Magnetic properties and crystal structures of $Sm_yGd_{2-y}Fe_{17-x}Si_x$ alloys prepared by induction melting

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Abstract The magnetic properties and crystal structures of $Sm_yGd_{2-y}Fe_{17-x}Si_x$ alloys ($0 \le x \le 2$ and $y = 0\sim1.67$) have been investigated using x-ray diffraction and magnetic measurements. The $Sm_yGd_{2-y}Fe_{17-x}Si_x$ specimens were crystallized to the rhombohedral Th_2Zn_{17}-structure with less than 5 mol% of impurities. The unit cells of the mixed rare-earth samples are smaller than those of Sm_2Fe_{17} and Gd_2Fe_{17}. For example, the T_C of $SmGdFe_{17}$ ($255°C$) is approximately 160 and $80°C$ higher than that of Sm_2Fe_{17} and Gd_2Fe_{17}, respectively. The T_Cs measured for $Sm_yGd_{2-y}Fe_{17-x}Si_x$ samples, 280 to $290°C$, are among the highest values observed for a $R_2Fe_{17-x}M_x$ intermetallic where M is a substituent other than cobalt.

Key words Rare-earth transition metal intermetallics, Induction melting, Sm-Gd-Fe-Si alloy

1. Introduction

Among various properties a good candidate for a permanent magnetic material must have are; a) high energy product, b) large anisotropy field, and c) high Curie temperature (T_C) [1]. In addition, the commercial production of permanent magnets based on such a material must be economically feasible and all leading candidates belong to the family of rare earth-transition metal (R-T) intermetallics.

Unfortunately, existing theoretical models that describe the magnetic behavior of these materials are unable to predict the magnetic properties of a given intermetallic.

Consequently, researchers must synthesize and characterize R-T intermetallics of varying compositions in order to identify potential candidates. The present study is expected to contribute to the existing data based on the magnetic properties of these intermetallics. It has been observed that the magnetic properties of R_2Fe_{17}-based solid solutions can be modified by partially substituting the iron sublattice with certain other elements, such as Mn, Ga, Al [2-7].

The magnetic behavior of R-T intermetallics depends on the type of the rare-earth. For example, the T_C of Gd_2Fe_{17} is approximately $80°C$ higher than that of Sm_2Fe_{17} [1]. In addition, as has been observed for intermetallics such as $R_2Fe_{17-x}Si_x$ [2-4], partial substitution

of the iron sublattice by silicon can lead to remarkable improvements in certain magnetic properties [2]. Another process by which the magnetic properties of these materials, especially the R_2Fe_{17} intermetallics, can be improved is interstitial nitrogenation or carbiding [1]. In order to investigate the combined affects of mixing rare earths, partially substituting the iron sub-lattice, and insertion of interstitial atoms, the crystallographic and magnetic properties of $Sm_yGd_{2-y}Fe_{17-x}Si_x$ ($x = 0, 2$; $y = 0\sim1.67$) alloys were studied.

2. Experimental Procedure

The samples were synthesized as ingots by induction melting stoichiometric amounts of elements of purity 99.99 % or better in a copper cold boat. Sample com-

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Table 1
Lattice parameters and Curie temperatures of $Sm_yGd_{2-y}Fe_{17-x}Si_x$ intermetallic alloys

Sample		a (Å)	c (Å)	V (Å^3)	T_C (°C)
x = 0	y = 0.33	8.524	12.450	785.08	116
	y = 0.67	8.530	12.435	781.18	204
	y = 1	8.538	12.439	769.72	255
	y = 1.33	8.540	12.433	766.08	280
	y = 1.67	8.541	12.428	777.36	280
x = 2	y = 0.33	8.504	12.433	777.86	290
	y = 0.67	8.490	12.450	772.51	280
	y = 1	8.488	12.440	778.93	270
	y = 1.33	8.490	12.441	771.38	268
	y = 1.67	8.497	12.442	770.49	266

positions are shown in Table 1. The 10 wt% of excess amount of samarium was added and a weight allowance (ca. 3 %) was made for possible loss of samarium during melting. The ingot specimen was preared by induction melting process. The ingots were wrapped in tantalum foil and annealed at 950°C for 7 days under flowing Ar atmosphere. The phase purity was identified by X-ray diffraction (XRD) using a SCINTAG diffractometer with Cu-K_α radiation. The presence of samarium in these samples prevents the use of neutron diffraction techniques to determine the site occupancies. Consequently, the compositions of the samples listed in Table 1, especially the Sm : Gd and Fe : Si ratios, may be somewhat different from the actual compositions. The lattice parameters were obtained by Rietveld analysis of powder XRD patterns. The thermo-magnetic behavior for measuring the Curie temperature was investigated using a vibrating sample magnetometer with Faraday-type balance.

