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Magnetic hardening of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds

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Abstract

The structure and magnetic properties of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ ($x=2$, 3, 4 and 5) compounds prepared by arc-melting were studied. X-ray diffraction shows that these samples are single phase with rhombohedral Th_2Zn_{17}-type structure. The unit-cell volumes v of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds increase monotonically with increasing Ga concentration from 832.7 Å^3 for $x=2$ to 847.4 Å^3 for $x=5$. The Curie temperature T_C and room temperature saturation magnetization M_s are found to decrease monotonically with increasing Ga concentration. The $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds exhibit an easy c-axis anisotropy at room temperature. The anisotropy field is 135 kOe for $x=2$, and decreases to 76 kOe at $x=4$. A room temperature coercivity exceeding 13 kOe is obtained in as-quenched $Sm_2Fe_{17-x}Ga_xC_{2.5}$ ribbons with $x=2$ and $x=3$.

Keywords: Magnetic hardening; Arc-melting; Melt-spinning

1. Introduction

Since the discovery of the $Sm_2Fe_{17}N_y$ hard magnetic materials [1], a number of investigations on the structure and magnetic properties of interstitial rare-earth iron intermetallic compounds with the 2:17-type structure have been reported. Introducing interstitial atoms into R_2Fe_{17} compounds by gas–solid reactions [1–5] leads to a considerable enhancement of the Curie temperature and a modification of the magnetocrystalline anisotropy of Sm_2Fe_{17} from easy plane to easy c-axis. Unfortunately these nitrides and carbides prepared by gas–solid reaction have poor high temperature stability. Recently, Shen et al. have shown that the high-carbon rare-earth iron compounds with 2:17-type structure can be formed not only by melt-spinning [6], but also by the substitution of Ga, Al or Si for Fe [7,8]. The structure and magnetic properties of some arc-melted compounds of $R_2(Fe,M)_{17}C_y$ with M ≡ Ga, Al or Si have been studied. It was found that the $Sm_2Fe_{14}Ga_3C_y$ compounds with $y \geqslant 1.5$ have Curie temperatures higher than 600 K and room temperature anisotropy fields higher than 90 kOe [7]. A high coercivity of 15 kOe at room temperature was obtained in the $Sm_2(Fe,Ga)_{17}C_y$ compounds by melt-spinning [7]. In this paper we report the effects of the substitution of Ga for Fe on the structure and magnetic properties in $Sm_2Fe_{17}C_{2.5}$.

2. Experimental

The $Sm_2Fe_{17-x}Ga_xC_{2.5}$ samples with $x=2$, 3, 4 and 5 were prepared by arc-melting in an argon atmosphere of high purity. The raw materials Sm, Fe, Ga and an Fe–C alloy were at least 99.9% pure. The ingots were melted at least four times to ensure homogeneity. An excess of 4.5% Sm was added to compensate for the evaporation loss of Sm during melting. The heat treatment of the arc-melted ingots was performed in a steel tube in a highly purified argon atmosphere at 1273–1450 K for 12–96 h. In order to obtain a high coercivity, the ingots were melt spun in argon atmosphere on the outside of a single copper wheel rotating with surface velocities between 10 and 47 m s^{-1}. The as-quenched ribbons were about 1 mm wide and 20–30 μm thick. X-ray diffraction measurements on powder samples and alloy ribbons were performed using Co Kα radiation to identify the phase component and to determine the crystallographic structure. The room temperature saturation magnetization and coercivity were measured by an extracting sample magnetometer in fields up to 65 kOe. The Curie temperatures were determined from the temperature dependence of magnetization measured by a vibrating sample magnetometer in a magnetic field of 1 kOe. The aligned samples for anisotropy field measurements were prepared by mixing the powder with epoxy resin and then aligning in a magnetic field of 10 kOe. The anisotropy field was

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determined from magnetization curves measured along and perpendicular to the alignment direction by using the extracting sample magnetometer with a magnetic field of up to 65 kOe at room temperature.

