The formation and magnetic properties of $Sm_2Fe_{15}Al_2C_x$ (x=0-2.0) compounds prepared by arc melting

1994 J. Phys.: Condens. Matter 6 L185

(http://iopscience.iop.org/0953-8984/6/14/001)

J. Phys.: Condens. Matter 6 (1994) L185–L190. Printed in the UK

LETTER TO THE EDITOR

The formation and magnetic properties of $Sm_2Fe_{15}Al_2C_x$ ($x = 0$–2.0) compounds prepared by arc melting

Zhao-hua Cheng, Bao-gen Shen, Fang-wei Wang, Jun-xian Zhang, Hua-yang Gong and Jian-gao Zhao

State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

Received 24 January 1994

Abstract. The structural and magnetic properties of compounds in the series $Sm_2Fe_{15}Al_2C_x$ have been investigated by means of x-ray diffraction (XRD) and magnetization measurements. Samples with $x = 0$, 0.5, 1.0, 1.5, and 2.0 were prepared by the arc-melting method. XRD patterns indicate that these carbides are single phase with a rhombohedral Th_2Zn_{17}-type structure, except for $Sm_2Fe_{15}Al_2$, which contains a few per cent of BCC α-Fe. The saturation magnetization for all samples is about 110 emu g^{-1} and has a small dependence on C concentration. The Curie temperature T_C enhancement is about 190 K for $x = 1.5$ compared with that of Sm_2Fe_{17}. All compounds exhibit an easy c-axis anisotropy at room temperature and the anisotropy field increases to about 110 kOe when $x \leqslant 1.5$. The substitution of Al, like Ga and Si, is found to not only help the formation of high-C-concentration rare-earth–Fe compounds with 2:17-type structure, but also increase the anisotropy field.

The relatively low values of Curie temperature for R_2Fe_{17} compounds and the fact that none of them exhibits an easy-axis anisotropy at room temperature restrict the possible application of these materials as permanent magnets. Much work has been done towards understanding the means by which the Curie temperature of these materials may be increased and the methods by which a room-temperature uniaxial anisotropy may be induced. These investigations have been made by either substituting other elements into the R_2Fe_{17} structure or preparing materials with additional atoms located interstitially. Recently, it was found that introducing interstitial atoms, such as H [1], C [2], and N [3], into R_2Fe_{17} intermetallics has improved the hard-magnetic properties considerably. Unfortunately these nitrides and carbides prepared by the gas–solid reaction method have a poor thermal stability, and decompose into the equilibrium phase (RN)RC and α-Fe on heating to above 600 °C. Our previous work showed that the heavy rare-earth–Fe compounds $R_2Fe_{17}C_x$ with high C concentration ($x \simeq 3.0$) could be obtained by melt spinning, and they were stable at high temperature (above 1000 °C), while it was still difficult to synthesize $Sm_2Fe_{17}C_x$ with $x > 1.5$. Last year, it was discovered that the addition of Ga to $R_2Fe_{17}C_x$ compounds can facilitate the formation of rare-earth–Fe compounds with high C concentration [4, 5]. In this letter, we report the formation and magnetic properties of novel hard-magnetic $Sm_2Fe_{15}Al_2C_x$ ($x = 0$–2.0) compounds.

Fe and C were first melted together in an induction furnace to form Fe–C alloy with a lower melting temperature. Then Fe, Sm, Al and Fe–C alloys were arc melted in an Ar atmosphere of high purity. Elements used were at least 99.9% pure. An excess of 10–20% Sm was added to compensate for the evaporation during melting. The ingot alloys were

0953-8984/94/140185+06$19.50

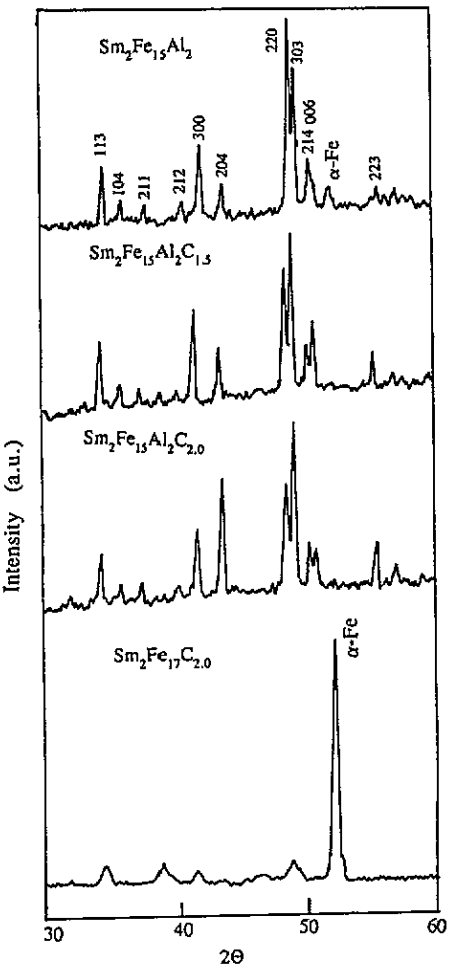

Figure 1. XRD patterns of $Sm_2Fe_{15}Al_2C_x$ ($x = 0$, 1.5, and 2.0) and $Sm_2Fe_{17}C_{2.0}$ compounds prepared by arc melting with Co Kα radiation.

