

Magnetic and Mössbauer spectral studies of $R_3Fe_{29-x}Mo_x$ compounds (R = Y, Nd, Sm, Gd, Tb, and Dy)

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Received 6 June 2004; received in revised form 13 September 2004; accepted 14 September 2004

Available online 28 November 2004

Abstract

The investigation of the structure, magnetic and Mössbauer properties for the series of $R_3(Fe,Mo)_{29}$ compounds has been performed, where R = Y, Nd, Sm, Gd, Tb, and Dy. The crystallographic structure of the ternary phase compounds has been investigated by Rietveld refinement of the X-ray diffraction patterns obtained at room temperature. The quality of the single-phase compounds was also checked by thermomagnetic measurements, from room temperature to above the Curie temperatures. From the magnetic isotherms for the free powder samples, measured at 4.2 K, the saturation magnetizations and the iron average magnetic moments have been derived. ^{57}Fe Mössbauer spectra of the $R_3(Fe,Mo)_{29}$ compounds have been measured at 15 K. The analysis of spectra, in a model which takes into account both the Fe atom nearest neighbor numbers and the Fe–Fe nearest neighbor bond lengths, indicates that the transferred contribution to the hyperfine field at the iron sites, due to rare earth moments, can be correlated with the rare earth effective spin.

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Keywords: Intermetallics; X-ray diffraction; Magnetic measurements; Nuclear resonances

1. Introduction

The magnetic $R_3(Fe,Mo)_{29}$ phase, located at the iron-rich corner of the rare earth–iron–transition metal (R–Fe–T) ternary phase diagrams, is stabilized by a third element (T) and has the monoclinic symmetry in the $A2/m$ space group [1]. The 3:29 structure is an intermediate structure between the well known rhombohedral Th_2Zn_{17} and tetragonal $ThMn_{12}$ structures [2], consisting in a combination of the 2:17 rhomb and 1:12 units in a ratio of 1:1. The families of the 3:29 compounds, such as $R_3(Fe,Ti)_{29}$, $R_3(Fe,Cr)_{29}$, $R_3(Fe,V)_{29}$, and $R_3(Fe,Mo)_{29}$ exhibit a complex structural arrangement and a large variety of interesting physical properties [2–6]. It has also been found that introduction of the interstitial N, H, or C atoms leads to remarkable improvements of the magnetic properties including Curie temperature, the saturation mag-

netization, and the magnetic anisotropy [7,8]. The hyperfine interactions at R and Fe atoms in $R_3(Fe,Ti)_{29}$, $R_3(Fe,V)_{29}$, and $R_3(Fe,Cr)_{29}$ were investigated [8–18] at various temperatures. The only ^{57}Fe Mössbauer study on magnetically aligned powder samples of $Nd_3(Fe,Mo)_{29}$ [19] have been intended to confirm that a spin reorientation takes place between 250 and 300 K. In order to understand the magnetic structure and intrinsic magnetic properties of these compounds, a systematic study on the role of the different rare earth elements in a series of similar compounds is definitely necessary.

In this work, an investigation of the structure, magnetic properties, and hyperfine interactions at iron atoms in the series of $R_3Fe_{29-x}Mo_x$ (R = Y, Nd, Sm, Gd, Tb, and Dy) compounds, with molybdenum as stabilizing element, has been performed. The lattice parameters, Curie temperature, saturation magnetization, average ^{57}Fe hyperfine field, and the rare earth contribution to hyperfine field of these compounds have been obtained.

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2. Experiment

The $R_3Fe_{29-x}Mo_x$ compounds with $R = Y, Nd, Sm, Gd, Tb,$ and Dy were prepared by argon-arc melting, as elsewhere described [5], in which the purity of the starting elements was at least 99.9%. The ingots were melted in a water cooled copper hearth and remelted at least four times to promote homogeneity. In order to compensate the loss of the rare earth during melting and annealing, the starting composition contained an excess amount of rare earth compared with the stoichiometric composition. The ingots were subsequently wrapped in Ta foil and annealed in sealed quartz tubes under a protective argon atmosphere at 1250–1450 K for a period from 1 to 3 days in order to maximize the amount of $R_3Fe_{29-x}Mo_x$ phase. At the end of the annealing process, the ingots were water quenched. The quality of the single-phase compounds was checked by both the thermomagnetic analysis (TMA) and X-ray diffraction (XRD).