3. Results and discussion

X-ray diffraction studies have shown that $Sm_2Fe_{17-x}Ga_xC_{2.5}$ samples with $x<2$ exhibit a multiphase structure with predominant α-Fe phase. For $x=2$, the arc-melted sample consists of the 2:17 phase and the α-Fe phase. However, high temperature annealing leads to formation of the 2:17-type single phase. When the Ga concentration x is larger than 3, the arc-melted $Sm_2Fe_{17-x}Ga_xC_{2.5}$ alloys are single phase with rhombohedral Th_2Zn_{17}-type structure. Fig. 1 shows X-ray diffraction patterns of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ with $x=2$ and $x=4$. As has been shown in other Ga-substituted rare-earth iron compounds [7,8], the partial substitution of Ga in $Sm_2Fe_{17}C_{2.5}$ helps the formation of the 2:17 phase. Table 1 summarizes the lattice constants a and c, and the unit-cell volumes v of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds with $2<x<5$. The substitution of Ga in $Sm_2Fe_{17}C_{2.5}$ leads to an increase in the unit-cell volume. An approximately linear dependence of the unit-cell volume on Ga concentration is observed, as shown in Fig. 2. The increase in cell volume per Ga atom is found to be about 4.9 Å^3.

The saturation magnetization M_s and Curie temperature T_C of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds are also shown in Fig. 2. The room temperature saturation magnetization is 97.8 emu g^{-1} for $x=2$ and it decreases monotonically with increasing Ga content to 52.1 emu g^{-1} at $x=5$. The T_C of $Sm_2Fe_{17-x}Ga_xC_2$ with $x=2$ is found to be 637 K, which is about 240 k higher than that of Sm_2Fe_{17} and about 80 K higher than that of $Sm_2Fe_{17}C$ [9]. However, further substitution of Ga decreases T_C. In general, in the Fe-rich rare-earth iron compounds the Curie temperature is mainly determined by the Fe–Fe exchange interactions, which are strongly dependent on interatomic distance. It has been shown previously that the introduction of interstitial carbon atoms or the partial substitution of Ga in Sm_2Fe_{17} led to a strong increase in the Curie temperature [4–10]. The enhancement of T_C is due mainly to lattice expansion. However, a decrease in the Curie temperature with increasing x in $Sm_2Fe_{17-x}Ga_xC_{2.5}$ is observed, although the substitution of Ga results in a monotonic increase in the unit-cell volume. This indicates that substitution of the larger Ga for Fe in rare-earth iron compounds with high carbon concentration decreases the overall Fe–Fe exchange interactions because the effect of a further increase in the Fe–Fe distance is overcompensated by a decrease in the number of Fe–Fe atom pairs, leading to a decrease in T_C.

The $Sm_2Fe_{17-x}Ga_xC_{2.5}$ samples with $x\leqslant 5$ are found to have a strong easy c-axis anisotropy at room temperature. This can be clearly seen from the X-ray diffraction patterns of magnetically aligned powder samples, as shown in Fig. 1

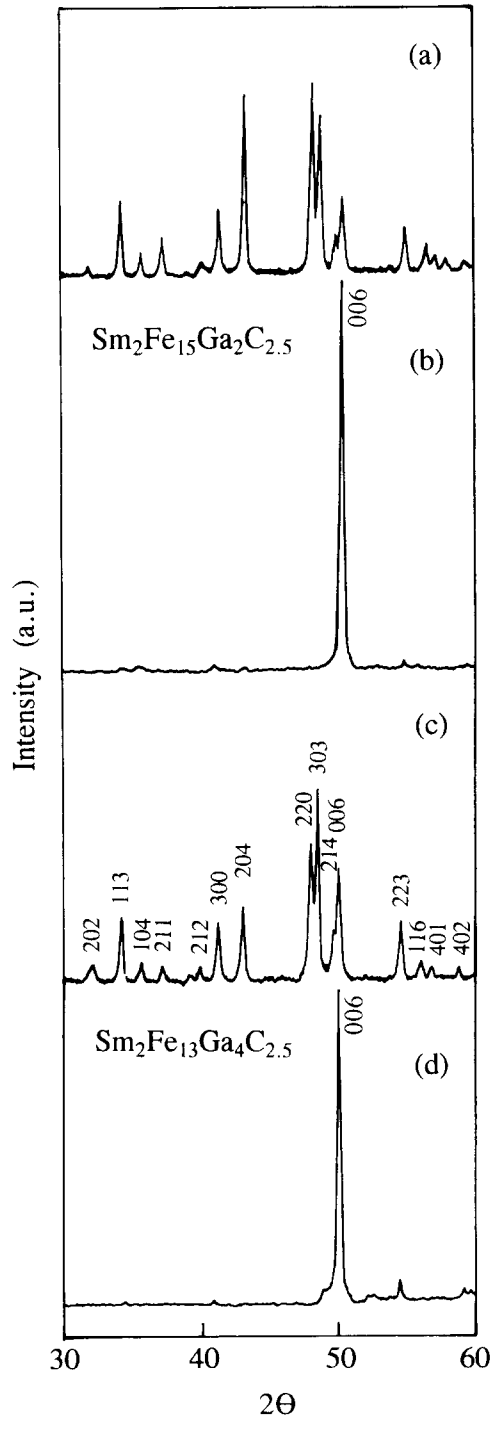

Fig. 1. X-ray diffraction patterns of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds with $x=2$ and $x=4$; (a) and (c) non-aligned samples, (b) and (d) aligned samples.