melted at least four times to ensure homogeneity, then annealed under an Ar atmosphere at 1400 K for 5 d, followed by quenching into H_2O. Ingots were then ground to yield powders and oriented in an applied field of 20 kOe in an epoxy resin. X-ray diffraction (XRD) was performed on powder samples using Co Kα radiation to determine the phase structure as well as lattice constants and unit cell volume. The Curie temperatures were derived from the temperature dependence of magnetization $\sigma(T)$ curves measured by a vibrating sample magnetometer in a field of 700 Oe. The anisotropy fields were obtained by the magnetization curves measured along and perpendicular to the aligned direction using the extracting sample magnetometer with a magnetic field up to 70 kOe. The saturation magnetization at 300 K was obtained by fitting the experimental data of $M(H)$ versus H using the law of approach to saturation.

Figure 1 shows the typical XRD patterns of $Sm_2Fe_{15}Al_2C_x$ with $x = 0$–2.0. For comparison, that of $Sm_2Fe_{17}C_{2.0}$ is also presented. It can be seen from figure 1 that the carbides containing Al are single phase with a rhombohedral Th_2Zn_{17}-type structure except

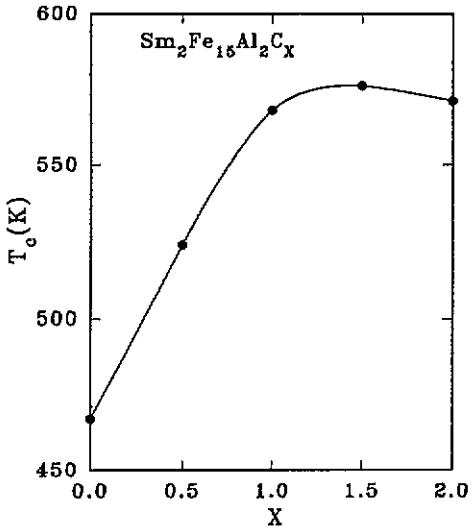

Figure 2. The Curie temperature of $Sm_2Fe_{15}Al_2C_x$ as a function of C concentration.

Table 1. The structural and magnetic parameters for Sm_2Fe_{17}, $Sm_2Fe_{17}C$ and $Sm_2Fe_{15}Al_2C_x$ compounds.

Compounds	a (Å)	c (Å)	v (Å^3)	T_C (K)	H_A (kOe)	M_s (emu g^{-1})	EMD
$x = 0$	8.613	12.478	802.2	467	—	—	c-axis
$x = 0.5$	8.652	12.491	809.8	524	80	110.3	c-axis
$x = 1.0$	8.674	12.506	814.0	568	108	112.3	c-axis
$x = 1.5$	8.680	12.520	817.0	576	111	110.2	c-axis
$x = 2.0$	8.682	12.580	821.0	571	102	112.6	c-axis
Sm_2Fe_{17} [9]	8.54	12.43	785	389	—	—	plane
$Sm_2Fe_{17}C$	8.644 [2]	12.476 [2]	807.0	552	53 [10]	—	c-axis

for $Sm_2Fe_{15}Al_2$, which contains a small amount of α-Fe. However, it is difficult to obtain a single-phase $Sm_2Fe_{17}C_x$ with $x > 1.0$ by arc melting. XRD patterns demonstrate that $Sm_2Fe_{17}C_{2.0}$ has a multi-phase structure with a predominant α-Fe phase. It is noteworthy that the addition of Al, like Ga and Si, to $R_2Fe_{17}C_x$ also plays an important role in the formation of the high-C rare-earth–Fe intermetallics with 2:17-type structure.

The lattice constants a and c and unit cell volume v of $Sm_2Fe_{15}Al_2C_x$ compounds with $x = 0$–2.0 are listed in table 1. For comparison, the values of Sm_2Fe_{17} and $Sm_2Fe_{17}C$ are also summarized. Both lattice constants and the unit cell volume are observed to increase on introducing interstitial C atoms. This result is similar to that for R_2Fe_{17} (C, N)$_x$ compounds. Comparing the unit cell volumes of $Sm_2Fe_{15}Al_2$ and $Sm_2Fe_{15}Al_2C$ with those of Sm_2Fe_{17} and $Sm_2Fe_{17}C$, it can be seen that the substitution of larger Al atoms for Fe in these materials results in an expansion of the unit cell volumes.

