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## LETTER TO THE EDITOR

# Magnetic properties of gas-phase prepared $R_2Fe_{15}Si_2C_x$ ( $R=Sm$ and $Er$ ) compounds

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**Abstract.** We have discovered that Si is able to stabilize the structure of the 2:17-type carbides. The carbides,  $R_2Fe_{15}Si_2C_x$  ( $R=Sm$  and  $Er$ ), were made by heating powders of  $R_2Fe_{15}Si_2$  in  $CH_4$  at 700 °C. For  $Sm_2Fe_{15}Si_2C_{2.3}$ , the Curie temperature is 630 K and the anisotropy field is estimated to be over 70 kOe at room temperature; however, the saturation magnetization is relatively low, only about 88 emu  $g^{-1}$ , also at room temperature.

$R_2Fe_{17}$  intermetallic compounds are not useful as permanent magnets because of their low Curie temperatures and planar anisotropies at room temperature [1]. Many investigations have been made to modify their magnetic properties. In one method a partial substitution for Fe by a third element is made.  $R_2Fe_{15}Si_2$  compounds have elevated Curie temperatures [2–4].  $Sm_2Fe_{15}Ga_2$  has a uniaxial anisotropy [5] and an increase in the Curie temperature [5, 6]. Off-stoichiometric  $R_2Fe_{14}Si_2$  ( $R=Nd, Gd, Dy, Ho, Er$  and  $Y$ ) compounds, which have the  $R_2Fe_{17}$  crystal structure, possess a uniaxial anisotropy; however, their Curie temperatures, although higher than those of  $R_2Fe_{17}$ , are rather too low for use as permanent magnets. For example,  $Nd_2Fe_{14}Si_2$  has a Curie temperature, saturation magnetization and anisotropy field of 518 K, 128 emu  $g^{-1}$  and 50 kOe, respectively [7].

In another method, N or C is introduced interstitially either by a gas-phase reaction [8, 9] or by alloying [10] to form  $R_2Fe_{17}Z_x$  ( $Z=N$  or  $C$ ). These nitrides or carbides can be made by heating powders of the corresponding  $R_2Fe_{17}$  compounds in  $N_2$  or  $CH_4$  at 500 to 550 °C and have excellent intrinsic magnetic properties. The most striking ones are an increase of the Curie temperature by about 400 °C and, for  $Sm_2Fe_{17}N_{2.4}$ , a high uniaxial anisotropy with an anisotropy field 140 kOe at room temperature [8]. However, these nitrides or carbides have a major drawback, namely their instability at high temperatures. They will completely decompose into  $\alpha$ -Fe and RN or RC at 700 °C. Because of this drawback, it is difficult for the nitrides or carbides to be made into useful permanent magnets with high remanences, at least by normal techniques.

In this paper, it is reported that  $R_2Fe_{15}Si_2C_x$  ( $R=Sm$  and  $Er$ ) can be made by heating the powders of their parent,  $R_2Fe_{15}Si_2$ , in  $CH_4$  at 700 °C, instead of at 500 to 550 °C. These  $R_2Fe_{15}Si_2C_x$  compounds still retain the  $R_2Fe_{17}$  structure, even after annealing at 900 °C for 2 h. In particular,  $Sm_2Fe_{15}Si_2C_{2.3}$  has a high Curie temperature and a large uniaxial anisotropy; hence, this carbide may be a promising candidate for use as a permanent magnet.

$R_2Fe_{15}Si_2$  ( $R=Sm$  and  $Er$ ) compounds were prepared by arc-melting of better than 99.8% pure primary materials in a purified argon atmosphere. The ingots were annealed in an argon atmosphere at 900 °C for 4 h followed by quenching in water. X-ray diffraction showed that samples were single phase except for a small amount of  $\alpha$ -Fe for  $Sm_2Fe_{15}Si_2$ . The

$R_2Fe_{15}Si_2$  compounds were ground into fine powders (the size of the particles was smaller than  $20 \mu\text{m}$ ). Then, the powders were heated in  $CH_4$  at a temperature of  $700^\circ\text{C}$  for 2 h to form the carbides,  $R_2Fe_{15}Si_2C_x$ . The C concentrations were determined, by weighing, to be  $x = 2.3$  and  $2.5$  for the Sm and Er carbides, respectively.