Table 1
Lattice parameters and unit-cell volumes of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds

Compound	a (Å)	c (Å)	v (Å^3)
$Sm_2Fe_{15}Ga_2C_{2.5}$	8.739	12.591	832.7
$Sm_2Fe_{14}Ga_3C_{2.5}$	8.752	12.624	837.4
$Sm_2Fe_{13}Ga_4C_{2.5}$	8.761	12.653	841.1
$Sm_2Fe_{12}Ga_5C_{2.5}$	8.790	12.664	847.4

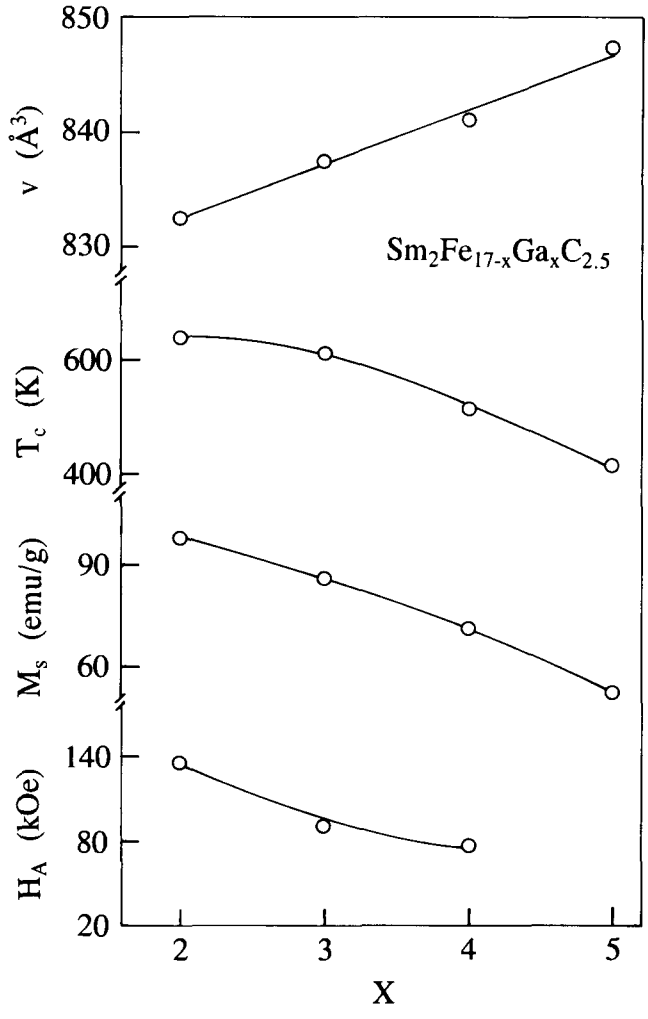

Fig. 2. Unit-cell volume v, Curie temperature T_C, saturation magnetization M_s and magnetocrystalline anisotropy field H_A at room temperature for $Sm_2Fe_{17-x}Ga_xC_{2.5}$ compounds as functions of Ga concentration.

for $Sm_2Fe_{17-x}Ga_xC_{2.5}$ with $x=2$ and $x=4$. The strong uniaxial magnetocrystalline anisotropy of the Sm-sublattice in $Sm_2Fe_{17-x}Ga_xC_{2.5}$ results not only from the addition of interstitial carbon atoms but also from the substitution of Ga. It was found previously that the substitution of Ga for Fe in Sm_2Fe_{17} leads to a change in the direction of easy magnetization from the basal plane to the c-axis at room temperature [10]. The addition of carbon atoms causes a more negative crystal field parameter A_{20} [11], accordingly increasing the anisotropy of the Sm-sublattice. Fig. 3 shows the magnetization curves of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ ($x=2$ and $x=4$) compounds measured along and perpendicular to the aligned directions at room temperature. The magnetocrystalline anisotropy field H_A estimated from magnetization curves is also shown in Fig. 2. The room temperature anisotropy field decreases with increasing Ga concentration. The H_A of the sample with $x=2$ is 135 kOe, which is 55 kOe higher than that of $Nd_2Fe_{14}B$.