Figure 2 illustrates the Curie temperature T_C of these compounds as a function of C concentration. At lower C concentration, the increase of T_C is due to the substitution of larger Al atoms. For $x = 0$ and 1.5, T_C is about 80 and 190 K higher that of Sm_2Fe_{17}, respectively. The Curie temperature T_C enhancement in Al-substituted carbides is also observed in $R_2Fe_{17-x}Al_x$ compounds with R=Y, Ho [6], and Sm [7]. It is commonly assumed that the magnetic ordering temperature in rare-earth–Fe compounds is dominated

by the exchange interactions of the Fe sublattice, which is strongly dependent on the Fe–Fe interatomic distance. The generally low values of Curie temperature in R_2Fe_{17} compounds result from the relatively small Fe–Fe distance in these materials. Jacobs *et al* [6] have shown that the exchange coupling between Fe atoms in $R_2Fe_{17-x}Al_x$ increases strongly with Al substitution. The increase in Curie temperature corresponds to an increase in the positive Fe–Fe exchange coupling as a result of increasing interatomic distance.

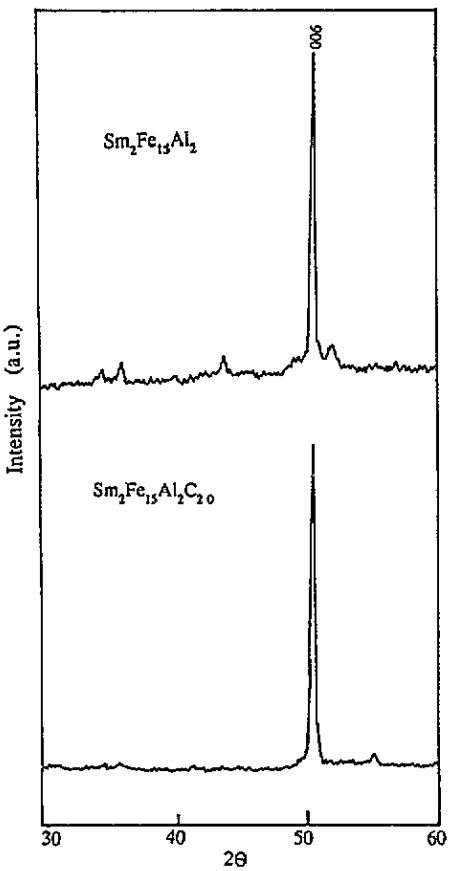

Figure 3. Room-temperature XRD patterns of $Sm_2Fe_{15}Al_2$ and $Sm_2Fe_{15}Al_2C_{2.0}$ aligned in an applied magnetic field of 20 kOe with Kα radiation.

The XRD studies of magnetic-field-oriented powders can provide information concerning the magnetocrystalline anisotropy. Figure 3 presents the room-temperature diffraction patterns of oriented $Sm_2Fe_{15}Al_2$ and $Sm_2Fe_{15}Al_2C_{2.0}$ powders. A uniaxial anistropy is observed for all compounds in the drastic increase in the (0, 0, 6) reflection and the disappearance of $(h, k, 0)$. In contrast to an earlier report [7] that the $Sm_2Fe_{15}Al_2$ compound exhibited a mixture of easy-plane and easy-axis behaviour, the present result clearly indicates that the sample with $x = 0$ exhibits an easy c-axis anisotropy. This fact suggests that its spin reorientation transition temperature is near room temperature. This study indicates that the addition of Al to Sm_2Fe_{17} compounds can develop a room-temperature uniaxial anisotropy without introducing interstitial N or C atoms.

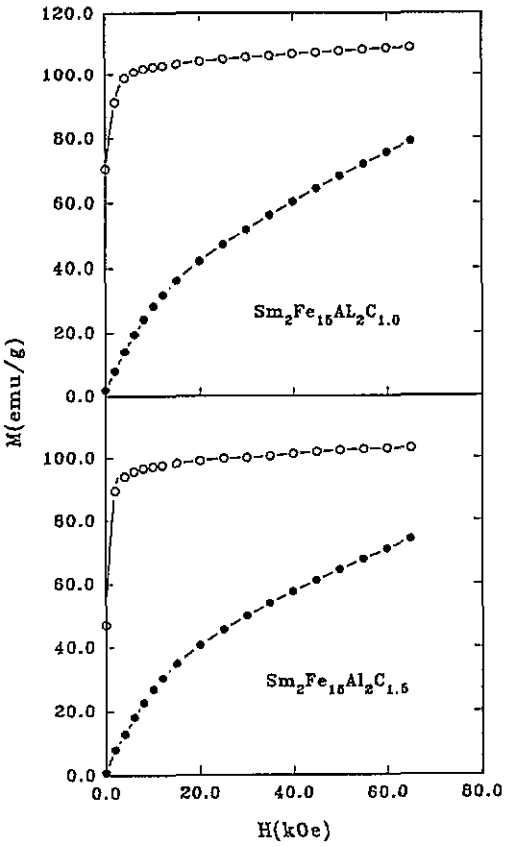

Figure 4. Magnetization curves of $Sm_2Fe_{15}Al_2C_x$ (x = 1.0 and 1.5) at 300 K along and perpendicular to the aligned direction (open and closed circles, respectively).

