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Magnetic properties of $Sm_2Fe_{14-x}Co_xAl_3$ compounds

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Compounds in the series $Sm_2Fe_{14-x}Co_xAl_3$ ($x = 0, 1, 2, 3, 4$ and 5) have been shown to be of the Th_2Zn_{17} structure. The Curie temperature is found to increase monotonically from a critical temperature, T_c equal to 471 K for the $x=0$ sample, to $T_c=681$ K for the $x=5$ sample. X-ray diffraction measurements of magnetic field oriented powders showed that all compounds exhibit a room temperature uniaxial anisotropy. Magnetization measurements show that the magnetic anisotropy of $Sm_2Fe_{14}Al_3$ can be increased substantially by the substitution of even a small quantity of Co for Fe. Results are discussed in terms of possible applications of these compounds as particulate recording media.

1. Introduction

The hard magnetic properties of the rare earth iron compounds of the composition R_2Fe_{17} have been studied extensively in recent years. These materials can be formed as single phased compounds across the entire rare earth series [1], and show high values of saturation magnetization [2]. Typical properties of these compounds include relatively low Curie temperatures [3] and easy plane anisotropy at room temperature. In fact none exhibit an easy axis anisotropy at room temperature and with the exception of Tm_2Fe_{17}, which exhibits an easy axis anisotropy below 72 K [4], there is no clear evidence in any R_2Fe_{17} binary compound for a spin reorientation transition at low temperature. In order to increase the Curie temperature of these materials and to induce room temperature uniaxial anisotropy, a number of investigations concerning the substitution of other elements into the R_2Fe_{17} structure, or the preparation of materials with interstitial atoms, have been undertaken. While the substitution of Co, Ni and Si for Fe [5–8], as well as the introduction of interstitial hydrogen, nitrogen and carbon [9–14], in many of these materials will substantially increase the Curie temperature, only a few cases of room temperature uniaxial anisotropy have been reported. Wang and Dunlap have shown that substitution of Al or Ga for Fe in Sm_2Fe_{17} can introduce uniaxial anisotropy [15, 16]. As well, the introduction of interstitial carbon and nitrogen has been shown to yield uniaxial anisotropy in the case of $Sm_2Fe_{17}C_y$ ($y > 0.5$) and $Sm_2Fe_{17}N_y$ ($y > 0.8$) [9, 12]. In some cases (e.g. for interstitial hydrogen) it has been shown that compositional changes that are advantageous in increasing the Curie temperature are detrimental to the formation of uniaxial anisotropy [17]. Recent studies of $Sm_2Fe_{17-x}Al_x$ compounds [15–17] have shown that a room temperature uniaxial anisotropy is produced for $x \geqslant 3$, and that the Curie temperature is a maximum for $x = 3$. Here new results are reported concerning investigation of the magnetic properties of $Sm_2Fe_{14-x}Co_xAl_3$ compounds. These studies demonstrate that the Curie temperature of $Sm_2Fe_{14}Al_3$ can be increased substantially by the substitution of small quantities of Co for Fe while simultaneously increasing the saturation magnetization and the magnetic anisotropy. The measured magnetic properties of these compounds suggest that they are suitable materials for use as recording media and possible applications in this area will be discussed.

2. Experimental procedure

The compounds $Sm_2Fe_{14-x}Co_xAl_3$ (with $x = 0, 1, 2, 3, 4, 5$) were prepared by arc melting high purity elemental components, followed by annealing at 1273 K for 72 h and quenching into ice water. Ingots were then ground to yield powder samples.

Room temperature X-ray diffraction measurements were made using a Siemens D500 scanning diffractometer with CuK_α radiation. Magnetic anisotropy studies were undertaken using X-ray diffraction methods on samples which had been field oriented. Powders with particles of diameter 125 μm or less were mixed with epoxy resin and aligned at room temperature in an external field of 1.0 T.

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Curie temperatures were obtained from calorimetry measurements carried out on a Fisher Series 300 quantitative differential thermal analyser. Magnetization measurements were made at 10 K in applied magnetic fields up to 3.0 T using a Quantum Design superconducting quantum interference device (SQUID) magnetometer.

3. Results

A room temperature X-ray diffraction pattern of the $Sm_2Fe_{10}Co_4Al_3$ compound is illustrated in Fig. 1a. Indices of the major peaks for the rhombohedral Th_2Zn_{17} phase are indicated in the figure. No significant diffraction from impurity phases is observed. Diffraction patterns of the other alloys studied in the present work show that these alloys are also single phase compounds of the rhombohedral Th_2Zn_{17} structure. Lattice parameters, as derived from the present measurements, are given in Table I. The lattice parameters are seen to decrease with increasing Co content. This change occurs isotropically as evidenced by the fact that the c/a ratios are independent of composition.

