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LETTER TO THE EDITOR

Structural and magnetic properties of the novel ternary compound $Y_3(Fe, Ti)_{29}$

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Abstract. The compound $Y_3(Fe, Ti)_{29}$ has been successfully prepared with a starting composition of $Y_9Fe_{86}Ti_5$ via annealing at 960 °C for three days. Thermo-gravimetric analysis (TGA) gives a magnetic ordering temperature of 386(3) K. Refinement of powder x-ray diffraction data with the newly discovered monoclinic $Nd_3(Fe, Ti)_{29}$ structure model gives the lattice parameters $a = 10.6068(9)$ Å, $b = 8.5224(6)$ Å, $c = 9.6926(8)$ Å and $\beta = 97.153(7)°$. ^{57}Fe Mössbauer measurements yield an average hyperfine field of 20.2(2) T at 293 K for the new $Y_3(Fe, Ti)_{29}$ compound, which corresponds to an average Fe magnetic moment of about $1.3\mu_B$.

A novel ternary $R_3(Fe_{1-x}T_x)_{29}$ phase (R = Nd [1, 2], Pr [3], Sm [4], Gd [5] and Tb [6]; T = Ti, V Cr and Mn [7, 8], and x in the range of 4–6 at.%) has recently been identified. The crystallographic structure of $Nd_3(Fe_{1-x}Ti_x)_{29}$ (3:29) phase has been determined by means of x-ray diffraction [9] and neutron diffraction [10]. The structure is monoclinic with the space group $P2_1/c$ (No 14). There are two Nd sites, namely 2a and 4e, and fifteen Fe(Ti) sites in the structure, one 2d and fourteen 4e sites. The new $Nd_3(Fe_{1-x}T_x)_{29}$ structure can be obtained by dumb-bell substitution in the hexagonal $CaCu_5$ (1:5) structure, $R_{1-\delta}(2T)_\delta T_5 \rightarrow RT_z$, by analogy with the well known rhombohedral Th_2Zn_{17} (2:17R), $\delta = 1/3$, and tetragonal $ThMn_{12}$ (1:12), $\delta = 1/2$, structures [2]; the 3:29 structure corresponds to $\delta = 2/5$ [2, 9]. In fact, this novel phase can be expressed as: $R_3(Fe_{1-x}Ti_x)_{29} = R_2Fe_{17} + R(Fe_{1-x}Ti_x)_{12}$ [11].

The new $R_3(Fe_{1-x}Ti_x)_{29}$ (R = Pr, Nd and Sm) compounds are ferromagnetic with a Curie temperature of 452 K for R = Sm. It has been shown that after introduction of interstitial nitrogen [1, 4, 12] or carbon [13] atoms, the Curie temperature and uniaxial anisotropy of $R_3(Fe_{1-x}Ti_x)_{29}X_y$ (X = N or C) are both greatly enhanced and comparable to those observed in $Sm_2Fe_{17}X_y$ [14]. These promising intrinsic magnetic properties suggest that the new $Sm_3(Fe_{1-x}Ti_x)_{29}N_y$ compound might be an alternative hard nitride magnet to $Sm_2Fe_{17}N_{3-\delta}$ and $NdFe_{11}TiN_{1-\delta}$ [14]. Crystal-field calculations for $R_3(Fe_{1-x}Ti_x)_{29}$ [11] indicate that the dominant crystal-field coefficient A_{20} has different signs at the two R sites which suggests that some improvement in the intrinsic magnetic properties of $R_3(Fe_{1-x}Ti_x)_{29}$ may result from mixing R elements with opposing Stevens coefficients α_J, e.g. Nd ($\alpha_J < 0$) and Sm ($\alpha_J > 0$).

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Here, we report the preparation and characterization of a new compound $Y_3(Fe_{1-x}Ti_x)_{29}$. A sample with the starting composition $Y_9Fe_{86}Ti_5$ was prepared by a procedure of arc melting the high-purity ($\geqslant$99.9%) constituents in an argon atmosphere, subsequent annealing under an argon atmosphere in sealed quartz tubes for a period of three days at 960 °C, and then quenching in water. The sample was characterized by (i) powder x-ray diffraction (XRD) using Co Kα radiation on a Siemens D5000 diffractometer (the XRD data were analysed using a computer program for Rietveld analysis at fixed wavelength [15]), (ii) scanning electron microscopy using a Jeol JSM840 microscope (SEM), (iii) thermo-gravimetric analysis (TGA) in the presence of a small magnetic field for the determination of the Curie temperatures (T_C) and (iv) ^{57}Fe Mössbauer spectroscopy carried out at 293 K in a standard transmission geometry using a ^{57}CoRh source, with α-Fe calibration.

