MAGNETIC PROPERTIES OF $R_3(Fe, Mo)_{29}N_x$ (R= Sm OR Y) INTERSTITIAL NITRIDES[①]

Pan Hongge

Department of Materials Science and Engineering,
Zhejiang University, Hangzhou 310027, P. R. China

ABSTRACT $R_3(Fe, Mo)_{29}N_x$ (R= Sm or Y) nitrides have been synthesized by gas phase reaction under an atmosphere of nitrogen. The nitrides retain the structure of parent compounds. The unit cell volume of the nitride is 4.7% for $Sm_3(Fe, Mo)_{29}N_x$ compound and 5.3% for $Y_3(Fe, Mo)_{29}N_x$ compound greater than that of corresponding parent. Introduction of nitrogenation leads to an increase of Curie temperatures T_C and saturation magnetization, and the $Sm_3(Fe, Mo)_{29}N_x$ nitride exhibits uniaxial anisotropy with an anisotropy of 20.5 T at 4.2 K and 14.6 T at 300 K.

Key words magnetic properties rare-earth transition-metal compounds gas phase reaction Curie temperature

1 INTRODUCTION

The $R_3(Fe, M)_{29}$ (R= rare earth; M= Ti, V, Mn, Mo, etc) compounds are newly discovered rare earth iron intermetallics. They almost exist across the whole lanthanide series and crystallize in monoclinic $Nd_3(Fe, Ti)_{29}$-type structure[1-8]. The magnetic properties of a series of $R_3(Fe, M)_{29}$ (R= Ce, Nd, Sm, Gd, Tb, Dy or Y; M= Ti, V, Mn, Cr or Mo) compounds have been studied in great detail[1-8]. The magnetic ordering temperature T_C are remarkably low. Taking $R_3(Fe, Mo)_{29}$ as an example, the Curie temperature T_C ranges from as little as 300 K for R= Ce to 494 K for R= Gd[5]. The saturation magnetization σ_S are also lower than that of corresponding 2∶17 compounds, for example R_2Fe_{17}. In 1990, Coey *et al*[9] have reported that considerable improvements with respective to Curie temperatures and saturation magnetization of R_2Fe_{17} were reached by gas phase interstitial modification. In this paper, we reported the intrinsic properties of $R_3(Fe, Mo)_{29}N_x$ (R= Sm or Y) nitrides.

2 EXPERIMENTAL

Ingots of $Sm_3(Fe_{0.966}Mo_{0.034})_{29}$ and $Y_3(Fe_{0.969}Mo_{0.031})_{29}$ compounds were prepared by argon arc melting with starting elements of at least 99.9% purity, with an excess amount of Sm and Y elements to compensate for its loss during melting. The ingots were melted in a water cooled copper hearth and remelted at least five times for homogeneity. The ingots were annealed at 1 453 K for 48 h under argon atmosphere, then quenched in water. In order to prepare the nitride, the ingots were pulverized into fine powders with an average size of 10~15 μm, and then the nitrogenation was performed by heating the fine powder in nitrogen at 100 kPa at 823 K for 2.5 h for $Sm_3(Fe_{0.966}Mo_{0.034})_{29}$ compound and 803 K for 2 h for $Y_3(Fe_{0.969}Mo_{0.031})_{29}$ compound, respectively. The nitrogen content was determined from the difference between the mass before and after nitrogenation.

X-ray diffraction with Cu-K_α radiation was used to identify the phases present in the compounds and to determine the lattice parameters. Thermomagnetic analysis (TMA) was per-

① Project 59501006 supported by the National Natural Science Foundation of China
Received Sep. 25, 1997; accepted Jan. 12, 1998

formed in a low field of about 0.04T in the temperature range from 300 K to above the Curie temperature. The Curie temperatures T_C were determined from σ^2-T plots by extrapolating σ^2 to zero. The magnetization curves were measured by extracting sample magnetometer (ESM) with a superconducting magnet of maximum magnetic field up to 7T. Saturation magnetization σ_S were derived from σ-$1/B$ plots based on the magnetization curves. The anisotropy fields B_a were estimated from the extrapolated intersection point of two magnetization curves measured with the magnetic field applied parallel and perpendicular, respectively, to the alignment direction of the cylinder samples.