The magnetic hardening of $Sm_2Fe_{17-x}Ga_xC_{2.5}$ was investigated by melt-spinning. As an example, Fig. 4 shows a room temperature hysteresis loop of as-quenched $Sm_2Fe_{14}Ga_3C_{2.5}$ ribbons. Room temperature coercivities exceeding 13 kOe are obtained in $Sm_2Fe_{17-x}Ga_xC_{2.5}$ ($x=2$ and $x=3$) ribbons prepared at quenching rates of 15–30 m s^{-1}. Quenching rates higher than 30 m s^{-1} lead to a rapid decrease in H_C. X-ray diffraction analysis shows that all diffraction lines in the pattern of the $Sm_2Fe_{17-x}Ga_xC_{2.5}$ ribbons prepared at velocities lower than 30 m s^{-1} can be indexed according to the Th_2Zn_{17} structure, indicating that these samples consist almost entirely of the hard magnetic phase. In an earlier study it was shown that a comparatively high coercivity ($H_C=4.6$ kOe) in $Sm_2Fe_{17}C_y$ could be attained only for high carbon concentrations [12]. However, the relatively high carbon concentration leads to formation of the α-Fe phase, which prevents the attainment of a high coercivity. Our studies indi-

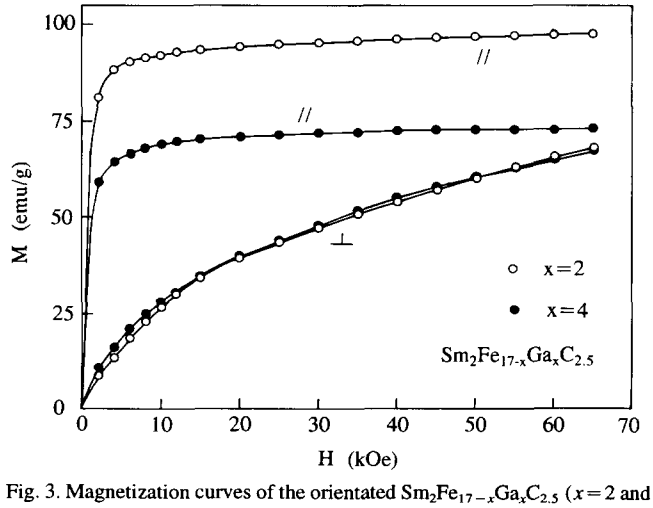

Fig. 3. Magnetization curves of the orientated $Sm_2Fe_{17-x}Ga_xC_{2.5}$ ($x=2$ and $x=4$) samples measured along and perpendicular to the aligned directions at room temperature.

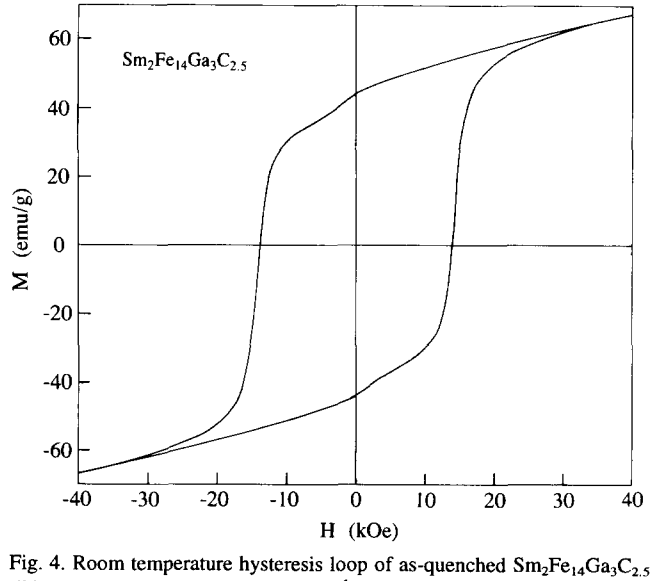

Fig. 4. Room temperature hysteresis loop of as-quenched $Sm_2Fe_{14}Ga_3C_{2.5}$ ribbons prepared at a speed of 20 m s^{-1}.

cate that the substitution of Ga in $Sm_2Fe_{17}C_y$ with high carbon concentration helps the formation of the 2:17-type structure and reduces the formation of the soft magnetic α-Fe phase. This, in consequence, results in a higher coercivity.

Acknowledgements

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