coercivity, we first analyze the temperature dependence of the anisotropy, which governs the temperature dependence of the coercivity, and then briefly discuss some micromagnetic aspects of the problem.
A. Temperature Dependence of the Anisotropy

As a rough approximation, the “intrinsic” coercivity $H_c = iH_c$ of permanent magnets scales as

$$H_c = 2\alpha K_1/\mu_0 M_s$$  \hspace{1cm} (1)

where $K_1$ is the first-order anisotropy constant, $M_s$ is the spontaneous magnetization, and $\alpha$ is a real-structure-dependent dimensionless factor.

Usually, $\alpha \approx 0.3$ for optimized permanent magnets [9]. The main contribution to the temperature dependence of the coercivity originates from the temperature dependence of $K_1$ [2], [8]. $M_s$ is much less temperature dependent [8], whereas $\alpha$ is essentially constant unless there are irreversible structural changes on heating.

Fig. 2 shows the temperature dependence of $K_1$ for a NdCo$_5$, YCo$_5$, and SmCo$_5$ [2]. The convergent character of the curves shows that the rare-earth anisotropy is less important at high temperatures: the magnetization of the rare-earth ions must be coupled to the magnet’s main transition-metal magnetization, but the rare-earth transition-metal intersublattice exchange is comparatively weak and easily overcome by thermal excitation [2], [6]. The striking anisotropy differences between isostructural rare-earth compounds—compare NdCo$_5$ and SmCo$_5$ in Fig. 2—are well known to reflect the shape of the rare-earth 4$f$ electron clouds (see e.g., [2], [8]). The shape of the 4$f$ shells is given by the Stevens factor $\alpha_f$ in the case of uniaxial crystals (hexagonal, tetragonal, and rhombohedral). The elements Ce, Pr, Nd, Tb, Dy, and Ho have oblate (pancake-like) 4$f$ shells ($\alpha_f < 0$), whereas Sm, Er, Tm, and Yb are characterized by prolate (cigar-like) 4$f$ shells ($\alpha_f > 0$). In a given crystalline environment, prolate and oblate ions give opposite anisotropy contributions, which explains the different low-temperature anisotropies of NdCo$_5$ and SmCo$_5$.

Gadolinium, which has a half-filled 4$f$ shell, and the “non-magnetic” rare earths Y, La, and Lu exhibit 4$f$ shells with spherical symmetry, so that $\alpha_f = 0$ and the corresponding anisotropy contribution is zero. The anisotropy of YCo$_5$ therefore originates from the Co sublattice. Figs. 1 and 2 show that the rare-earth contribution to anisotropy and coercivity is negligible at high temperatures.

B. Micromagnetic Effects

The intrinsic temperature dependence of the anisotropy (Fig. 2) provides a qualitatively correct explanation of the coercivity. However, Fig. 1 shows that the high-temperature coercivity of YCo$_5$ is actually somewhat higher than that of SmCo$_5$. This supports our original idea that Sm may well be replaced by Y, but it doesn’t make sense from a purely intrinsic point of view. In terms of Eq. (1), the reason for this effect is well known: small structural changes may give rise to disproportionally large changes in the parameter $\alpha$ [2]. The YCo$_5$ and SmCo$_5$ magnets are structurally very similar (nanostructured random-anisotropy magnets), but there remain small differences in stoichiometry and grain structure that produce easily measurable coercivity deviations.

It is important to keep in mind that the present predictions refer to single-phase materials. Two-phase materials, such as cellular 1 : 5/2 : 17 magnets, exploit subtle differences between the anisotropies of the phases involved, and the coercivity may actually reach a maximum at high temperatures [2]–[5].

As a first-order approach, the magnets can be considered as isotropic and weakly interacting ensembles of Stoner–Wohlfarth particles. In the hard-magnetic limit [10], the energy product can be approximated by

$$(BH)_{\text{max}} = \mu_0 M_s^2/16 \left(1 - \frac{\mu_0 M_s^2}{K_1} + \frac{4A}{R^2 K_1} \right)$$  \hspace{1cm} (2)

where $A$ is the exchange stiffness and $R$ is an appropriately defined average grain radius. For $K_1 = 8$, the “ideal” isotropic energy product of $\mu_0 M_s^2/16$ is reproduced, whereas the second and third terms in the bracket describe the onset of soft magnetism and the remanence enhancement, respectively. Equation (2) shows that very hard isotropic materials with sufficiently small grain sizes exhibit a remanence-enhanced energy product.
but it is difficult to predict to what extent this remains true at elevated temperatures, where the magnets become softer.

V. CONCLUSIONS

We have shown that the high-temperature performance of the Y–Co magnets is comparable to that of similarly processed isotructural Sm–Co magnets. This may be used to develop rare-earth-free high-temperature permanent magnets having moderate energy products.

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REFERENCES


