

# EFFECTS OF Al CONTENT ON STRUCTURAL STABILITY AND MAGNETIC PROPERTIES OF $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$ COMPOUNDS<sup>①</sup>

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**ABSTRACT** The magnetic properties of  $\text{Sm}_2\text{Fe}_{17-x}\text{Al}_x$  ( $x = 0, 1, 2, 3$ ) compounds and their hydrides are systematically investigated. The results reveal that all compounds have the  $\text{Th}_2\text{Zn}_{17}$ -type structure. As the content of Al increases, the Curie temperature of  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  compounds increases, and their saturation magnetization decreases. It is first discovered that the Curie temperatures of the hydrides of  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  compounds measured from heating process are higher than those measured from cooling process, and their difference is denoted as  $\Delta T_c^*$ . The correlation between  $\Delta T_c^*$  and the structural stability of  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  compounds as well as their corresponding hydrides is discussed.

**Key words**  $\text{Sm}_2\text{Fe}_{17}$  Al hydride Curie temperature structural stability

## 1 INTRODUCTION

It is well known that  $\text{Sm}_2\text{Fe}_{17}\text{N}_y$  compounds have good intrinsic magnetic properties<sup>[1]</sup>. Because of the poor stability of  $\text{Sm}_2\text{Fe}_{17}\text{N}_y$  at high temperatures, much attention has been paid to the studies of  $\text{Sm}_2(\text{Fe}, \text{M})_{17}$  ( $\text{M} = \text{Al}, \text{Ga}, \text{Si}, \text{Ti}, \text{V}, \text{Cr}, \text{Mn}, \text{Co}, \text{Ni}, \text{Zr}, \text{Nb}, \text{Mo}$ )<sup>[2-7]</sup> compounds. Partial substitution of Ga, Si, Al for Fe does not change the structure of  $\text{Th}_2\text{Zn}_{17}$ -type, but their anisotropy field has been changed from easy-plane to easy-axis. However, there are few reports on the effect of alloying elements on the structural stability of  $\text{Th}_2\text{Zn}_{17}$ -type compounds so far. There exists Miedama rule of reversible stability between compounds and their corresponding hydrides<sup>[8]</sup>. Therefore, the stability of the compounds can be determined by the stability of their corresponding hydrides.

In this paper, the effects of partial substitur-

tion of Fe by Al in  $\text{Sm}_2\text{Fe}_{17}$  compounds on the Curie temperature and the saturation magnetization of  $\text{Sm}_2\text{Fe}_{17}$  compounds and their hydrides at room temperature are reported.

## 2 EXPERIMENTAL

The purities of the raw materials used in the experiments are as follows:  $\text{Fe} \geq 99.95\%$ ,  $\text{Al} \geq 99.99\%$ ,  $\text{Sm} \geq 99.5\%$ . All samples are prepared by arc melting of a mixture of composing elements in a water-cooled copper crucible under mobile argon atmosphere. The mixture of stoichiometrically calculated elements was put into the crucible with rare earth at the lower layer, aluminum in the intermediate layer and iron on the upper layer. The samples were turned over and remelted three times for homogeneity.

All samples were subjected to a homogenization treatment under the condition of 1273 K, 48 h. The sample powder with diameter of about 30

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$\mu\text{m}$  was obtained by mechanical crushing and grinding. Then the powder was hydrogenized with a  $p$ - $c$ - $t$  measurement equipment. The hydrogen contents were determined by volume constant difference method.

In order to identify the phase composition and structural parameters of the compounds and their hydrides, X-ray diffraction analysis was carried out with  $\text{CuK}\alpha$  radiation.

The magnetization curves were determined by a vibration sample magnetometer at low field (0.04 T). To make a comparison, all the samples were heated to a same temperature at a same rate. The Curie temperature  $T_c$  was obtained from  $\sigma^2$ - $T$  curve, which was derived from  $\sigma$ - $t$  curve.