3. Results and Discussion

The XRD data suggest that all of the parent samples crystallized in the rhombohedral Th_2Zn_{17}-type structure with only small amount (<5 mol%) of impurities [1]. The impurity content increased with the silicon content. The most prominent impurity phases were α-iron and iron

silicide.

In order to investigated that the effect of silicon on the variation of crystallographic and magnetic properties of $Sm_yGd_{2-y}Fe_{17-x}Si_x$ alloys, XRD patterns and lattice parameters with composition (x = 0, 2 and y = 0.33~ 1.67) were presented and compared between two alloys systems in Fig. 1.

Table 1 also gives lattice parameters and unit cell volumes. It is interesting to note that the unit cell of $SmGdFe_{17}$ is smaller than that of Sm_2Fe_{17} and Gd_2Fe_{17}. However, in contrast to most $R_2Fe_{17-x}Si_x$ intermetallics for which the unit cell contracts with increasing silicon content [2, 3], the unit cell of $SmGdFe_{17-x}Si_x$ samples is larger than that of $SmGdFe_{17}$. In the case of $Sm_yGd_{2-y}Fe_{17-x}Si_x$ samples, however, the cell volume increases very slightly with increasing samarium content.

As shown in Table 1 of the magnetic properties, the Curie temperature of $SmGdFe_{17}$ is approximately 160 and 80°C higher than that of Sm_2Fe_{17} and Gd_2Fe_{17}, respectively. It appears that the Curie temperature of $Sm_yGd_{2-y}Fe_{17}$ intermetallics may vary through a maximum as the samarium content is increased. In contrast to other $R_2Fe_{17-x}Si_x$ intermetallics [2] partial substitution of iron by silicon in $SmGdFe_{17-x}Si_x$ does not have a significant effect on the Curie temperature. However, the T_C of the $SmGdFe_{17-x}Si_x$ intermetallics reported herein are among the highest values ever observed for a $R_2Fe_{17-x}M_x$ intermetallic (M : a substituent, other than cobalt).
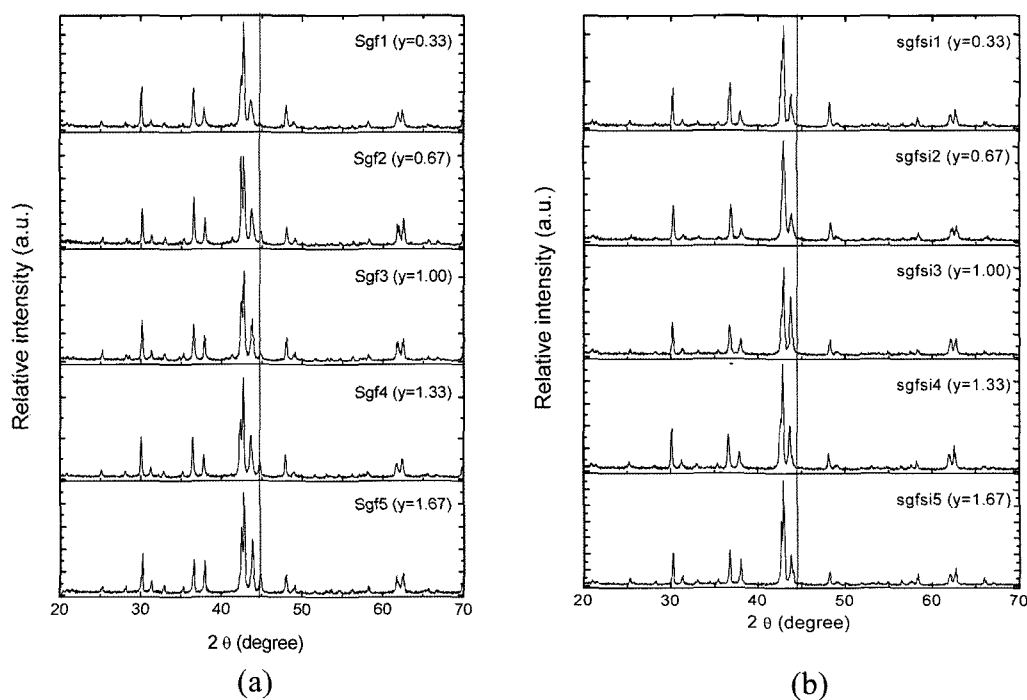

(a)   (b)