TMA was performed in a low field of about 0.02 T with a Weiss-type balance in the temperature range from room temperature to above the Curie temperature T_C . The Curie temperatures were determined from $M^2(T)$ plots, derived from the thermomagnetic magnetization curves $M(T)$, by extrapolating M^2 to zero.

The identification of the phases and the structural study were performed by powder XRD using a Siemens diffractometer in the Bragg–Brentano geometry, with $Cu\ K\alpha$ radiation, at room temperature. The crystallographic structure was refined by the full pattern Rietveld method using the FULLPROF.98 program [20]. An overall temperature factor was used and a pseudo-Voigt function was selected as the peak shape function. The atomic positions for the related phase $Y_3(Fe_{0.969}Mo_{0.031})_{29}$ [21] were used as the starting parameters in the refinement process.

The magnetization curves of the free powder samples were measured at 4.2 K by means of a Quantum Design SQUID magnetometer with a maximum magnetic external field up to 7 T. The saturation magnetizations M_s at 4.2 K were derived from $M(1/B)$ plots and by extrapolating $1/B$ to zero, based on the high field data of the magnetization curves.

The Mössbauer spectra were taken at 15 K, using a conventional constant-acceleration spectrometer with a closed-cycle cryostat for the low temperature measurements. A ^{57}Co source in a rhodium matrix was used and the velocity calibration was made using an α -iron absorber at room temperature. The Mössbauer absorbers contained about $35\ mg/cm^2$ of powdered sample, which had been sieved to a 0.05 mm or smaller diameter particle size. In the fitting procedures, described in Section 3, the spectra were analyzed as a convolution of four independent magnetic subspectra (sextets) based on site assignments defined by Wigner–Seitz cell environments. A fifth sextet was introduced for α -Fe in case of Nd and Gd compounds and the estimated errors for the hyperfine fields are in the range of $\pm 0.3\ T$.

3. Results and discussion

XRD and TMA confirmed that all of the investigated compounds were of single phase and crystallized in $Nd_3(Fe,Ti)_{29}$ -type structure except for a very small amount of unreacted α -Fe as the second phase in Nd and Gd samples. The α -Fe content calculated from the Rietveld analysis was 3.2 wt.% for $Nd_3(Fe,Mo)_{29}$ and 4.1 wt.% for $Gd_3(Fe,Mo)_{29}$ samples, respectively. As an illustration, Figs. 1 and 2 show the calculated XRD patterns (solid lines) compared with the experimental results (circles) of $R_3(Fe,Mo)_{29}$ compounds with $R = Y$ and Nd. The lowest part in Figs. 1 and 2 gives the

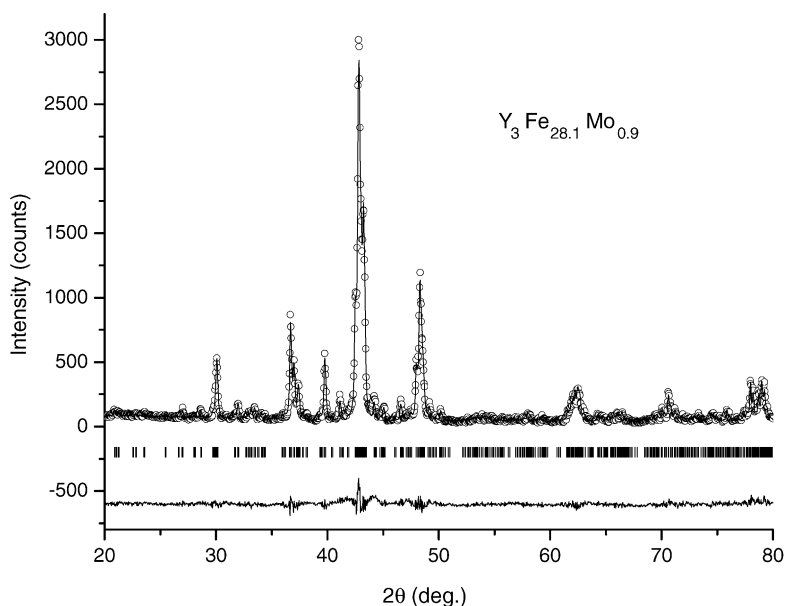


Fig. 1. The X-ray powder diffraction pattern of the $Y_3Fe_{28.1}Mo_{0.9}$ compound refined according to the space group $A2/m$.

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