### 3 RESULTS AND DISCUSSION

X-ray diffraction results indicate that  $\text{Sm}_2\text{Fe}_{17-x}\text{Al}_x$  ( $x = 0, 1, 2, 3$ ) compounds are of  $\text{Th}_2\text{Zn}_{17}$ -type structure with space group  $R\bar{3}m$ . After hydrogenation treatment,  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}\text{H}_y$  hydrides have the same structure as their parent compounds. The only difference is that the unit cell volume of hydrides has expanded.

The thermomagnetization curves of the  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  and their hydrides  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}\text{H}_y$  are shown in Fig. 1. The heating rate adopted in all measurements is 10 K/min, and all the samples are heated to 653 K. The lattice parameters of  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  and their hydrides are listed in Table 1, the Curie temperatures and saturation magnetizations of the  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  and their hydrides are listed in Table 2. It can be seen from Fig. 1 and Table 2 that, with increasing Al content, the Curie temperatures of the compounds increase, but their saturation magnetizations at room temperature decrease.

After hydriding treatment, the Curie temperature of the hydrides is higher than that of the parent compounds. The increasing magnitude decreases with the increase of Al content.

In Fig. 1, an interesting and significant phenomenon is found, i.e. for the same hydrided sample, the  $\sigma$ - $T$  curves determined by heating and cooling processes are different. Table 2

indicates that for the same hydride, the Curie temperature obtained from heating process is higher than that obtained from cooling process. Its difference is denoted as  $\Delta T_c^*$ .  $\Delta T_c^*$  of hydrides with different Al contents are different, and  $\Delta T_c^*$  increases with increasing Al content in the compounds.

The experimental results of Miedema *et al.*<sup>[8]</sup> indicate that for a rare earth-transition metal compound, its stability and the stability of its corresponding hydride agree with the rule of irreversible stability, namely, the more stable the hydride is, the less stable the compound is.

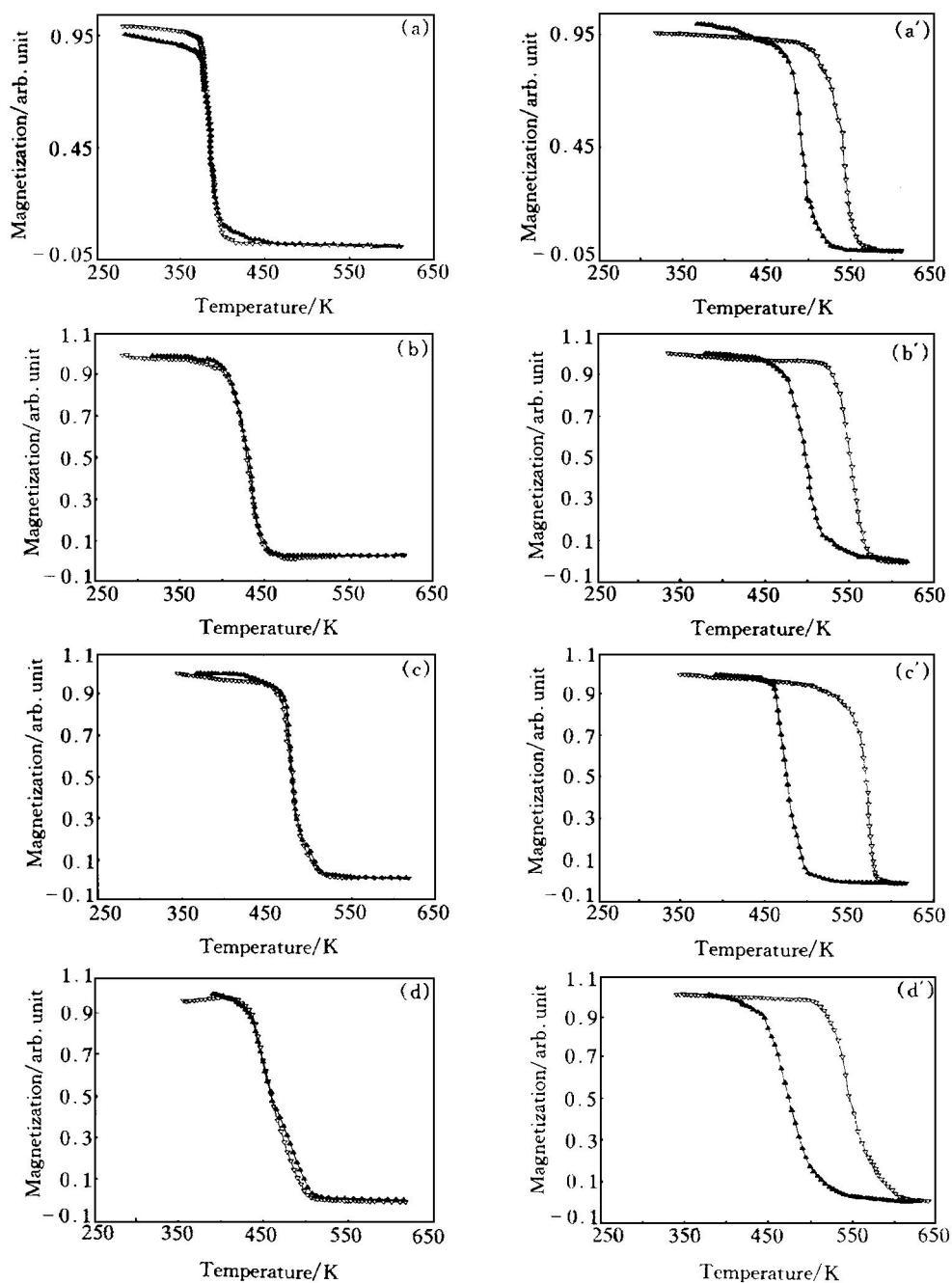
For  $\text{Sm}_2\text{Fe}_{17}$  compound, its first hydriding peak is in the range of 523 ~ 623 K<sup>[9]</sup>. As the temperature increases further, the hydride begins to release hydrogen. When the temperature reaches above 923 K, the compound begins to absorb hydrogen again. Therefore, the fact that the  $\sigma$ - $T$  curves obtained from heating process and cooling process do not coincide is ascribed to the variation of hydrogen content during the heating process. For a compound series with the same elements and crystal structure, the Curie temperature variation depends mainly on the quantity of each element.