A typical room temperature CuK_α X-ray diffraction pattern obtained for the $Sm_2Fe_{14-x}Fe_xAl_3$ compounds after alignment in an applied magnetic field of 1.0 T is illustrated in Fig. 1b. Indices of the existent diffraction peaks are shown in the figure. X-ray diffraction patterns of the other compounds studied here showed the existence of the same peaks as seen in Fig. 1b. The characteristic magnetic anisotropy of the sample is determined on the basis of the X-ray peaks present. The present results indicate an alignment of the samples along the c-axis and are consistent with the existence of a uniaxial anisotropy in all samples.

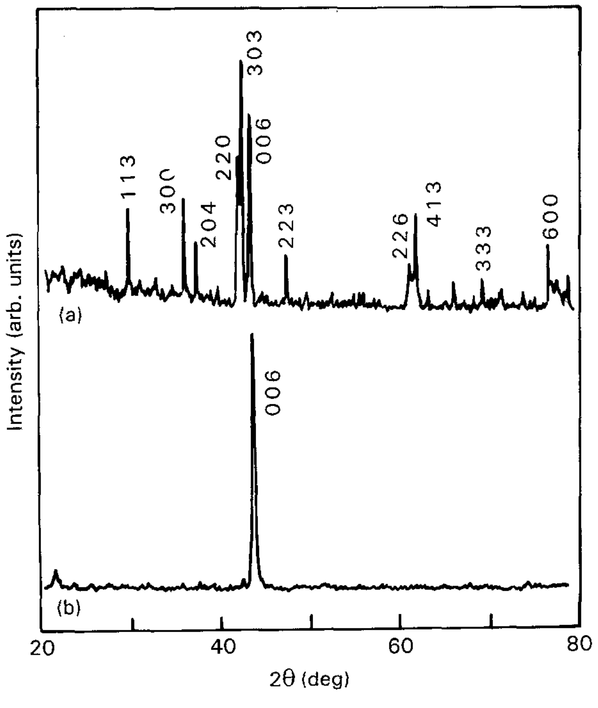

Figure 1 Room temperature CuK_α X-ray diffraction pattern for $Sm_2Fe_{10}Co_4Al_3$: (a) unoriented and (b) field oriented. Indices for the Th_2Zn_{17} phase are indicated.

Curie temperatures measured for the $Sm_2Fe_{14-x}Co_xAl_3$ series of alloys are shown as a function of composition in Fig. 2, and indicate that the substitution of Co for Fe results in a consistent and substantial

TABLE I Lattice parameters obtained from X-ray diffraction studies of single phase $Sm_2Fe_{14-x}Co_xAl_3$ compounds with the Th_2Zn_{17} structure

x	a (nm)	c (nm)	c/a
0	0.8642	1.2562	1.453
1	0.8641	1.2531	1.450
2	0.8624	1.2527	1.452
3	0.8609	1.2515	1.453
4	0.8595	1.2506	1.454
5	0.8595	1.2486	1.452

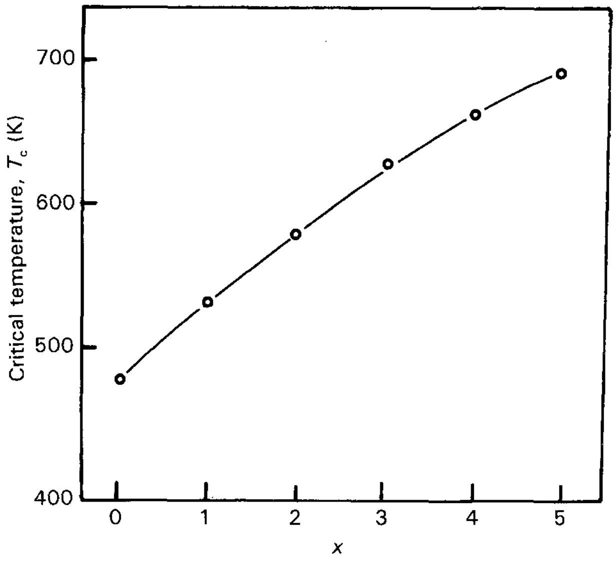

Figure 2 Composition dependence of the measured Curie temperature of $Sm_2Fe_{14-x}Co_xAl_3$ compounds.

