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Magnetic properties of iron-rich mixed rare-earth $Sm_{2-x}Tb_xFe_{17-y}Si_y$ compounds

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A series of $Sm_{2-x}Tb_xFe_{17-y}Si_y$ solid solutions with $x=0$, 1, and 1.5 and $y=1$, 2, and 3 were prepared by induction melting stoichiometric amounts of high purity elements. The x-ray diffraction data confirm that the postannealed samples are 2:17 intermetallics of the $R\bar{3}m$ space group. The lattice parameters and the unit cell volumes were calculated using a modified Rietveld program. The fitted intensities showed behavior related to a disordered rhombohedral structure as inferred from neutron data. It was observed that for a particular y, the unit cell volume decreased almost linearly with increasing Tb content ($x=0$, 1, and 1.5). The unit cell volume decreases with increasing Si content (y) for a particular x as is also observed for the single rare-earth 2:17 compounds. The Curie temperature measurements show that with increasing Tb content x a decrease in the Curie temperatures of about 3% for $y=1$, 10% for $y=2$, and 15% for $y=3$ was observed. For a particular x, a nearly linear type increase in the Curie temperature resulted with increasing silicon content ($y\leqslant 3$). It was observed that with increasing x there was a decrease of about 40% in the magnetization for $y=1$ and 2, whereas there was a decrease of about 35% for $y=3$. For a particular x, the magnetization decreased almost linearly with increasing Si content ($y=1-3$). © *2002 American Institute of Physics.* [DOI: 10.1063/1.1451701]

I. INTRODUCTION

It is well established that the type of rare-earth element in R–T (R=rare earth, T=transition metal) intermetallics plays an important role in determining their magnetic properties. For example, the Curie temperature T_c of Gd_2Fe_{17} is approximately 80 °C higher than that of Sm_2Fe_{17}.[1] Partial substitution of the transition metal sublattice by nontransition metal elements can improve certain magnetic properties. For example, partial substitution of Fe by Si in several R–T intermetallic compounds leads to significant increases in T_c.[2,3] It was recently reported that the T_cs of some mixed Sm/Gd 2:17 structures were unusually high.[4] The purpose of this article is to present results from the investigation of crystallographic and magnetic properties of another mixed rare-earth system and the partial substitution of the Fe sublattice by Si in nominally 2:17 intermetallics.

II. EXPERIMENTAL METHOD

A series of $Sm_{2-x}Tb_xFe_{17-y}Si_y$ solid solutions with $x=0$, 1, and 1.5 and $y=1$, 2, and 3 were prepared by induction melting stoichiometric amounts of high purity elements. The samples were then annealed for 1 week at 1000 °C in an argon atmosphere. The x-ray diffraction (XRD) data using a SCINTAG diffractometer with Cu K_α radiation confirm that the postannealed samples are 2:17 intermetallics of the $R\bar{3}m$ space group with some alpha iron phase. The lattice parameters and the unit cell volumes were calculated using a modified RIETVELD program.[5] The thermomagnetic ($M-T$) behavior was investigated using a vibrating sample magnetometer (VSM) at the University of Missouri-Rolla to determine the T_c's for the samples. The saturation magnetization for the samples at 4 T was obtained using the VSM at the Laboratoire des Materiaux et du Genie Physique, France.

III. RESULTS AND DISCUSSION

The XRD data show that all the samples crystallized in the rhombohedral Th_2Zn_{17}-type structure.[1] XRD patterns indicate the presence of small amounts of alpha iron, which is assumed not to influence our data significantly. All peaks are indexed to the rhombohedral 2:17 phase $R\bar{3}m$, but, on the whole, the weak reflections that arise from the inequivalence of the 9*d* and 18*h* sites and from the regular stacking of rare-earth atoms and Fe dumbbells are reduced in their intensities compared to the lines originating from the $CaCu_5$ parent cell as shown in Fig. 1. This may be attributed to the disorder associated with the breaking of the perfect $a-b-c$ stacking of the c axes which leads, in turn, to a near equivalence of the 9*d* and 18*h* Fe sites as reported for the Nd/Dy system.[6] The detailed results of these studies will be published elsewhere. As has been observed for other $RFe_{17-y}Si_y$ intermetallics, the unit cell contracts with increasing Si content.[2] It was observed that for a particular y, the unit-cell volume decreased almost linearly as Sm was replaced with Tb. This is quite expected because of the smaller size of Tb compared to Sm. Cell volume as a function of the content of Tb and Si is shown in Figs. 2(a) and 2(b) respectively.