For the hydrides of  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  compounds, only hydrogen content changes before and after heating during the  $\sigma$ - $T$  measurement process. During the cooling process, the hydrogen content is lower than that during heating process. This correlates with the stability of hydrides. The temperature at which the hydride is stable is an important parameter showing the stability of the hydride.

For  $\text{R}_2\text{Fe}_{17}\text{H}_y$  hydrides<sup>[10]</sup>, since their Curie temperatures increase with increasing hydrogen contents, the variation of Curie temperature reflect the variation of hydrogen content in the hydrides. The larger the variation of hydrogen content is, the worse the stability of the hydride is and vice versa.  $\Delta T_c^*$  is such a parameter that denotes Curie temperature difference between two hydrides with different hydrogen contents. The magnitude of  $\Delta T_c^*$  indicates the stability of the hydrides. For the hydrides of  $\text{Sm}_2\text{Fe}_{17-x}\text{Al}_x$ ,  $\Delta T_c^*$  increases as  $x$  increases.

It confirms that the stability of the hydrides of  $\text{Sm}_2(\text{Fe}, \text{Al})_{17}$  compounds decreases with

increasing Al content. According to the Miedema's rule of invariable stability, the stability of



**Fig.1** Thermomagnetic ( $M-T$ ) curves of  $\text{Sm}_2\text{Fe}_{17-x}\text{Al}_x$  ( $x=0, 1, 2, 3$ )

compounds (a-d) and their hydrides (a' - d') by VSM at 0.04 T

(a), (a') -  $x=0$ ; (b), (b') -  $x=1$ ; (c), (c') -  $x=2$ ; (d), (d') -  $x=3$

