

Structural and magnetic properties of (Sm , Pr) 3 (Fe 1 - x Al x) 27.5 Ti 1.5 [x = 0.1 , 0.2 , 0.3]

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Structural and magnetic properties of $(\text{Sm}, \text{Pr})_3(\text{Fe}_{1-x}\text{Al}_x)_{27.5}\text{Ti}_{1.5}$ [$x=0.1, 0.2, 0.3$]

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The structural and magnetic properties of $(\text{Sm}_{0.9}\text{Pr}_{0.1})_3(\text{Fe}_{1-x}\text{Al}_x)_{27.5}\text{Ti}_{1.5}$ [$x=0.1, 0.2, 0.3$] compounds have been investigated by x-ray diffraction, magnetization, and Curie temperature measurements. The lattice parameters are found to increase with increasing Al concentration. The saturation magnetization is seen to decrease with the substitution of Al and is attributed to the modification in the DOS by Al. The Curie temperature increased for low concentrations of Al and then decreased. The easy direction of magnetization (EMD) in all the compounds is normal to the (204) plane. The anisotropy fields decrease with increasing Al concentration. © 2005 American Institute of Physics. [DOI: 10.1063/1.1851880]

INTRODUCTION

The discovery of $\text{Nd}_3(\text{Fe}, \text{Ti})_{29}$ compounds has opened up a novel class rare earth (R)-iron compounds, namely $\text{R}_3(\text{Fe}, \text{M})_{29}$ (3:29) compounds¹⁻³ as potential permanent magnet materials. These compounds have been reported to form in monoclinic structure with $A2/m$ space group,⁴ a combination of alternate stacking of $\text{Th}_2\text{Zn}_{17}$ (2:17) and ThMn_{12} (1:12) type segments with two inequivalent sites for R atoms and 11 inequivalent sites for Fe/M atoms.⁴⁻⁶ Li *et al.*⁶ have reported that the dominant crystal field parameter A_2^0 which determines the R anisotropy has opposite signs at the two inequivalent sites of R atoms. At the 2a site with 1:12 like environment, $A_2^0 > 0$ and at the 4i site with 2:17 like environment, $A_2^0 < 0$. The product of second order Steven's coefficient (α_J) and A_2^0 should be positive for axial anisotropy. Shah *et al.*⁷ have investigated the magnetic properties of $(\text{Sm}_{1-x}\text{Pr}_x)_3\text{Fe}_{27.5}\text{Ti}_{1.5}$ compounds and showed that Sm ($\alpha_J > 0$) occupies the 2a site while Pr ($\alpha_J < 0$) occupies the 4i site. Sheloudko *et al.*⁸ have reported preferential occupancy of Pr and Nd at 4i sites in $\text{Pr}_2\text{Er}(\text{Fe}, \text{Ti})_{29}$ and $\text{SmNd}_2(\text{Fe}, \text{Ti})_{29}$ respectively. Substitution of Al has been reported to increase Curie temperature and uniaxial anisotropy in 2:17 compounds.⁹ In this paper, the investigation of the role of Al on the structural and magnetic properties of $(\text{Sm}_{0.9}\text{Pr}_{0.1})_3\text{Fe}_{27.5}\text{Ti}_{1.5}$ compounds is presented.

EXPERIMENTAL DETAILS

All the compounds were prepared by arc melting Sm and Pr of 99.9% purity, Fe of 99.95% purity and Al and Ti of 99.99% purity. The melting was done several times to ensure homogeneous mixing of the constituents. About 12% of Sm has been added in excess to compensate for the loss of Sm during melting. Each ingot was wrapped in a Ta foil and sealed in an evacuated quartz tube and homogenized at 1100 °C for 7 days and then quenched in ice water mixture. Structural characterization was done using powder x-ray diffraction patterns taken employing Fe-K_α radiation. Magnetization and Curie temperature measurements were carried out

using a vibrating sample magnetometer (PAR-Model 155). Anisotropy fields (H_A) were determined from the magnetization curves of magnetically aligned samples (parallel to the plane of the sample geometry, arrested in 5 min epoxy resin) using the singular point detection method (SPD). Easy magnetization direction (EMD) was determined from the XRD patterns of magnetically aligned samples (perpendicular to the substrate).

