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Structural and magnetic properties of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$ ($x=0.1-0.4$)

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The structural and magnetic properties of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$ ($x=0.1-0.4$) were investigated by x-ray diffraction, magnetization, and Curie temperature (T_C) measurements. The unit cell volume increases with the Al substitution, in accordance with the larger atomic radius of Al. The saturation magnetization and the Curie temperature decrease with increasing Al content, and the decrease is attributed to the modification of the density of states of $3d$ band due to the $3p$ band of Al and to the magnetic dilution effect. The easy direction of magnetization is along the b axis for $x=0.4$. The anisotropy field (H_A) decreases with increasing Al concentration. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2172911]

I. INTRODUCTION

A class of R-TM intermetallics, namely, $R_3(Fe,M)_{29}$, attracted much attention in recent years due to their promising intrinsic magnetic properties such as large magnetization, high Curie temperature, etc., for permanent magnet applications.[1–3] These compounds are found to form in $Nd_3(Fe,Ti)_{29}$-type monoclinic structure with $A2/m$ space group,[4] a combination of alternate stacking of rhombohedral Th_2Zn_{17} (2:17)- and tetragonal $ThMn_{12}$ (1:12)-type segments with two inequivalent sites for R and 11 inequivalent sites for (Fe,M).[4–6] Substitution of Fe by Co in 3:29 compounds has a remarkable effect on their magnetic properties.[7–10] Pan *et al.*[11] have reported the effect of Ga substitution on the intrinsic magnetic properties of $R_3(Fe,Mo)_{29}$ ($R=Y,Sm$). Ga substitution changes the anisotropy of $Sm_3(Fe,Mo)_{29}$ from planar to conical. However, this leads to magnetic dilution as well. Substitution of Al has been reported to improve the magnetic properties in 2:17 compounds.[12,13] Since the 3:29 phase is structurally related to both 2:17 and 1:12 phases, it is of interest to investigate the effect of Al substitution in 3:29 materials. Hu *et al.*[14] on the basis of neutron diffraction studies on $Nd_nFe_{m-x-y}V_xM_y$ $[(n,m)=(1,12),(2,17),(3,29)]$ have shown that in the 3:29 compounds the easy magnetization direction (EMD) is along the a axis. Venkatesh *et al.*[15] have reported the effect of Al substitution at the Fe site on the magnetic properties of $(Sm_{0.9}Pr_{0.1})_3Fe_{27.5}Ti_{1.5}$. Even though there was a mention of $Nd_3(Fe,Al)_{29}$ by Cadogan *et al.*,[2] structural and magnetic details are not available. In the present paper, we report structural and magnetic studies of these $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$ alloys, for $x=0.1-0.4$.

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II. EXPERIMENTAL DETAILS

The compounds were prepared by arc melting the constituent elements of high purity (Nd—99.9%, Fe—99.95%, Al—99.9%, and Ti—99.99%) in an arc furnace under argon atmosphere. The compounds were melted several times to ensure homogeneity. The weight loss after the final melting was found to be less than 0.5%. The melted ingots were wrapped in tantalum foil, sealed in quartz tube in vacuum $[10^{-6}\ \text{torr} < \text{pressure} < 10^{-5}\ \text{torr}]$, homogenized at 1373 K (1100 °C) for 5 days, and were quenched in ice and water mixture. Structural characterization was done by taking powder x-ray diffraction patterns employing Fe $K\alpha$ radiation. Magnetization and Curie temperature measurements were carried out using the vibrating sample magnetometer (VSM) (Model No. PAR 155) up to an applied field of 1.2 T. The powders were aligned in a magnetic field of 2.5 T for investigating the anisotropy and easy magnetization direction (EMD). X-ray diffraction (XRD) patterns of magnetically aligned powder samples with the alignment direction normal to the substrate were taken in order to study the magnetic anisotropy of the compounds, and those that are aligned parallel to the substrate were used to determine the anisotropy fields from magnetization measurements.

III. RESULTS AND DISCUSSION

X-ray diffraction patterns of the compounds are shown in Fig. 1(a). All the compounds have formed with 3:29 phase as the major phase, with 1:12 phase as the secondary phase, and traces of α-Fe. The lattice parameters are listed in Table I. The lattice is seen to expand with Al concentration; this increase is in accordance with atomic radii of Fe and Al, as reported in 2:17 compounds.[12,16]

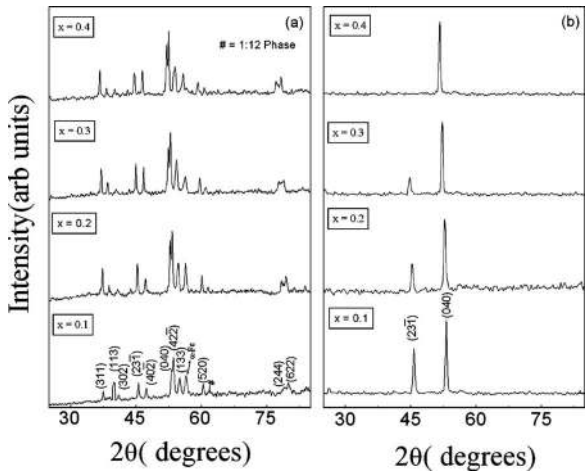

FIG. 1. XRD patterns of random (a) and oriented powders (b) of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$.