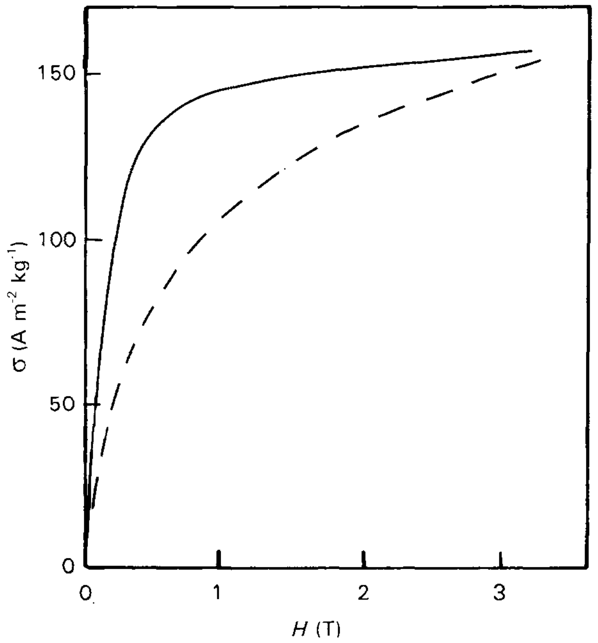

Figure 3 Easy axis (——) and hard axis (– – –) magnetization curves obtained at 10 K for the compound $Sm_2Fe_{11}Co_3Al_3$.

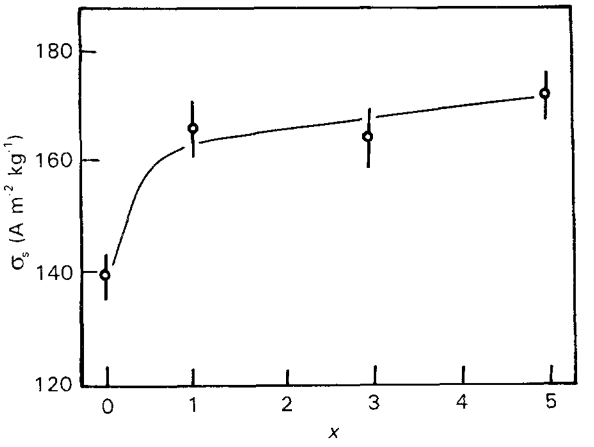

Figure 4 Composition dependence of the saturation magnetization (measured in an applied field of 3.0 T) of the $Sm_2Fe_{14-x}Co_xAl_3$ compounds obtained at a temperature of 10 K. The solid line is not a fitted curve.

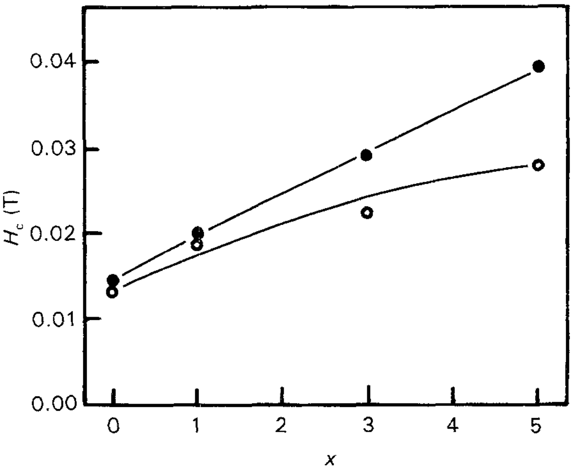

Figure 5 Composition dependence of the coercivity of the $Sm_2Fe_{14-x}Co_xAl_3$ compounds obtained at a temperature of 10 K; easy axis (o) and hard axis (●) data are shown.