3 RESULTS AND DISCUSSION

The X-ray diffraction patterns of the $R_3(Fe, Mo)_{29}N_x$ (R = Sm or Y) nitrides are shown in Fig. 1. It can be seen that the nitrides retain the structures of parent compounds which crystallized in $Nd_3(Fe, Ti)_{29}$ type[10] structure and the peaks of the $R_3(Fe, Mo)_{29}$ nitrides shift to a smaller angle compared with that of the parent compound showing that the unit cell volume of $R_3(Fe, Mo)_{29}$ nitrides have been expanded. The unit cell parameters a, b, c, β and the unit cell volume V of the $R_3(Fe, Mo)_{29}$ nitrides and their parents are listed in Table 1. the unit cell volume V is 4.7% greater for $Sm_3(Fe, Mo)_{29}$ nitride and 5.3% greater for $Y_3(Fe, Mo)_{29}$ nitride than corresponding parent compound. The nitrogen content x is about 3.9 for $Sm_3(Fe, Mo)_{29}N_x$ nitride and 3.8 for $Y_3(Fe, Mo)_{29}N_x$ nitride which are similar to that of the $Nd_3(Fe, Ti)_{29}N_x$ nitride[2].

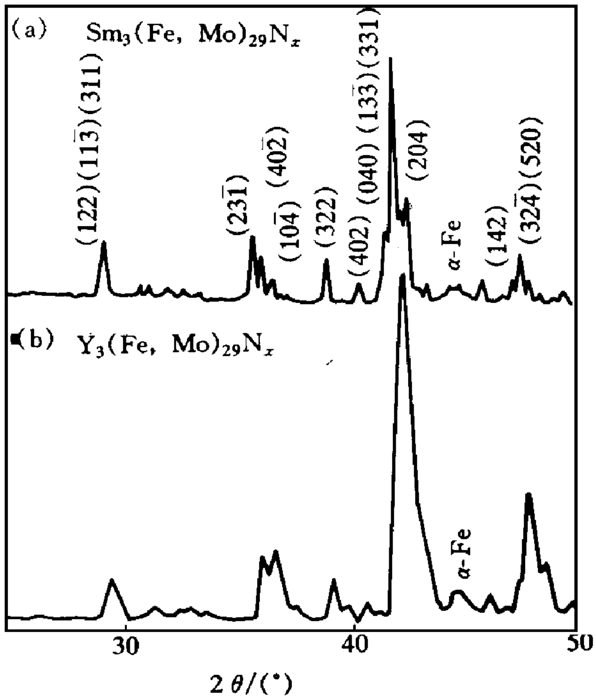

Fig. 1 X-ray diffraction patterns for $R_3(Fe, Mo)_{29}N_x$ (R= Sm or Y) nitrides

The magnetization as a function of temperature for isotropic polycrystalline samples of $R_3(Fe, Mo)_{29}N_x$ (R = Sm or Y) nitrides are shown in Fig. 2, together with those of the $R_3(Fe, Mo)_{29}$ (R= Sm or Y) parents for comparison. The Curie temperatures T_C were derived to be 704 K for $Sm_3(Fe, Mo)_{29}N_x$ nitride, which is 58.2% higher than that of the parent and 659 K for $Y_3(Fe, Mo)_{29}N_x$ nitride, which is 75.2% higher than that of the parent. These results are also listed in Table 2.

The saturation magnetization σ_S of $R_3(Fe, Mo)_{29}N_x$ (R= Sm or Y) nitrides and their parents at 4.2K and 300K are listed in Table 2.

Table 1 Lattice parameters and unit cell volumes of $R_3(Fe, Mo)_{29}N_x$ nitrides (R= Sm or Y)

Compound	a/ Å	b/ Å	c/ Å	β/(°)	V/ Å^3	$\frac{\Delta V}{V}$/%
$Sm_3(Fe, Mo)_{29}$	10.622	8.568	9.738	96.83	879.92	–
$Sm_3(Fe, Mo)_{29}N_x$	10.805	8.703	9.870	97.07	921.09	4.7
$Y_3(Fe, Mo)_{29}$	10.568	8.505	9.672	96.89	861.86	–
$Y_3(Fe, Mo)_{29}N_x$	10.713	8.666	9.839	96.57	907.40	5.3