The saturation magnetization M_s at 300 K, taken to be the magnetization at 4 T in our case for varying Tb and Si

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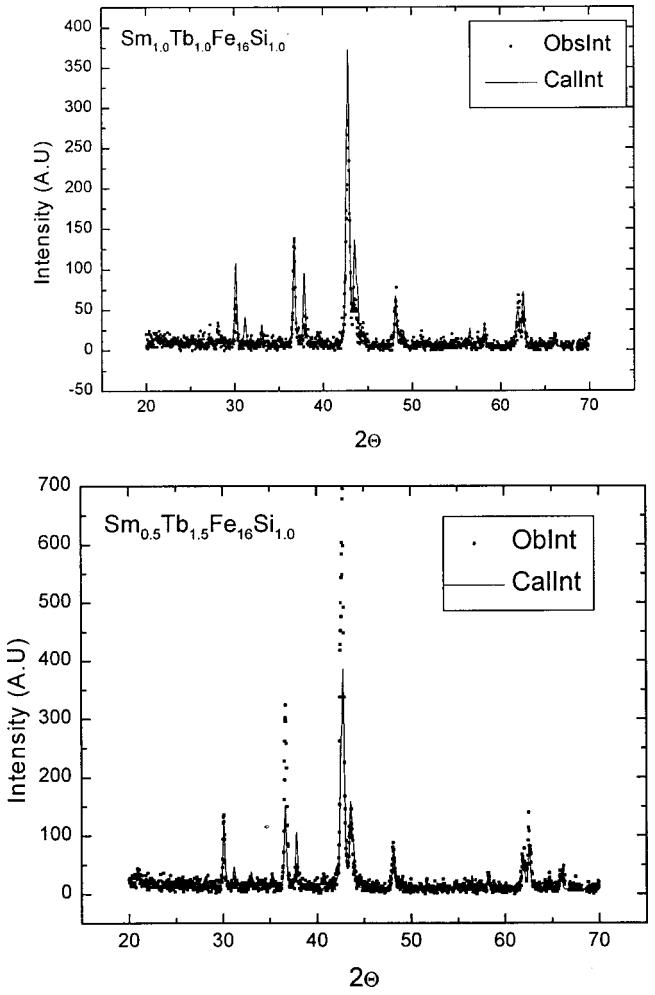

FIG. 1. XRD pattern and the calculated fit for $SmTbFe_{16}Si_1$ and $Sm_{0.5}Tb_{1.5}Fe_{16}Si_1$ showing the reduced intensities in the calculated fit.

contents, is shown in Figs. 3(a) and 3(b), respectively. It must be noted that the M_s values shown in Figs. 3(a) and 3(b) have not been corrected for the small amounts of impurities present in some of the samples. It was observed that for increasing x from $x=0$ to $x=1.5$ there is a decrease of about 40% in magnetization for $y=1$ and 2, whereas there is a decrease of about 35% for $y=3$. The main cause of this reduction is the antiferromagnetic coupling of the Tb with the Fe sublattices, which causes their contributions to partially cancel. For a particular x, the magnetization decreased almost linearly with increasing Si content.

The results of the thermomagnetic behavior of the Curie temperature T_c as a function of the content of Tb and Si are shown in Figs. 4(a) and 4(b), respectively. With increasing Tb content, a decrease in T_c of about 3% for $y=1$, 10% for $y=2$, and 15% for $y=3$ is observed. This is contrary to the effect without Si for which the Tb samples are known to have higher Curie temperatures than the corresponding Sm samples of the 2:17 type. The cause of this is at present unknown, but may be related to the combined effect of cell volume contraction from the Si and Tb simultaneously.[7] For a particular x, a nearly linear type increase in T_c is observed with increasing content of silicon ($y \leqslant 3$). No efforts have thus far been made to exceed $y=3$, but the present results