▽ - Heating; ▲ - Cooling

**Table 1** Lattice parameters of  $\text{Sm}_2\text{Fe}_{17-x}\text{Al}_x$  ( $x = 0, 1, 2, 3$ ) compounds and their hydrides

| Compounds  | $a/\text{\AA}$ | $c/\text{\AA}$ | $V/\text{\AA}^3$ | $\frac{\Delta V}{V}/\%$ |
|--|----------------|----------------|------------------|-------------------------|
| $\text{Sm}_2\text{Fe}_{17}$                          | 8.549          | 12.444         | 787.6            |                         |
| $\text{Sm}_2\text{Fe}_{17}\text{H}_{5.2}$            | 8.662          | 12.523         | 813.7            | 3.3                     |
| $\text{Sm}_2\text{Fe}_{16}\text{Al}$                 | 8.577          | 12.503         | 796.7            |                         |
| $\text{Sm}_2\text{Fe}_{16}\text{AlH}_{3.1}$          | 8.680          | 12.578         | 820.8            | 3.0                     |
| $\text{Sm}_2\text{Fe}_{15}\text{Al}_2$               | 8.612          | 12.538         | 805.2            |                         |
| $\text{Sm}_2\text{Fe}_{15}\text{Al}_2\text{H}_{2.8}$ | 8.698          | 12.600         | 825.5            | 2.5                     |
| $\text{Sm}_2\text{Fe}_{14}\text{Al}_3$               | 8.635          | 12.548         | 810.3            |                         |
| $\text{Sm}_2\text{Fe}_{14}\text{Al}_3\text{H}_{2.6}$ | 8.704          | 12.598         | 826.6            | 2.0                     |

**Table 2** Curie temperatures and saturation magnetizations of  $\text{Sm}_2\text{Fe}_{17-x}\text{Al}_x$  ( $x = 0, 1, 2, 3$ ) compounds and their hydrides at room temperature

| Compounds  | $T_c/\text{K}$ | $\Delta T_c/\text{K}$ | $\Delta T_c^*/\text{K}$ | $M_s(300\text{ K})/(\text{Am}^2\cdot\text{kg}^{-1})$ |
|--|----------------|-----------------------|-------------------------|--|
| $\text{Sm}_2\text{Fe}_{17}$                          | 398            |                       | -                       | 118.0  |
| $\text{Sm}_2\text{Fe}_{17}\text{H}_{5.2}$            | 554            | 156                   |                         | 149.6  |
| $\text{Sm}_2\text{Fe}_{17}\text{H}_z^*$              | 509            | 111                   | 45                      |  |
| $\text{Sm}_2\text{Fe}_{16}\text{Al}$                 | 449            |                       | -                       | 112.3  |
| $\text{Sm}_2\text{Fe}_{16}\text{AlH}_{3.1}$          | 571            | 122                   |                         | 139.1  |
| $\text{Sm}_2\text{Fe}_{16}\text{Al}_1\text{H}_z^*$   | 511            | 62                    | 60                      |  |
| $\text{Sm}_2\text{Fe}_{15}\text{Al}_2$               | 488            |                       | -                       | 103.6  |
| $\text{Sm}_2\text{Fe}_{15}\text{Al}_2\text{H}_{2.8}$ | 574            | 86                    |                         | 137.4  |
| $\text{Sm}_2\text{Fe}_{15}\text{Al}_2\text{H}_z^*$   | 502            | 14                    | 72                      |  |
| $\text{Sm}_2\text{Fe}_{14}\text{Al}_3$               | 498            |                       | -                       | 95.4   |
| $\text{Sm}_2\text{Fe}_{14}\text{Al}_3\text{H}_{2.6}$ | 580            | 82                    |                         | 123.3  |
| $\text{Sm}_2\text{Fe}_{14}\text{Al}_3\text{H}_z^*$   | 501            | 3                     | 79                      |  |

The superscript “\*” indicates the hydrides after the  $\alpha T$  measurement by VSM.

$\text{Sm}_2\text{Fe}_{17-x}\text{Al}_x$  compounds increases with increasing aluminum content. Therefore, aluminum is an element of stabilizing effect for  $\text{Th}_2\text{Zn}_{17}$ -type structure.

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