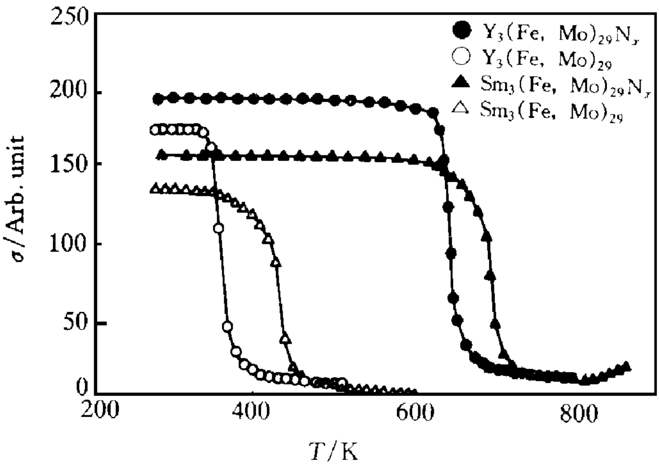

Fig. 2 Magnetization as a function of temperature for $R_3(Fe, Mo)_{29}$ (R= Sm or Y) compounds and their nitrides

Table 2 Curie temperatures T_C and saturation magnetization σ_S of $R_3(Fe, Mo)_{29}N_x$ nitrides (R= Sm or Y) compared with those of parents

Compound	T_C/K	σ_S/(Am²·kg⁻¹) 4.2K	300K
$Sm_3(Fe, Mo)_{29}$	445.0	135.0	107.0
$Sm_3(Fe, Mo)_{29}N_x$	704.0	152.0	137.0
$Y_3(Fe, Mo)_{29}$	376.0	147.0	143.3
$Y_3(Fe, Mo)_{29}N_x$	659.0	162.1	106.5

The temperature dependence of the σ_S of $R_3(Fe, Mo)_{29}N_x$ nitrides and average Fe moments μ are presented in Fig. 3 and Table 3. The increase in σ_S is attributed to the increase in average Fe moments after nitrogenation. The increase in Curie temperature T_C upon nitrogenation may partly be explained in terms of lattice expansion of the nitride which leads to an increase in the average nearest-neighbor Fe-Fe exchange interaction. A theoretical analysis shows that the increase in Curie temperature T_C may also be ascribed to the increase in magnetization upon nitrogenation and the decrease in the spin up density of states at the Fermi level E_F associated with narrowing of the 3d band[11].

Fig. 4 shows the X-ray diffraction patterns for a magnetically aligned powder samples. It can be seen that the easy magnetization direction

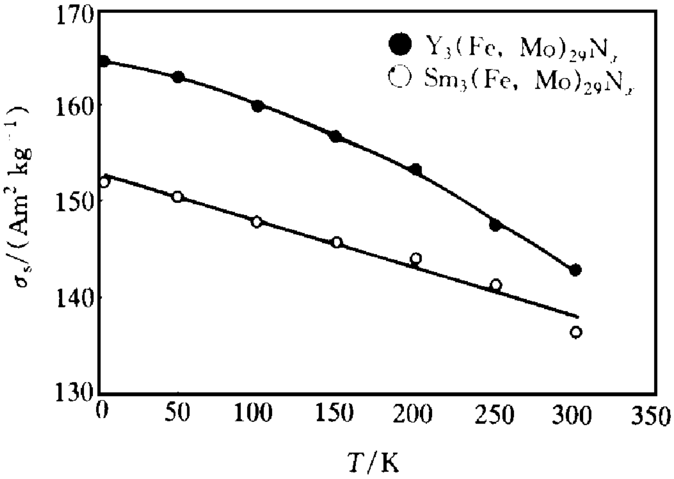

Fig. 3 Saturation magnetization σ_S of $R_3(Fe, Mo)_{29}N_x$ (R= Sm or Y) nitrides at 4.2, 50, 100, 150, 200, 250 and 300 K