It is well known that the net anisotropy in rare-earth–Fe intermetallics is determined by the sum of the Fe sublattice and the rare-earth sublattice anisotropies. In R_2Fe_{17} compounds, the magnetization of the Fe sublattice exhibits a planar anisotropy. The rare-earth sublattice anisotropy can be approximately described by the product of the second-order Stevens coefficient α_j and the second-order crystal field parameter A_{20} on the basis of the crystal-field-induced single-ion model [8]. A negative product $\alpha_j A_{20}$ gives an uniaxial contribution from the rare-earth sublattice to the total anisotropy. In the case of Sm_2Fe_{17} compounds, $\alpha_j > 0$ and $A_{20} < 0$, the Sm^{3+} sublattice exhibits an uniaxial anisotropy. The substitution of Al for Fe increases the uniaxial anisotropy, suggesting an increase in the magnitude of the negative A_{20}, and, consequently, an increase in the anisotropy of the Sm sublattice.

Figure 4 shows an example of the magnetization curves measured along and perpendicular to the aligned direction at room temperature for the samples with $x = 1.0$ and 1.5. The anisotropy field H_A, estimated from the measurement curves, and saturation magnetization M_s of $Sm_2Fe_{15}Al_2C_x$ are summarized in table 1. The room-temperature anisotropy field of $Sm_2Fe_{15}Al_2C_x$ is found to increase with increasing C concentration when $x \leqslant 1.5$. For the samples with $x = 1.0$ and 1.5, the anisotropy field is about 110 kOe, which is much higher than that of $Sm_2Fe_{17}C$ ($H_A = 53$ kOe) and $Nd_2Fe_{14}B$ ($H_A = 80$ kOe), but the saturation magnetization is somewhat lower than that of Al-free compounds with the same C concentration. In order to improve the saturation magnetization, efforts to reduce

the Al concentration are in progress.

In summary, the $Sm_2Fe_{15}Al_2C_x$ compounds with $0 \leqslant x \leqslant 2.0$ can be prepared by arc melting. The addition of Al to these materials can not only facilitate the formation of the 2:17-type rare-earth–Fe compounds with high C concentration, but also improve the hard-magnetic properties. For the samples with $x = 1.0$ and 1.5, the Curie temperature is comparable with that of $Nd_2Fe_{14}B$, but the anisotropy field is much higher than that of $Nd_2Fe_{14}B$ and $Sm_2Fe_{17}C$. The present work suggests that $Sm_2(Fe, Al)_{17}C_x$ magnets with appropriate C concentration could be a starting material for sintering permanent magnets.

The authors wish to express their gratitude to T S Ning and M Hu for their assistance in XRD experiments. This work is supported by the National Natural Science Foundation of China.

References

[1] Wang X Z, Donnelly K, Coey J M D, Chevalier B, Etourneau J and Berlureau T 1988 *J. Mater. Sci.* **23** 329
[2] Zhong X P, Radwanski R J, de Boer F R, Jacobs T H and Buschow K H J 1990 *J. Magn. Magn. Mater.* **86** 333
[3] Coey J M D and Sun H 1990 *J. Magn. Magn. Mater.* **87** L251
[4] Shen B G, Kong L S, Wang F W and Cao L 1993 *Appl. Phys. Lett.* **63** 2288
[5] Shen B G, Wang F W, Kong L S, Cao L and Guo H Q 1993 *J. Magn. Magn. Mater.* **127** L267
[6] Jacobs T H, Buschow K H J, Zhou G F, Li X and de Boer F R 1992 *J. Magn. Magn. Mater.* **116** 220
[7] Wang Z and Dunlap R A 1993 *J. Phys.: Condens. Matter* **5** 2407
[8] Buschow K H J 1991 *Rep. Prog. Phys.* **54** 1123
[9] Sun H, Coey J M D, Otani Y and Hurley D P F 1990 *J. Phys.: Condens. Matter* **2** 6465
[10] Kou X C, Grossinger R, Jacobs T H and Buschow K H J 1990 *J. Magn. Magn. Mater.* **88** 1