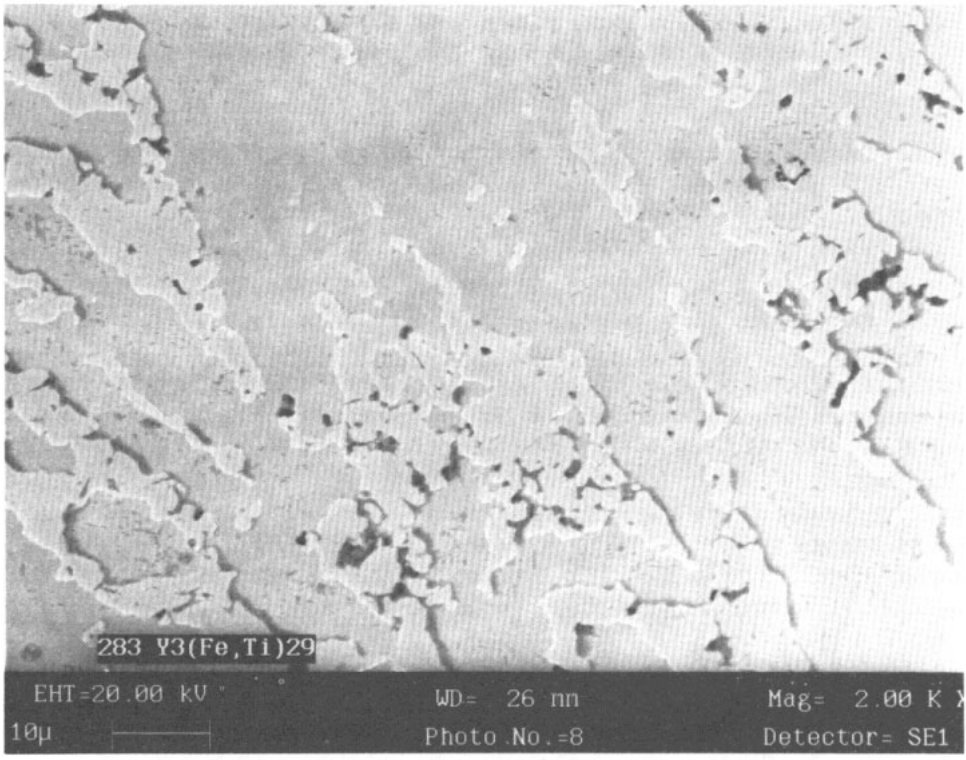

Figure 1. A SEM micrograph of the annealed $Y_9Fe_{86}Ti_5$ sample.

XRD and TGA analyses show that in the as-cast sample, the dominant phase is the $Y_2(Fe_{1-x}Ti_x)_{17}$ phase which has the hexagonal Th_2Ni_{17}-type structure. SEM analysis of the annealed sample shown in figure 1 indicates that the dominant phase is $Y_3(Fe_{1-x}Ti_x)_{29}$ with an impurity phase of $Y(Fe_{1-x}Ti_x)_{12}$, which amounts to ~20% (based on the relative area of the SEM micrograph). Our results show that the annealing treatment is very important in stabilizing the 3:29 phase. Furthermore, it should be mentioned that, according to our

systematic studies on the phase formation in the Y–Fe–Ti system, single-phase 3:29 is very difficult to obtain. The sample featured in this letter is that having the maximum amount of $Y_3(Fe_{1-x}Ti_x)_{29}$ phase, ~80%.

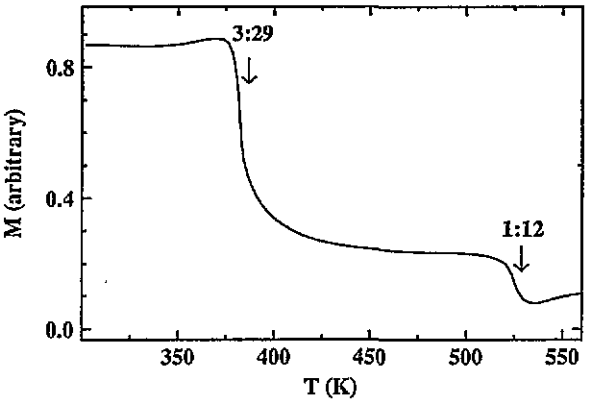

Figure 2. Thermo-gravimetric analysis (TGA) data for the annealed sample of $Y_9Fe_{86}Ti_5$, where the dominant phase is 3:29 with an impurity phase of 1:12 (~20%).

Figure 2 shows the thermo-gravimetric analysis (TGA) data which indicate ferromagnetism in $Y_3(Fe_{1-x}Ti_x)_{29}$ with a Curie temperature of 386(3) K. One can see from this figure that there exists a tail for temperatures higher than the Curie temperature of 3:29 phase, which is consistent with the SEM observation that the ferromagnetic impurity is the 1:12 phase. In addition, the amount of 1:12 phase is again about 20%.

The powder XRD pattern for $Y_3(Fe_{1-x}Ti_x)_{29}$ is shown in figure 3. The theoretical XRD pattern, which was generated by employing the atomic positions of the monoclinic $Nd_3(Fe_{1-x}Ti_x)_{29}$ structure [9] and including a contribution from the impurity 1:12 phase with the tetragonal $ThMn_{12}$ structure, is represented by the solid line. The refinement gives excellent agreement with the experimental data and yields lattice parameters of $a = 10.6068(9)$ Å, $b = 8.5224(6)$ Å, $c = 9.6926(8)$ Å and $\beta = 97.153(7)°$ for $Y_3(Fe_{1-x}Ti_x)_{29}$.

