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Magnetic and structural properties of $SmCo_{7-x}Cu_x$ alloys

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We report the structural and magnetic properties of $SmCo_{7-x}Cu_x$, where $x=0$, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.7. X-ray diffraction shows that these alloys from the disordered hexagonal $TbCu_7$-type structure. For large values of $x(x\geqslant 0.8)$ the hexagonal $TbCu_7$-type structure cannot be formed. X-ray diffraction on magnetically aligned samples show that these samples have uniaxial anisotropy. The lattice parameters (a and c) are dependent on the Cu concentration, and the unit cell volume is found to increase with x. The saturation magnetization decreases with x at both room temperature and 25 K. The Curie temperature increases with x for small values of x while it decreases with x for large values of x. A maximum value of $T_C=852\,^{\circ}\mathrm{C}$ is found in these alloys.
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I. INTRODUCTION

In the last 30 years, there has been an intensive search for new iron-rich or cobalt-rich rare-earth intermetallic compounds for magnetic applications including materials for room temperature permanent magnets, high temperature permanent magnets, magnetic recording, etc. The compounds ($R_xFe_yCo_z$) include materials with atomic ratios of rare-earth to iron and cobalt 1:5, 1:7, 1:12, and 2:17 with different types of structure. Most of the R–Fe compounds have low Curie temperature (T_C), relatively low saturation magnetization (M_s), small magnetic anisotropy, in-plane anisotropy, and are unstable at high temperature which lowers their potential as materials for high temperature applications.[1–7] The disordered $TbCu_7$-type or so-called 1:7 structure shows interesting magnetic properties when Co or other elements are substituted for Fe.

The metastable $TbCu_7$-type structure of $Sm(Fe,Ti)_7$ or $Sm(Fe,V)_7$ can be formed under certain preparation conditions. Saito *et al.*[8] studied $SmFe_{11}Ti$ alloy ribbons and found that the structure changes from a tetragonal $ThMn_{12}$-type structure to a hexagonal $TbCu_7$-type structure by changing the roller velocity. They also found that ribbons with a $ThMn_{12}$-type structure give the maximum hard magnetic properties. Xiao *et al.*[9] studied the Sm–Fe–Ti system and found that this system crystallizes in the metastable $TbCu_7$-type structure with an easy in-plane magnetization and has a T_C of 243 °C. The metastable $TbCu_7$-type structure transforms to a $ThMn_{12}$ structure (with $T_C=305\,^{\circ}\mathrm{C}$ and easy in-plane magnetization) at an annealing temperature higher than 740 °C. Katter *et al.*[10] studied Sm–Fe–N and found that $Sm_{10.6}Fe_{89.4}N_y$ forms the $TbCu_7$-type structure with a coercivity of 6.0 kOe, a remanence of 684 emu/cm^3, and an energy product $(BH)_{\max}$ of 8.74 MGOe, while $Sm_{12}Fe_{88}$ crystallizes in the rhombohedral Th_2Zn_{17} structure. This study also showed that T_C and M_s change from 200 °C and 987 emu/cm^3 for $Sm_{10.6}Fe_{89.4}$ to 470 °C and 1114 emu/cm^3 for $Sm_{10.6}Fe_{89.4}N_y$. A study of R–Cu compounds by Buschow and Van Der Gast[11] showed that for R=Gd, Tb, Dy, and Y a compound of the approximate composition RCu_7 can be formed with the $TbCu_7$ structure and these compounds decompose with annealing at low temperatures into RCu_5 and elementary Cu. They also found that c/a is about 0.84 for RCu_7 compounds while it is about 0.80 for RCu_5 compounds. Huang *et al.*[12] found c/a ratios of 0.82–0.83 for $Sm(Co,Zr)_7$ alloys and we also found the same ratios in our $Sm(Co,Ti)_7$ alloys.[13] Suzuki *et al.*[14] studied $Sm_{10}(Fe,V)_{90}N_y$ and found that the substitution of vanadium for iron in $Sm_{10}(Fe,V)_{90}$ alloys gives a great range of stability in the $TbCu_7$-type structure, where this structure can be formed for $5<V<10$. They also found that nitrogenation of the samples improves the magnetic properties including a $(BH)_{\max}$ value of 8.0 MGOe and a T_C value of 477 °C for $Sm_{10}Fe_{82.5}V_{7.5}N_y$. Chen *et al.*[15] studied $SmCo_x$ alloys by melt spinning and found that the alloys exhibit a single phase $SmCo_5$ and Sm_2Co_{17} structure for $x=5.0$ and $x=8.8$, respectively, while a three-phase structure ($Sm_2Co_{17}, SmCo_5, SmCo_3$) appears for $5.0<x<8.5$. Recently, studies by Lefever *et al.*[16,17] and by Huang *et al.*[12] showed that a small amount of Zr substitution could contribute to the stabilization of the hexagonal $TbCu_7$ structure and improve the magnetic anisotropy in Sm–Co–Zr compounds. An anisotropy field (H_A) value of 180 kOe and a T_C value of 750 °C for $SmCo_{6.5}Zr_{0.5}$ have been reported by Huang *et al.*[12] The $TbCu_7$-type structure could be indexed according to the $CaCu_5$-type structure with significant deviation of the lattice constants and x-ray peaks' intensities.