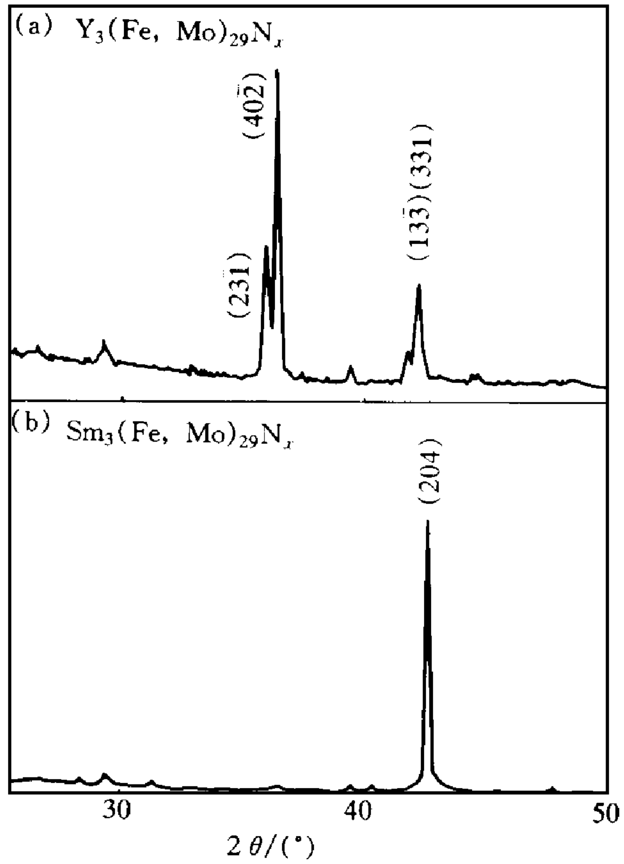

Fig. 4 X-ray powder diffraction patterns of magnetically aligned samples of $R_3(Fe, Mo)_{29}N_x$ (R= Sm or Y) nitrides

of $Sm_3(Fe, Mo)_{29}N_x$ nitride is uniaxial along [102]. Introduction of nitrogen leads to the occurrence of uniaxial anisotropy in the nitride. The temperature dependence of the anisotropy field B_a is shown in Fig. 5. The anisotropy field

Table 3 Temperature dependence of average Fe moments μ of $Y_3(Fe, Mo)_{29}N_x$ nitride and its parent compound

Compound	Average Fe moments, μ						
	4.2K	50K	100K	150K	200K	250K	300K
$Y_3(Fe, Mo)_{29}$	1.80	1.77	1.72	1.64	1.55	1.42	1.24
$Y_3(Fe, Mo)_{29}N_x$	2.08	2.05	2.02	1.97	1.92	1.86	1.80

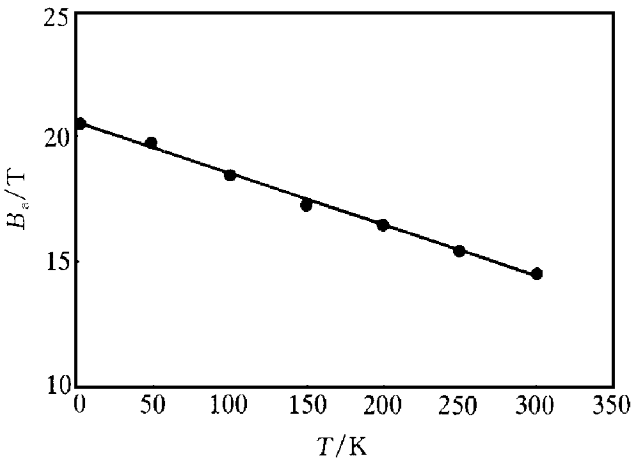

Fig. 5 Temperature dependence of anisotropy field B_a of $Sm_3(Fe, Mo)_{29}N_x$ nitride

B_a of $Sm_3(Fe, Mo)_{29}N_x$ nitride at 4.2K and 300 K are 20.5T and 14.6T, respectively. The value of the B_a decreases monotonically with increasing temperature. It can also be seen that the X-ray pattern of the aligned $Y_3(Fe, Mo)_{29}N_x$ nitride powder is similar to that of the aligned $Y_3(Fe, Mo)_{29}$ powder[12]. The easy magnetization direction in the $Y_3(Fe, Mo)_{29}N_x$ nitride is of planar type. It is revealed that the anisotropy of Fe sublattice in $R_3(Fe, Mo)_{29}N_x$ (R= rare earth) nitrides is of planar type.

4 CONCLUSIONS

In conclusion, the $R_3(Fe, Mo)_{29}N_x$ (R= Sm or Y) nitrides have been synthesized by gas phase reaction under an atmosphere of nitrogen. The nitrides retain the structure of parent compounds. After nitrogenation, the Curie temperature T_C and saturation magnetization σ_S strongly increase, the easy magnetization direction change from planar to uniaxial for $Sm_3(Fe, Mo)_{29}N_x$ nitride, but the sublattice Fe remain easy plane.

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(Edited by Peng Chaoqun)