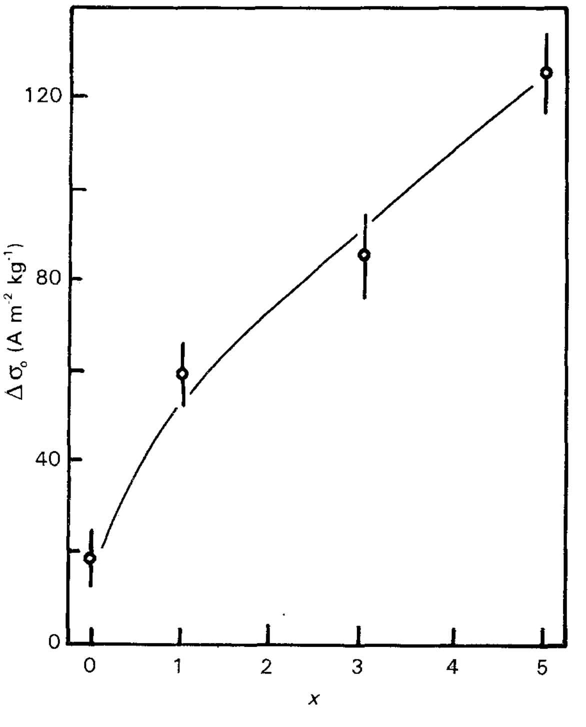

Figure 6 Composition dependence of the values of $\Delta\sigma_o$ of the $Sm_2Fe_{14-x}Co_xAl_3$ compounds at 10 K.

increase in magnetic ordering temperature. Magnetization measurements were made at 10 K for magnetically aligned samples oriented in both the easy and hard magnetic directions, as illustrated in Fig. 3. Values of the saturation magnetization have been obtained from easy axis magnetization curves and are shown as a function of compound composition in Fig. 4. The results show that the addition of small quantities (i.e. $x = 1$) of Co to the present compounds increases the magnetization, although further increases in the Co content have a comparatively smaller effect. As well, hysteresis loops as obtained from easy axis magnetization measurements have yielded values of the coercivity as illustrated in Fig. 5. The coercivities of these compounds are relatively small, typically of the order of 0.01 T. The difference between the easy axis magnetization and the hard axis magnetization, $\Delta\sigma = \sigma_{easy} - \sigma_{hard}$, has been extrapolated to zero applied field. This is a measure of the magnetic anisotropy and is illustrated in Fig. 6. The present results indicate a consistent increase in the anisotropy field as Co is substituted for Fe in these compounds.

4. Discussion

The uniaxial anisotropy observed for all compounds studied in this work may be compared with the results for Sm_2Fe_{17}, which exhibits an easy plane anisotropy at room temperature. In contrast to earlier reports [7] that the substitution of Al or small amounts of Co for Fe in Sm_2Fe_{17} does not alter the planar anisotropy; previous investigations of $Sm_2Fe_{17-y}Al_y$ [15] and the present results on the $Sm_2Fe_{14-x}Co_xAl_3$ series of compounds clearly show that uniaxial anisotropy is developed in alloys with sufficient Al and Co content. In many cases previous studies have dealt with limited ranges of compositions and in some cases the magnetic anisotropy was not considered carefully or was assumed to be planar on the basis of other studies.

The net anisotropy in the R_2Fe_{17} compounds is determined by the sum of the Fe sublattice anisotropy and the rare earth sublattice anisotropy [3, 10, 11, 18]. Since the Fe sublattice exhibits a planar anisotropy, a uniaxial anisotropy of the rare earth sublattice is necessary to yield a material with a net easy axis behaviour. A uniaxial anisotropy exists when the product of the second order Stevens coefficient, α_J, and the second order crystal field parameter, A_2^0 is negative. As A_2^0 is typically negative in the 2:17 compounds [3, 10], it is in cases where the second order Stevens coefficient is positive that easy axis anisotropy is favoured. This explains the tendency for Sm (which has positive α_J) compounds to exhibit uniaxial anisotropy, while compounds of many other rare earths with negative α_J exhibit planar anisotropy. Previous studies [15] of $Sm_2Fe_{17-x}Al_x$ compounds have shown that increasing the Al content of the compound increases the magnitude of the negative A_2^0 and increases the uniaxial contribution of the Sm sublattice to the total anisotropy. As Sm_2Co_{17} shows a room temperature uniaxial anisotropy it is not surprising that the substitution of Co for Fe in $Sm_2Fe_{14}Al_3$ does not alter the uniaxial anisotropy.

The Curie temperature of rare earth–transition metal compounds is primarily dependent on the magnitude of the exchange coupling between transition metal atoms, J_{TT} [7, 8]. The generally low values of the Curie temperature in R_2Fe_{17} compounds result in a weak transition metal exchange coupling due to the relatively small Fe–Fe distance in these materials. The increase in Curie temperature observed seen in many substituted rare earth–transition metal compounds and compounds with interstitial atoms typically results from an increased exchange coupling corresponding to an increase in lattice parameter.