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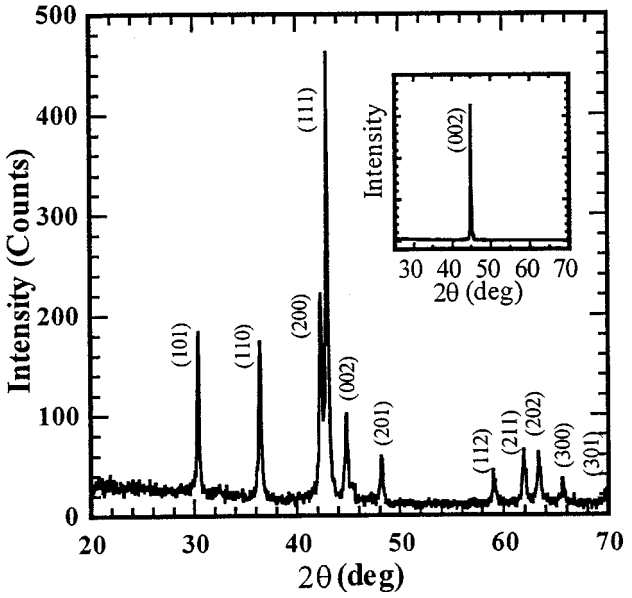

FIG. 1. Typical x-ray diffraction pattern for a $SmCo_{6.7}Cu_{0.3}$ alloy. The inset shows a typical x-ray diffraction pattern for the same sample after it has been magnetically aligned.

The aim of this article is to study the effect of Cu substitution for Co on the magnetic and structure properties of $SmCo_{7-x}Cu_x$ alloys.

II. EXPERIMENTAL PROCEDURE

Bulk samples of $SmCo_{(7\pm\delta)-x}Cu_x$, where δ is between 0 and 2 and $x=0$, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.7, were prepared by arc melting the elements of at least 99.9% purity in a water-cooled copper boat in a flowing-argon gas atmosphere. The alloys were melted four to five times to insure homogeneity. The phase purity for all the samples was determined by x-ray diffraction using Cu K_α radiation. The magnetization of the alloys was measured by a superconducting quantum interference device (SQUID) magnetometer in the temperature range 25–300 K and in fields from 0 to 50 kOe. High temperature magnetic measurements were done by a vibrating sample magnetometer (VSM) in the temperature range 300–1273 K.