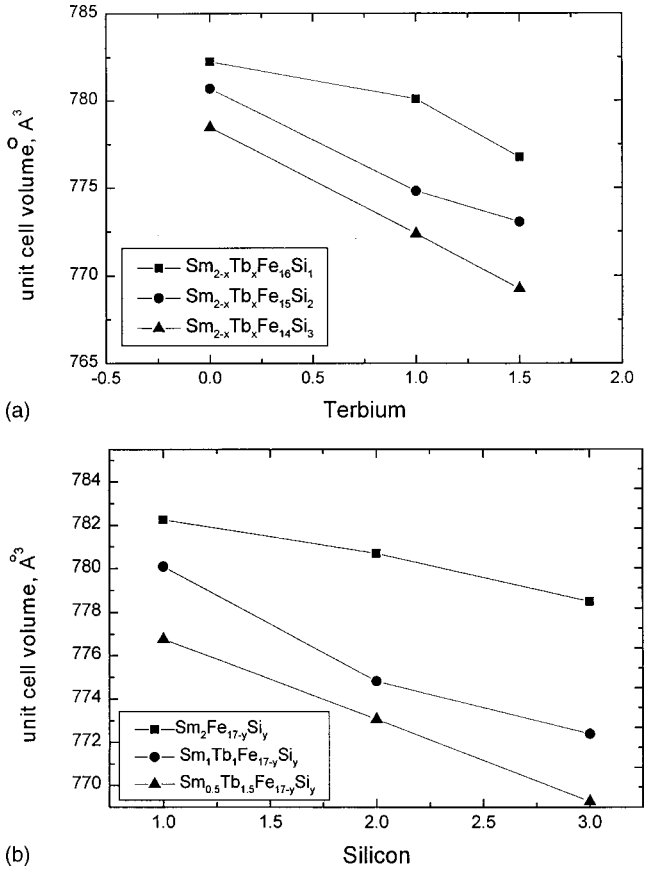

FIG. 2. (a) Cell volume vs terbium content and (b) cell volume vs silicon content.

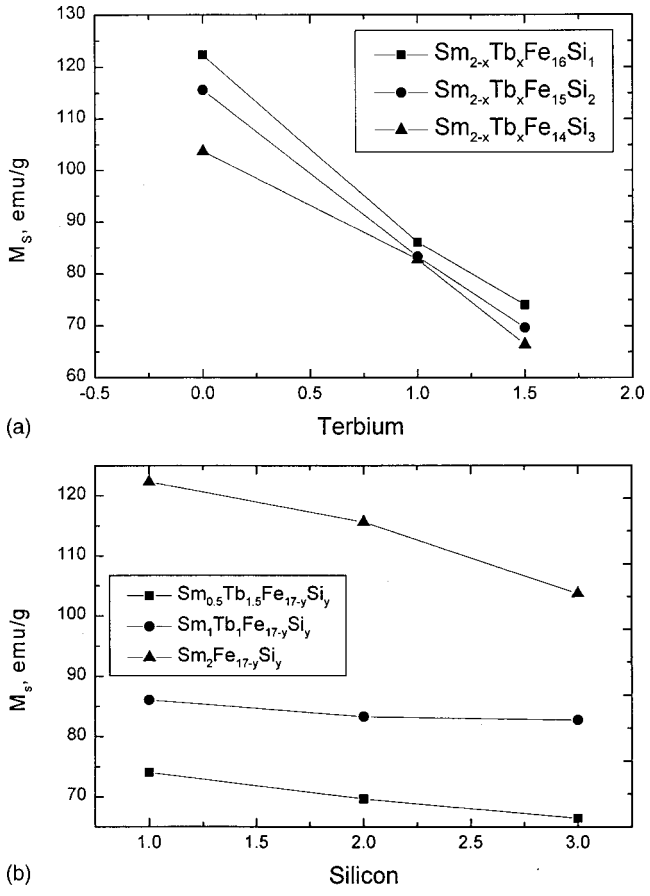

FIG. 3. (a) M_s vs terbium content and (b) M_s vs silicon content.