RESULTS AND DISCUSSION

X-ray diffraction patterns of the compounds are shown in Fig. 1(a). All the compounds have formed the 3:29 phase as the major phase and small amounts of 1:12 phase and traces of α -Fe. The lattice parameters are listed in Table I. The lattice is seen to expand with Al concentration with β remaining the same. This increase is in accordance with the atomic radii of Fe and Al, as reported in 2:17 compounds.^{9,10}

Figure 2 shows magnetization curves at room temperature where, the saturation field decreases with increasing Al concentration. The saturation magnetization values (M_S)

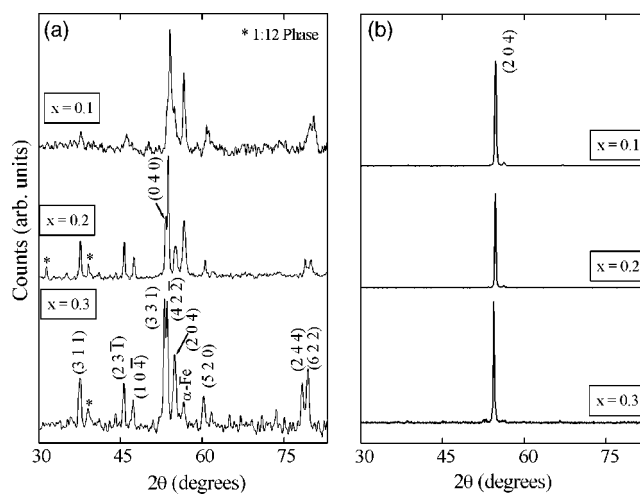


FIG. 1. XRD patterns of (a) random and (b) aligned powders of $(\text{Sm}_{0.9}\text{Pr}_{0.1})_3(\text{Fe}_{1-x}\text{Al}_x)_{27.5}\text{Ti}_{1.5}$.

TABLE I. Lattice parameters, room temperature magnetization, anisotropy field, and Curie temperature of $(\text{Sm}_{0.9}\text{Pr}_{0.1})_3(\text{Fe}_{1-x}\text{Al}_x)_{27.5}\text{Ti}_{1.5}$ compounds.

x	0 ^a	0.1	0.2	0.3
a (Å)	10.635	10.642	10.650	10.668
b (Å)	8.568	8.584	8.586	8.583
c (Å)	9.736	9.750	9.782	9.797
β (deg)	96.96	96.95	96.93	96.93
V (Å) ³	880	884	888	891
M_s (emu/g)	...	108	89	57
T_C (K)	474	458	466	444
H_A (kOe)	...	17	15	14

^aInterpolated from the data in Ref. 7.

were determined from the Honda plots and are shown in Table I. The Curie temperature (T_C) was determined through the magnetization measurements carried out at a field of 50 Oe and the T_C increases up to $x=2$ and then decreases.

The effect of Al substitution on the magnetization is discussed based on the reports on 2:17 compounds. The decrease in the magnetization has been explained as due to the modification in the DOS of the 3d band of Fe due to the 3p band of Al, situated at the bottom of the 3d band.¹¹ The spin-up band of Fe may be said to be nearly completely filled, as in the case of 2:17 systems, leading to a decrease in the magnetization with Al due to the charge transfer from the 3p band of Al to the 3d band of Fe. However, the rate of decrease in the present case is more than that in the 2:17 system. Therefore, the spin up band in this case may be filled more than in the 2:17 systems.

The Curie temperature is seen to increase from 458 K for $x=0.1$ to 466 K for $x=0.2$ and then to decrease to 444 K for $x=0.3$. Suresh *et al.*^{9,12} have reported an increase of T_C of ErPrFe_{17} from 290 K to 435 K and subsequent decrease with Al substitution. The higher T_C in the 3:29 system than in the 2:17 system may be due to the fact that 3:29 system contains more Fe than the 2:17 system. From the above comparisons, it seems likely that there is a critical Al to R ratio for which T_C reaches a maximum value.