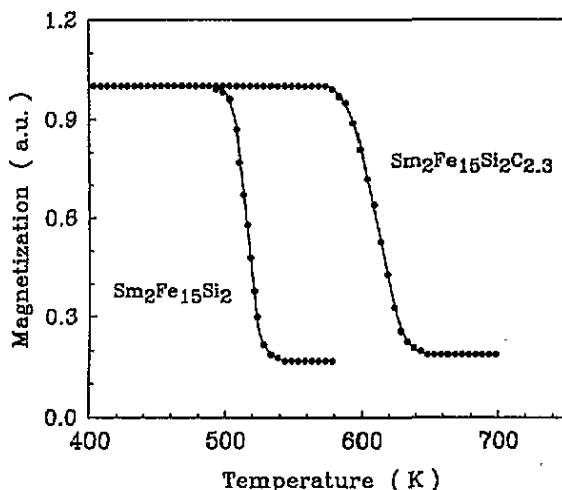
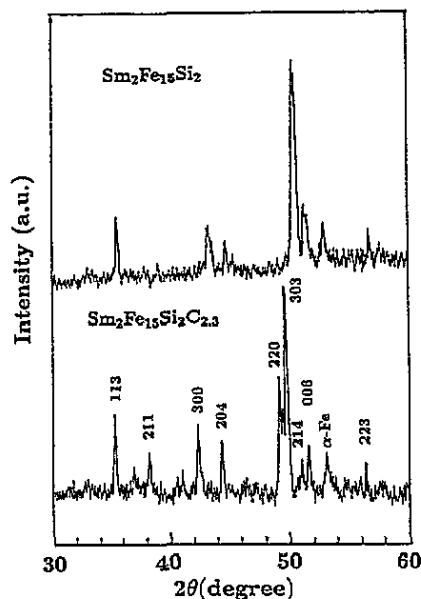


Figure 1. X-ray patterns for  $Sm_2Fe_{15}Si_2C_{2.3}$  and  $Sm_2Fe_{15}Si_2$ . Figure 2. Curie temperatures for  $Sm_2Fe_{15}Si_2C_{2.3}$  and  $Sm_2Fe_{15}Si_2$ .

X-ray diffraction using a diffractometer with  $Co K_\alpha$  radiation was employed to determine the crystal structure of  $R_2Fe_{15}Si_2C_x$  carbides. X-ray patterns of  $Sm_2Fe_{15}Si_2C_{2.3}$  and its parent are shown in figure 1 and lattice parameters are listed in table 1. The carbides retain their virginal crystal structure, namely the  $Th_2Zn_{17}$  structure for  $Sm_2Fe_{15}Si_2C_{2.3}$  and the  $Th_2Ni_{17}$  structure for  $Er_2Fe_{15}Si_2C_{2.5}$ . However, the lattice parameters,  $a$  and  $c$ , increase such that the volumes of the cells expand by 7.5% and 6.5% for  $Sm_2Fe_{15}Si_2C_{2.3}$  and  $Er_2Fe_{15}Si_2C_{2.5}$ , respectively, compared with their parents.

Table 1. Crystal structural and magnetic parameters for  $R_2Fe_{15}Si_2C_x$  ( $R=Sm$  and  $Er$ ) and their parents.

	$a$ (nm)	$c$ (nm)	$\Delta V/V$ (%)	$T_c$ (K)	$\sigma_s$ (emu $g^{-1}$ )	$H_a$ (kOe)	$H_{hf}$ (kOe)
$Sm_2Fe_{15}Si_2C_{2.3}$	0.875(2)	1.263(2)	7.5	630	88	> 70	186
$Sm_2Fe_{15}Si_2$	0.848(2)	1.250(3)		530	122		217
$Er_2Fe_{15}Si_2C_{2.5}$	0.862(1)	0.838(1)	6.5	620	86		174
$Er_2Fe_{15}Si_2$	0.842(2)	0.827(2)		465	77		205

Curie temperatures were determined by a thermomagnetic scan in a magnetic field of 0.5 kOe, as shown in figure 2 for  $Sm_2Fe_{15}Si_2C_{2.3}$  and its parent. The Curie temperatures are

630 K and 620 K for  $Sm_2Fe_{15}Si_2C_{2.3}$  and  $Er_2Fe_{15}Si_2C_{2.5}$ , respectively, which are 100 K and 150 K higher than those for their parents. The Curie temperature is primarily determined by the strength of the exchange interaction between Fe-Fe atom pairs, which is sensitive to the distance between them. The increase of Curie temperature is related to expansion of the cell volume.

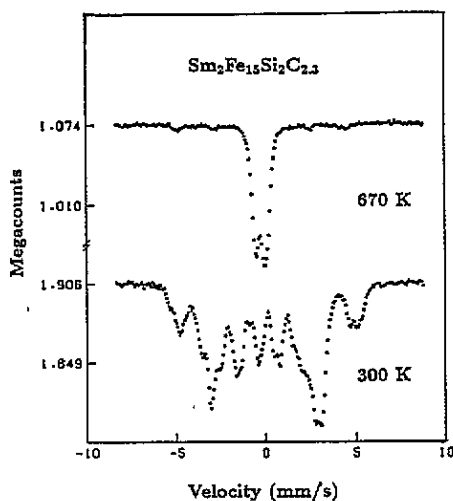


Figure 3. Mössbauer spectra of  $Sm_2Fe_{15}Si_2C_{2.3}$  at room temperature and 670 K.