Fig. 1. XRD patterns of (a) $Sm_yGd_{2-y}Fe_{17}$ and (b) $Sm_yGd_{2-y}Fe_{15}Si_2$ intermetallic compounds (line as marked : α-Fe phase).
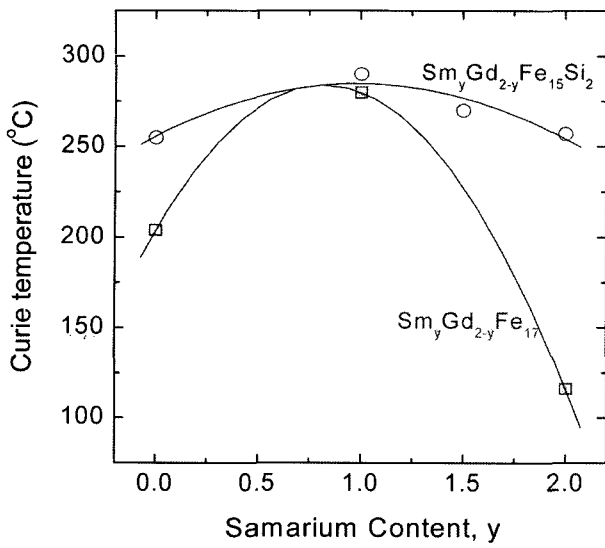
Fig. 2. Composition dependence of the Curie temperature of $Sm_yGd_{2-y}Fe_{17}$ and $Sm_yGd_{2-y}Fe_{15}Si_2$ with samarium content.

Note that the Curie temperatures of the $Sm_yGd_{2-y}Fe_{15}Si_2$ samples are higher than that of $Gd_2Fe_{15}Si_2$ and $Sm_2Fe_{15}Si_2$ [1]. In fact, the dependence of the Curie temperature of $Sm_yGd_{2-y}Fe_{15}Si_2$ on the samarium content is similar to that described above for $Sm_yGd_{2-y}Fe_{17}$ as shown in Fig. 2. In general, the addition of silicon increases the Curie temperature of $R_2Fe_{14}B$ which is an improved process to formulate better magnetic materials with higher

Curie temperatures [3].

The substitution of small amounts of silicon for iron increases the Curie temperature of $Sm_yGd_{2-y}Fe_{17}$. This can be explained from the previous works which the silicon reduces the extent of negative magnetic exchange interactions by preferentially replacing iron atoms on the sites in the $R_2Fe_{14}B$ structure which share the shorter iron-iron bond distances [2, 3].

The unit cell dimensions as a function of Sm content in $Sm_yGd_{2-y}Fe_{17}$ and $Sm_yGd_{2-y}Fe_{15}Si_2$ are shown in Fig. 3 and Fig. 4, respectively. The lattice and atomic positional parameters were used to calculate the compositional dependence of the Wigner-Seitz cell volumes in $Sm_yGd_{2-y}Fe_{17}$. The silicon affinity for a given transition metal site is correlated with the number of rare earth near neighbors for those site from the Wigner-Seitz cell configuration. The rare earth coordination of a transition metal site can be shared with its rare earth near neighbors, which is obtained from Wigner-Seitz cell calculations for each rare-earth transition metallic compounds [2-7]. As shown in Fig. 3 and Fig. 4, the lattice volume was decrease with samarium content in $Sm_yGd_{2-y}Fe_{17}$ but increased slightly in $Sm_yGd_{2-y}Fe_{15}Si_2$ structure. It can be revealed that the lattice parameter in c-axis is affected dominantly with composition dependence of silicon as modified $Sm_yGd_{2-y}Fe_{17}$.
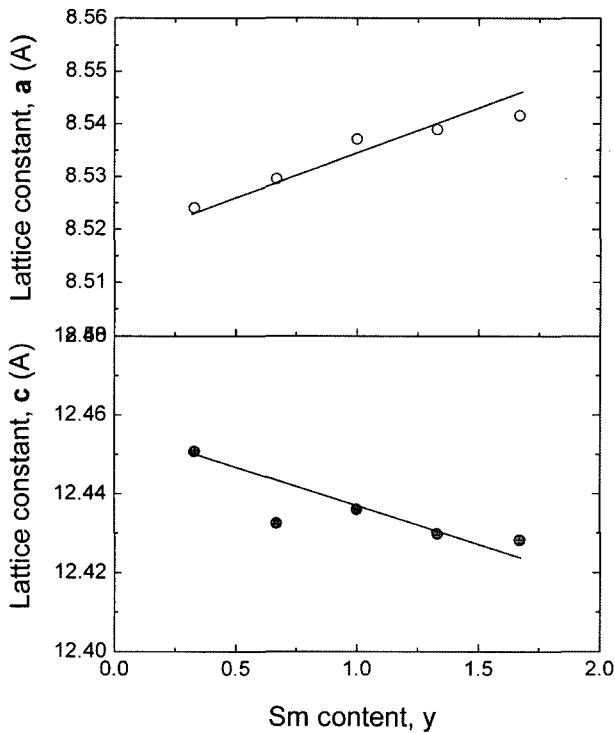

Fig. 3. The variation of lattice parameters of $Sm_yGd_{2-y}Fe_{17}$ with samarium content.