X-ray diffraction patterns for the magnetically aligned samples are shown in Fig. 1(b). The reflections from only (204) planes are seen in the pattern. The easy magnetization

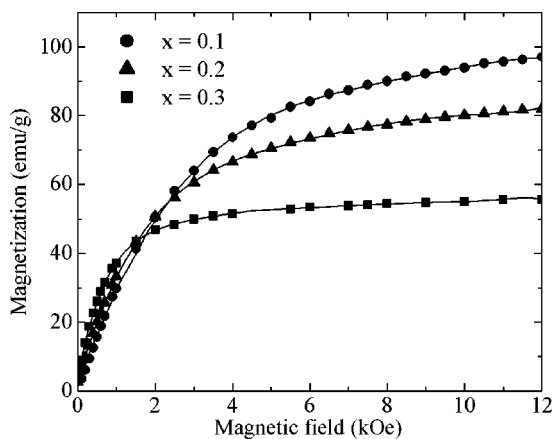


FIG. 2. Magnetization curves of $(\text{Sm}_{0.9}\text{Al}_{0.1})_3(\text{Fe}_{1-x}\text{Al}_x)_{27.5}\text{Ti}_{1.5}$ compounds at 300 K.

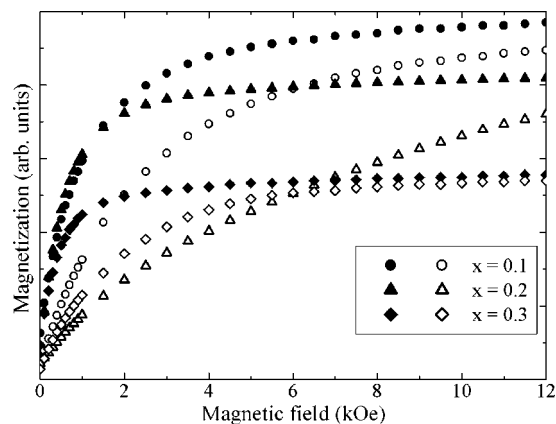


FIG. 3. Magnetization curves of aligned samples of $(\text{Sm}_{0.9}\text{Al}_{0.1})_3(\text{Fe}_{1-x}\text{Al}_x)_{27.5}\text{Ti}_{1.5}$ compounds along (solid symbols) and perpendicular (open symbols) to the direction of alignment.

direction has been found to be making an angle of 66.69° with a axis, in $x=0.1$ sample whereas, the angle is 66.64° in the other two compounds. The (204) plane in the 3:29 system is equivalent to the (004) plane of 2:17 setting or (301) plane of 1:12 setting, obtained from the conversions reported by Han *et al.*¹³ In other words, the easy magnetization is along the c -axis in the 2:17 setting and is along [301] in the 1:12 setting. Magnetization curves on magnetically aligned samples are shown in Fig. 3. The area between the magnetization curves in the case of the $x=0.2$ sample seems to be more than in the case of the other two. However, the anisotropy field (H_A) estimated from the singular point detection method (SPD) decreases with increasing Al as shown in Table I.

The net anisotropy in these compounds is the sum of the Fe sublattice anisotropy (planar) and rare earth sublattice anisotropy (depends on the sign of the product of second order Steven's coefficient α_J and the second order crystal field parameter A_2^0). Since the rare earth sublattice anisotropy is determining the net anisotropy, and Sm having a large and positive α_J , many $\text{R}_3(\text{Fe},\text{M})_{29}$ compounds with $\text{R}=\text{Sm}$ have a tendency to exhibit uniaxial anisotropy after nitrogenation^{7,14,15} or carbonation.¹⁶ However, in $(\text{Sm}_{0.9}\text{Pr}_{0.1})_3\text{Fe}_{27.5}\text{Ti}_{1.5}$ the substituent Al seems to do the job of increasing the contribution from the Fe sublattice to the net anisotropy, causing it to become uniaxial. In fact the effect of Al on the Co sublattice anisotropy has been demonstrated in $\text{Gd}_2\text{Co}_{17-x}\text{Al}_x$.¹⁷ In $\text{Sm}_2\text{Fe}_{1-x}\text{Al}_x$ compounds as well, such an effect has been observed for $x > 2$.¹⁸

CONCLUSIONS

The effect of Al substitution for Fe in $(\text{Sm}_{0.9}\text{Pr}_{0.1})_3\text{Fe}_{27.5}\text{Ti}_{1.5}$ ($x=0.1, 0.2, 0.3$) compounds have been investigated. Expansion of unit cell volume is explained as due to the larger atomic radius of Al replacing smaller Fe. Decrease in room temperature saturation magnetization and increase in Curie temperature are attributed to the charge transfer from Al to Fe. Uniaxial anisotropy in all compounds (in 2:17 setting) is observed.

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