Figure 4 shows the ^{57}Fe Mössbauer spectrum collected at 293 K, and the theoretical fit to this spectrum is represented by the solid line. Given the large number of Fe sites (=15) in the monoclinic $Nd_3(Fe_{1-x}Ti_x)_{29}$ structure and the effects of possible random site occupancy of Ti atoms, only the average hyperfine field can be deduced from the Mössbauer spectra of 3:29 compounds. Therefore, our analysis employed three magnetically split sextets with variable line-widths (inclusion of extra subspectra in the fitting procedure had little effect on the average hyperfine-field value). The fitted value of the ^{57}Fe average hyperfine field thus deduced is 20.6(2) T. After correcting for the 20% contribution of the $Y(Fe,Ti)_{12}$ phase, whose average ^{57}Fe hyperfine field is 22.0 T [16], we find an average hyperfine field of 20.2(2) T for the $Y_3(Fe_{1-x}Ti_x)_{29}$ phase.

In conclusion, we have successfully prepared the new ternary intermetallic compound $Y_3(Fe,Ti)_{29}$. Detailed analysis of its powder x-ray diffraction pattern shows that this new

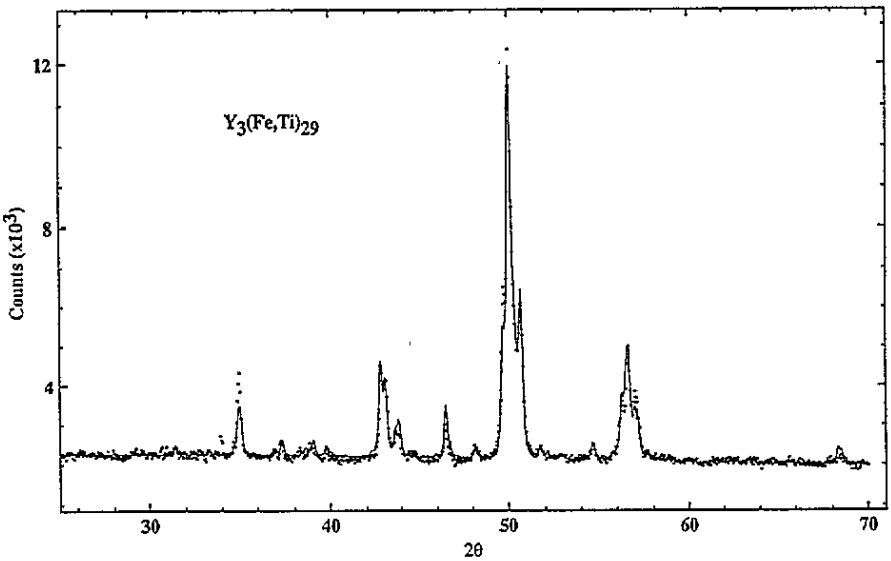

Figure 3. The powder x-ray diffraction pattern (Co Kα) of $Y_9Fe_{86}Ti_5$. The solid line represents the theoretical fit to the experimental data shown by points (see the text).

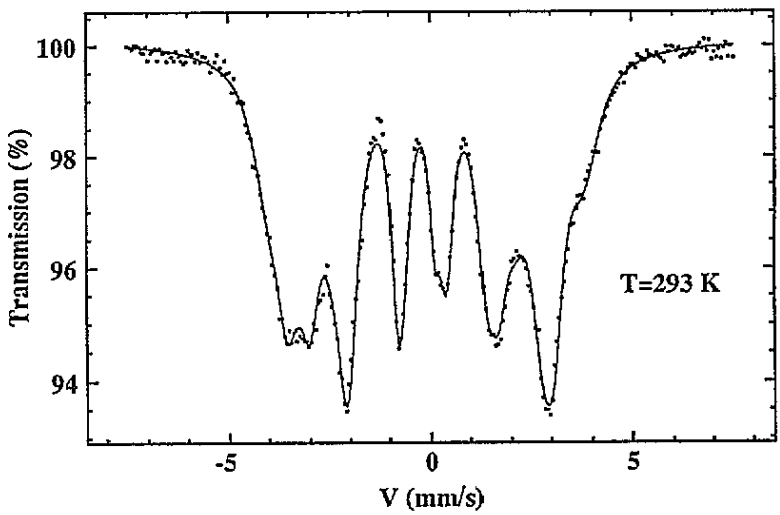

Figure 4. The ^{57}Fe Mössbauer spectrum for $Y_9Fe_{86}Ti_5$. The solid line represents the theoretical fit to this spectrum.

3:29 compound crystallizes in the novel monoclinic $Nd_3(Fe,Ti)_{29}$ structure. The magnetic ordering temperature of $Y_3(Fe,Ti)_{29}$ is 396(3) K, and ^{57}Fe Mössbauer measurements yield an average hyperfine field of 20.2(2) T at 293 K, which corresponds to an average Fe

magnetic moment of about $1.3\mu_B$. Our present work extends the list of 3:29 compounds with a non-magnetic R, which makes it possible to isolate the magnetic properties of the 3d sublattice for this series of compounds.

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