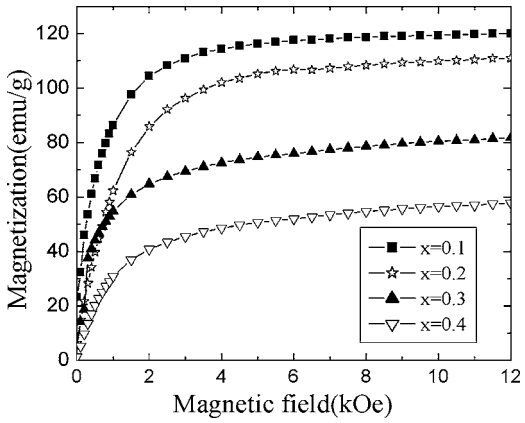

FIG. 2. Magnetization curves (at 300 K) of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$.

Figure 2 shows the magnetization curves of all samples at 300 K, and the magnetizations of all the compounds have reached almost their respective saturation values (M_S), which were obtained from the Honda plots and are shown in Table I. There is a decrease in the magnetization with Al concentration. This could be explained on the basis of the reports on 2:17 compounds. The decrease is attributed to the modification in the density of states (DOS) of 3*d* band of Fe due to the low lying 3*p* band of Al.[17] With the increase in Al concentration, there might be a charge transfer from Al to the 3*d* band of Fe. The spin-up and spin-down densities of states are both unfilled for the parent compound. They get progressively filled up and the spin-up subband gets completely filled with the increase in Al concentration. Consequently, the magnetic moment of the 3*d* sublattice decreases, thereby decreasing the magnetization. A similar reduction in the magnetic moment has also been observed in other 3:29 compounds.[15,18] In addition, magnetic dilution effect on Al substitution also contributes for the reduction in magnetization at high Al concentrations.

The Curie temperature was determined through the magnetization measurements carried out at a field of 50 Oe (Fig. 3). The Curie temperature is seen to decrease from 420 K for $x=0.1$ to 357 K for $x=0.4$. The modification of DOS near the Fermi level leading to a net decrease in the exchange splitting of the spin-up and spin-down bands as in the case of Al substituted Nd_2Fe_{17} (Refs. 17 and 19) could result in the decrease of Curie temperature.

It has been established that for the magnetic anisotropy of the 3:29 compounds the reflections from the (4 0−2), (0 4 0), and (2 0 4) planes play an important role.[3] The XRD patterns taken for the perpendicularly oriented samples [Fig. 1(b)] show that the substitution of Al causes a change in the EMD from cone to axial, as seen from the strong (0 4 0) reflection and the absence of (*h k* 0) and (*h k l*) reflections, for the $x=0.4$ compound. 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). Courtois *et al.*[20] predicted that the EMD of the Fe sublattice anisotropy favors the *b* plane and *R* sublattice anisotropy favors the *b* axis for the rare earths having negative α_J and the *b* plane for the rare earths having positive α_J. Thus, in $Nd_3(Fe_{0.6}Al_{0.4})_{27.5}Ti_{1.5}Al$ seems to cause the planar anisotropy of the Fe sublattice to decrease. As the anisotropy of the Nd sublattice is axial (α_J is negative), this may influence the reduced planar anisotropy of the Fe sublattice, making the total anisotropy become axial.

Magnetization measurements have been carried out at 300 K along the hard and easy directions (Fig. 4). By extrapolating the magnetization curve along the hard direction to meet the curve along the easy direction, the anisotropy

TABLE I. Lattice parameters, room temperature magnetization, anisotropy field, and Curie temperature of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$ compounds.

x	0[a]	0.1	0.2	0.3	0.4
a (Å) (±0.005)	10.638	10.740	10.794	10.855	10.942
b (Å) (±0.005)	8.589	8.667	8.701	8.774	8.847
c (Å) (±0.005)	9.745	9.885	10.015	10.097	10.110
β (deg) (±0.02)	96.93	96.98	97.02	96.06	97.09
V (Å)^3	884	913	933	956	971
M_S (emu/g) (300 K)	143	122	116	87	63
T_C (K)	437	420	415	376	357
H_A (kOe)	37	24	18	14	12

[a]Interpolated from the data in Ref. 3.

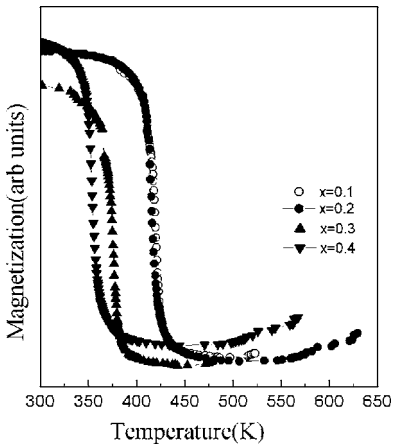

FIG. 3. Temperature variation of magnetization of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$.

fields (H_A) have been determined. The variation of anisotropy field with the concentration is shown in Table I. The anisotropy field is found to decrease with increasing Al concentration.

IV. CONCLUSIONS

The magnetic properties of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$ ($x=0.1-0.4$) are reported. The unit cell parameters increase anisotropically with the Al content. The saturation magnetization and the Curie temperature decrease with Al addition and is attributed to the modification of DOS of $3d$ band by the hybridization with the $3p$ band of Al. In the $x=0.4$ compound, the easy magnetization direction is along the b axis, and this could be due to the reduction in the planar anisotropy of the Fe sublattice.

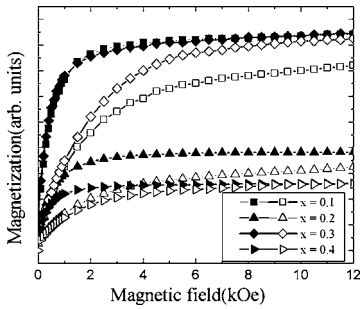

FIG. 4. Magnetization curves of magnetically aligned samples of $Nd_3(Fe_{1-x}Al_x)_{27.5}Ti_{1.5}$ along the easy axis (open symbols) and hard axis (closed symbols) at 300 K.

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