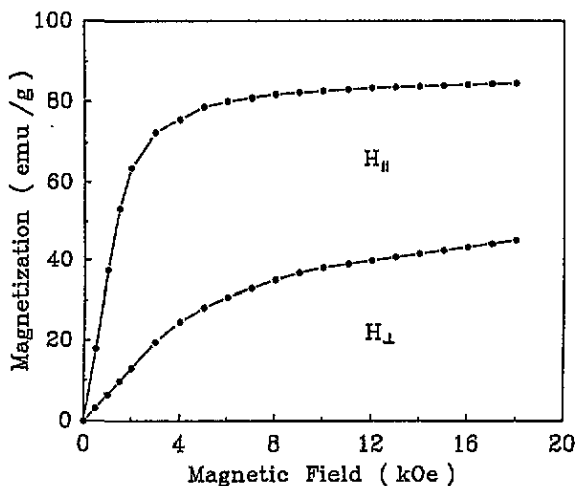


Figure 4. Magnetization curves of aligned  $Sm_2Fe_{15}Si_2C_{2.3}$  parallel and perpendicular to the aligned direction.

Magnetizations  $\sigma(H)$  were measured using a vibrating-sample magnetometer in applied fields up to 18 kOe at room temperature; the saturation magnetizations were found by fitting experimental  $\sigma(H)$  against  $1/H$  plots using the law of approach to saturation. The results are also listed in table 1. For  $Er_2Fe_{15}Si_2C_{2.5}$ , the saturation magnetization is  $86 \text{ emu g}^{-1}$ , which is 12% larger than  $77 \text{ emu g}^{-1}$  for its parent. However, for  $Sm_2Fe_{15}Si_2C_{2.3}$ , the saturation magnetization is  $88 \text{ emu g}^{-1}$ , which is less than that for its parent. This feature is different from nitride,  $Sm_2Fe_{15}Si_2N_x$ , for which the saturation magnetization is 4.3% larger than for its parent [11].

$^{57}Fe$  Mössbauer spectra were collected by a conventional constant-acceleration spectrometer. The Mössbauer spectra at room temperature and 670 K are shown in figure 3 for  $Sm_2Fe_{15}Si_2C_{2.3}$ . The average hyperfine fields are 186 and 174 kOe for the Sm and Er carbides, respectively, which are about 15% less than 217 and 205 kOe for their parents. If the proportional coefficient,  $145 \text{ kOe}/\mu_B$  [12], between the average hyperfine field and the average Fe moment is used, the Fe moment is estimated to be about 1.3 and  $1.2 \mu_B$  for the two carbides, respectively.

Aligned samples were prepared by mixing the fine powders with epoxy resin and then by placing the mixture in a magnetic field of 10 kOe. In order to determine the anisotropy field, magnetization curves with the field applied parallel and perpendicular to the alignment direction were obtained.  $Sm_2Fe_{15}Si_2$ ,  $Er_2Fe_{15}Si_2$  and  $Er_2Fe_{15}Si_2C_{2.5}$  have an easy planar anisotropy, whereas  $Sm_2Fe_{15}Si_2C_{2.3}$  exhibits a large uniaxial anisotropy, as shown in figure 4. The anisotropy field is estimated to be over 70 kOe as extrapolated from the maximum applied field of 18 kOe.

Most importantly, it is found that Si seems to take a key role on stabilizing structure of the 2:17-type of carbides. For  $\text{Sm}_2\text{Fe}_{17}$ , a large amount of  $\alpha$ -Fe develops when the heating temperature is above 550 °C in  $\text{CH}_4$  or in  $\text{N}_2$ ; the compound is completely decomposed into  $\alpha$ -Fe and SmC or SmN about 600–700 °C. However, for  $\text{Sm}_2\text{Fe}_{15}\text{Si}_2$ , only a small amount of  $\alpha$ -Fe is formed after heating up to a temperature of 700 °C in  $\text{CH}_4$ . A Mössbauer spectrum at  $T = 670$  K shows that the amount of  $\alpha$ -Fe is less than 5%. In addition, even though annealed at 900 °C for 2 h,  $\text{Sm}_2\text{Fe}_{15}\text{Si}_2\text{C}_{2.3}$  retains the  $\text{R}_2\text{Fe}_{17}$  crystal structure; however, the amount of  $\alpha$ -Fe increases. Thus,  $\text{Sm}_2\text{Fe}_{15}\text{Si}_2\text{C}_{2.3}$  may be a promising candidate as a permanent magnet material if its remanence can be raised.

It was found that Si is able to stabilize the structure of the 2:17-type carbides.  $\text{R}_2\text{Fe}_{15}\text{Si}_2\text{C}_x$  ( $R = \text{Sm}$  and  $\text{Er}$ ) was made by heating powder of  $\text{R}_2\text{Fe}_{15}\text{Si}_2$  in  $\text{CH}_4$  at 700 °C. These Si-containing carbides retain the crystal structure as same as their parents. The Curie temperatures, for the Sm and Er carbides, increase by 100 K and 150 K, respectively, compared with their parents. The anisotropy field for  $\text{Sm}_2\text{Fe}_{15}\text{Si}_2\text{C}_{2.3}$  is estimated to be over 70 kOe at room temperature. However, the saturation magnetization is relatively low, only about 88 emu  $\text{g}^{-1}$  at room temperature.

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