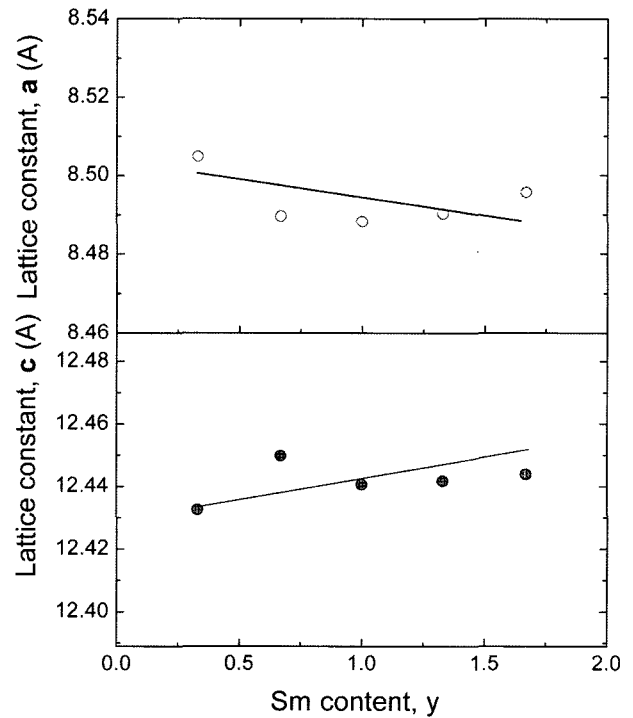
Fig. 4. The variation of lattice parameters of $Sm_yGd_{2-y}Fe_{15}Si_2$ with samarium content.

4. Conclusions

For a given silicon concentration, the Curie temperatures (T_C) of the mixed-rare earth $Sm_yGd_{2-y}Fe_{17-x}Si_x$ intermetallics are higher than those of the two end members of $x = 0$ and $x = 2$. Furthermore, the T_Cs measured for $Sm_yGd_{2-y}Fe_{17-x}Si_x$ samples are among the highest values ever observed for a $R_2Fe_{17-x}M_x$ intermetallic not containing cobalt. The resulting data of cell volume and Curie temperature indicates that there are distinct differences in the composition dependence of $Sm_yGd_{2-y}Fe_{17-x}Si_x$.

References

[1] K.H.J. Buschow, in Interstitial Intermetallic Alloys, edited by F. Grandjean, G.J. Long and K.H.J. Buschow (Kluwer Academic Publishers, Boston, 1995) pp. 349-409.

[2] G.K. Marasinghe, O.A. Pringle, G.J. Long, W.B. Yelon and F. Grandjean, "A neutron diffraction and Mössbauer spectral study of the structure and magnetic properties of the $Y_2Fe_{14-x}Si_xB$ solid solutions", J. Appl. Phys. 76 (1994) 2960.

[3] G.K. Marasinghe, O.A. Pringle, G.J. Long, W.J. James, D. Xie, J. Li, W.B. Yelon and F. Grandjean, "Neutron-diffraction and Mössbauer effect study of the preferential silicon site occupation and magnetic structure of $Nd_2Fe_{14-x}Si_xB$", J. Appl. Phys. 76 (1994) 2960.

[4] P.C. Ezekwenna, G.K. Marasinghe, W.J. James, O.A. Pringle, H. Luo, Z. Hu, Y.B. Yelon and Ph. l'Heritier, "Magnetic and structural properties of $Nd_2Fe_{17-x}Mn_x$ solid solutions", J. Appl. Phys. 81(8) (1997) 4533.

[5] P.C. Ezekwenna, G.K. Marasinghe, J.H. Nam, W.J. James, Y.B. Yelon, M. Ellouze and Ph. l'Heritier, "A magnetic and crystallographic study of $(Sm/Gd)_2Fe_{17-y}Si_yC_z$ solid solutions", J. Appl. Phys. 87(9) (2000) 6716.

[6] Z. Hu, W.B. Yelon, et al., "A magnetic, neutron diffraction, and Mössbauer spectral study of $Nd_2Fe_{15}Ga_2$ and $Tb_2Fe_{17-x}Ga_x$ solid solutions", J. Appl. Phys. 76 (1994) 443.

[7] W.B. Yelon, et al., "Neutron diffraction and Mössbauer effect study of several $Nd_2Fe_{17-x}Al_x$ solid solution", J. Appl. Phys. 73(10) (1993) 6029.