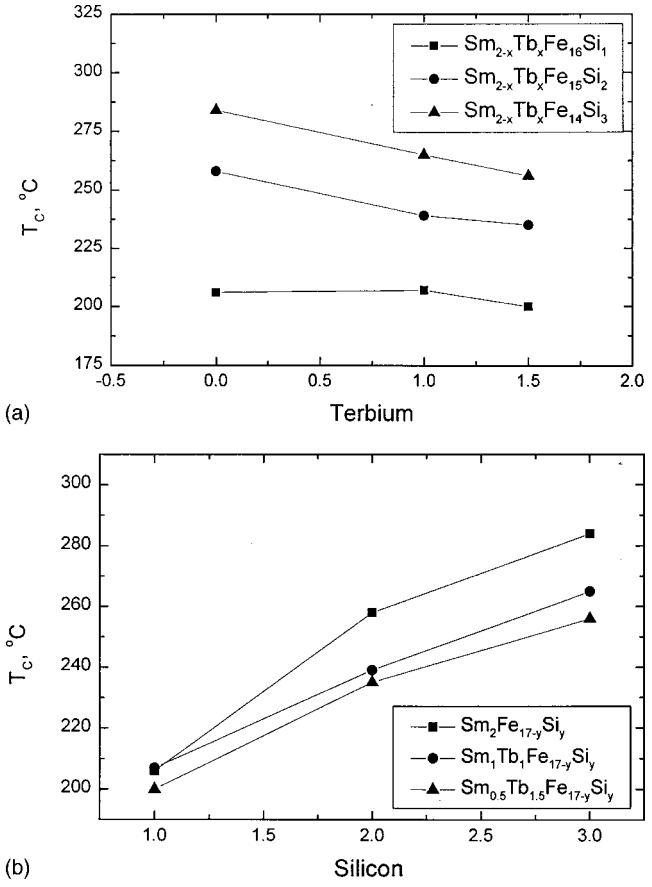

FIG. 4. (a) T_c vs terbium content and (b) T_c vs silicon content.

suggest that this may be useful even though $y=3$ appears to be the maximum concentration for a single, light rare earth. Table I compares the Curie temperatures for $Sm_2Fe_{17-x}Si_x$ $(x=1,2,3)$ with those from literature in parenthesis.[8]

IV. CONCLUSIONS

A behavior similar to that observed in the Nd/Dy system is likely exhibited by the Sm/Tb system in that the structure lies between the ordered rhombohedral phase formed with light rare earths and the hexagonal system formed with heavy rare earths. Like the Nd/Dy system studied with neutrons,[6] at certain values of x and y it shows a new disordered rhombohedral structure. Since this structure contains excess Fe dumbbells (compared to the 33% nominal value for the 2:17 phase) its properties may be a sensitive function of the starting composition. The present samples, at close to nominal stoichiometry, show the expected effects of Si and Tb on the cell volume and magnetization, but a somewhat surprising effect of Tb on the Curie temperature.[7] Further studies will investigate the effect of deviation from nominal stoichiometry on these properties.

TABLE I. The lattice parameters, unit cell volumes, M_s and T_c in $Sm_{2-x}Tb_xFe_{17-y}Si_y$.

Compound	a (Å)	c (Å)	V (Å^3)	M_s (emu/g)	T_c (°C)
$Sm_2Fe_{16}Si_1$	8.522	12.437	782.25	122.30	206 (202)
$Sm_2Fe_{15}Si_2$	8.492	12.490	780.10	115.60	258 (277)[a]
$Sm_2Fe_{14}Si_3$	8.487	12.449	776.76	103.70	284 (278)[a]
$SmTbFe_{16}Si_1$	8.488	12.529	780.70	86.03	207
$SmTbFe_{15}Si_2$	8.482	12.433	774.81	83.30	239
$SmTbFe_{14}Si_3$	8.466	12.451	773.06	82.71	265
$Sm_{0.5}Tb_{1.5}Fe_{16}Si_1$	8.469	12.530	778.48	74.03	200
$Sm_{0.5}Tb_{1.5}Fe_{15}Si_2$	8.457	12.467	772.38	69.64	235
$Sm_{0.5}Tb_{1.5}Fe_{14}Si_3$	8.454	12.426	769.26	66.41	256

[a]See Ref. 8.

ACKNOWLEDGMENTS

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