The Curie temperature can be increased by up to about 80 K in Sm_2Fe_{17} by the substitution of Al for Fe [15]. The Co containing alloys studied here show that a further, substantial increase in the Curie temperature results for the partial substitution of Co for Fe in the $Sm_2Fe_{14}Al_3$ compound. This indicates a strong ferromagnetic Fe–Co coupling which increases the Curie temperature more rapidly with increasing Co content than the possible reduction in T_c caused by the decrease in Fe–Fe neighbour distance.

5. Conclusions

In conclusion, the possible applications of the materials studied in the present work may be considered. The possibility of producing commercially viable hard magnetic materials in cost-effective Fe-based rare earth compounds has been one of the motivating factors for much of the recent work on 2 : 17 materials; the present compounds are promising in this respect as they exhibit uniaxial anisotropy, high values of the Curie temperature and high saturation magnetization. However, the values of the coercivities observed in this work are, like those of many rare earth–transition metal compounds, relatively small. This is evidenced as well by the differences between the easy axis and hard axis magnetization curves. The materials studied here, however, have magnetic properties which are compatible with their application as particulate recording materials [e.g. 19]. The 2 : 17 Sm–Fe–Co–Al compounds offer certain advantages in this area:

1. good thermal stability,
2. relative ease of preparation,
3. high saturation magnetizations,
4. suitable coercivities, and
5. relatively low cost of components.

In this latter case the compound with $x = 1$ has the advantage of an increased saturation magnetization and magnetic anisotropy over the $Sm_2Fe_{14}Al_3$ compound, while minimizing the Co content of the material. Increasing the Co content further increases the magnetic anisotropy, although it yields little benefit to the saturation magnetization and is clearly disadvantageous from a cost standpoint.

The present results indicate the possibility of the use of these materials in particulate recording media and suggest the suitability of further investigations directed towards this area of application.

References

1. J. M. D. COEY and H. SUN, *J. Magn. Magn. Mater.* **87** (1990) L251.
2. K. H. J. BUSCHOW, in "Ferromagnetic Materials", Vol. 2, edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1980) p. 297.
3. Y. OTANI, D. P. F. HURLEY, H. SONG and J. M. D. COEY, *J. Appl. Phys.* **69** (1991) 5584.
4. K. H. J. BUSCHOW, *Rep. Prog. Phys.* **40** (1977) 1179.
5. K. S. V. L. NARASIMHAN, W. E. WALLACE and R. D. HUTCHENS, *IEEE Trans. Magn.* **10** (1974) 729.
6. D. McNEELY and H. OESTERREICHER, *J. Less Common Metals* **44** (1976) 183.
7. B. P. HU, X. L. RAO, J. M. XU, G. C. LIU, X. L. DONG, H. LI, L. YIN and Z. R. ZHAO, *J. Magn. Magn. Mater.* **114** (1992) 138.
8. T. H. JACOBS, K. H. J. BUSCHOW, G. F. ZHOU, X. LI and F. R. DE BOER, *J. Magn. Magn. Mater.* **116** (1992) 220.
9. X. C. KOU, R. GROSSINGER, T. H. JACOBS and K. H. J. BUSCHOW, *Physics B* **168** (1991) 181.
10. J. M. D. COEY, H. SUN and D. P. F. HURLEY, *J. Magn. Magn. Mater.* **101** (1991) 310.
11. Q. N. QI, M. D. KUZ'MIN, H. SUN and J. M. D. COEY, *J. Alloys & Compounds* **182** (1992) 313.
12. M. KATTER, J. WECKER, C. KUHRT, L. SCHULTZ and R. GROSSINGER, *J. Magn. Magn. Mater.* **117** (1992) 419.
13. X. Z. WANG, K. DONNELLY, J. M. D. COEY, B. CHEVALIER, J. ETOURNEAU and T. BERLUREAU, *J. Mater. Sci.* **23** (1988) 329.
14. B. P. HU and J. M. D. COEY, *J. Less Common Metals* **142** (1988) 295.
15. Z. WANG and R. A. DUNLAP, *J. Phys. Condens. Matter* **5** (1993) 2407.
16. *Idem, Phil. Mag. B* **69** (1994) 103.
17. R. A. DUNLAP and Z. WANG, *Can. J. Phys.* **71** (1993) 574.
18. J. M. D. COEY, H. LI, J. P. GAVIGAN, J. M. CADOGAN and B. HU, in "Concerted European Action on Magnets", edited by I. V. Mitchell, J. M. D. Coey, D. Givord, I. R. Harris and R. H. Anitsch (Elsevier, London, 1989) p. 76.
19. L. WENBE and K. HONG, *IEEE Trans. Magn.* **21** (1985) 1503.

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