III. RESULTS AND DISCUSSION

Figure 1 shows a typical x-ray diffraction pattern for a $SmCo_{6.7}Cu_{0.3}$ alloy. From Fig. 1 we see that the sample crystallizes in the hexagonal $TbCu_7$-type structure. Samples with different Sm to (Co, Cu) atomic ratios showed different structures. For example, if the Sm-to-(Co, Cu) atomic ratios are more than (1/7) a hexagonal $CaCu_5$-type structure formed and if the atomic ratio is less than (1/7) a hexagonal Th_2Ni_{17}-type structure formed. This is in agreement with other observations by Khan[18,19] for $RCo_{5\pm x}$. In this article we are interested in alloys with the $TbCu_7$-type structure; therefore, we present the results for the samples with 1:7 composition. All the samples with the 1:7 composition showed the $TbCu_7$-type structure except for $x=0$ where a minor 2:17-type structure appears. X-ray diffraction shows that there is a shift in the peaks with increasing Cu concentration, which is due to the difference in the atomic volume. Table I summarizes the lattice parameters a and c obtained from the x-ray diffraction patterns for different concentrations. It can be seen that there is a small increase in a and c. The c/a ratio for these compounds is about 0.81–0.82 which is in agreement with other values of 0.82–0.83 by Huang *et al.*[12] for $Sm(Co, Zr)_7$ alloys and our same values for $Sm(Co, Ti)_7$ alloys.[13] The unit cell volume V obtained from the lattice parameters a and c are listed in Table I. It can be seen from the table that there is a volume expansion by substituting Cu for Co; this expansion is due to the larger atomic volume of Cu, which is in agreement with our observations for other alloys.[20] Samples for magnetic anisotropy studies were prepared by mixing a fine powder of diameter <38 μm with 5-min epoxy on a glass sample holder and then aligning in a magnetic field of 20 kOe for about 1 h. The inset of Fig. 1 shows a typical x-ray diffraction pattern for the $SmCo_{6.7}Cu_{0.3}$ alloy. From this figure we see that the sample, after alignment, shows the (002) peak only indicating a uniaxial magnetocrystalline anisotropy. X-ray diffraction measurements on other aligned samples showed the same results. Magnetic measurements on aligned samples showed that the magnetization in the direction parallel to the aligning field is much higher than that along the direction perpendicular to the aligning field. Figure 2 shows a typical initial magnetization curve for $SmCo_{6.6}Cu_{0.4}$ measured at a temperature of 25 K using the SQUID magnetometer. This figure indicates that the sample is magnetically ordered. Magnetic measurements for other samples showed that all the samples studied are magnetically ordered and the magnetization depends on the Cu concentration. We find the saturation magnetization by using the law of approach to saturation, by plotting M versus $1/H$ and extrapolating M to $(1/H)=0$. The saturation magnetization values for the samples measured at 300 and 25 K are listed in Table I. It is clear from the table that the saturation magnetization decreases with increasing Cu concentration, (x), which is due to the replacement of magnetic element (Co) by a nonmagnetic element (Cu). The deviation of the dependence of M_s on x from linear dependence can be due to experimental error and/or the estimation of M_s by extrapolation. The magnetization as a function of temperature is measured with a VSM under an applied field of 3 kOe for all the samples. Table I also gives the dependence of T_C on Cu concentration. The Curie temperature increases with x reaching a maximum at $x=0.2$ (T_C

TABLE I. Lattice parameters *(a)* and *(c)*, unit cell volume (V), saturation magnetization (M_s) at $T=300$ and 25 K, and Curie temperature (T_c) of $SmCo_{7-x}Cu_x$ alloys as a function of copper concentration *(x)*.

X	a (Å)	C (Å)	V (Å^3)	M_s (emu/g) $T=300$ K	M_s (emu/g) $T=25$ K	T_c (°C)
0.0	4.935	4.010	84.576	102	103	770
0.1	4.967	4.003	85.538	85	86	850
0.2	4.968	4.060	85.606	84	84	852
0.3	4.974	4.060	85.817	82	83	828
0.4	4.975	4.009	85.939	71	72	769
0.5	4.978	4.010	86.045	61	63	758
0.7	4.981	4.011	86.159	57	58	760

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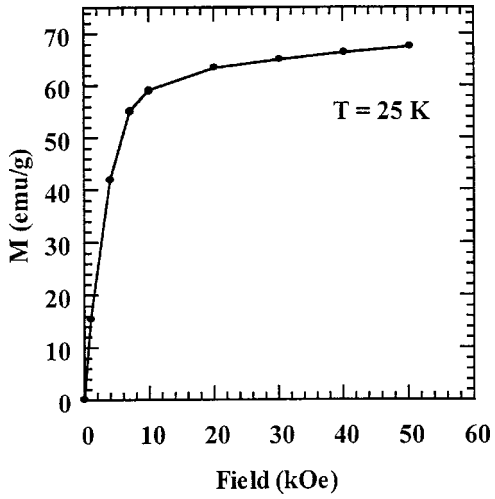

FIG. 2. Typical initial magnetization curve for a $SmCo_{6.6}Cu_{0.4}$ alloy measured at a temperature of 25 K.

$=852\,^{\circ}C$) then decreases with x as can be seen from the table. The maxima in T_C and c/a occur at the composition $x=0.2$. The nonmonotonic dependence of T_C and c/a on x remains to be understood. This maximum value for T_C is higher than that of the $SmCo_5$ compound ($T_C=750\,^{\circ}C$).

IV. CONCLUSIONS

Samples of the form of $SmCo_{7-x}Cu_x$ ($x=0$, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.7) have been prepared and studied. X-ray diffraction shows that these alloys form the hexagonal $TbCu_7$-type structure. We find that the hexagonal $TbCu_7$-type structure cannot be formed at large values of $x(x\geqslant 0.8)$. X-ray diffraction on magnetically aligned samples show that these samples have uniaxial anisotropy. The lattice parameters (a and c) are dependent on the Cu concentration. The unit cell volume is found to increase with x. We find that the saturation magnetization decreases with x at room temperature and at a temperature of 25 K. We find that Curie temperature increases with x reaching a peak at $x=0.2$ ($T_C=852\,^{\circ}C$) then decreases with x. These properties are promising for high temperature permanent-magnet applications.

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