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Influence of H and N insertion on the magnetostriction and thermal expansion of $YFe_{10}V_2Z_x$ (Z = N, H) compositions

M.R. Alinejad[a,*], N. Tajabor[a], H. Khandan Fadafan[a], D. Fruchart[b], D. Gignoux[b]

[a]*Department of Physics, Ferdowsi University of Mashhad, Mashhad 91775-1436, Iran*
[b]*Dept. Matière Condensée Matériaux, Fonctionnalités former Laboratoire de Cristallographie du CNRS, INSTITUT NEEL, BP 166, 38042 Grenoble Cedex 9, France*

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Abstract

Experimental results on the thermal expansion and magnetostriction of $YFe_{10}V_2$ composites are reported and the influence of H and N interstitial atoms is studied. The anisotropic magnetostriction is about 30% larger in the composite than in the starting alloy. Also, the anisotropic magnetostriction remains positive after insertion of H (N) ion while the sign of volume magnetostriction changes by hydrogenation. The anisotropic magnetoelastic interactions are enhanced by insertion of H and especially N interstitial atoms. The results are discussed considering the effect of H and N, and of temperature on magnetic anisotropy and microstructure.

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Keywords: Thermal expansion; Magnetostriction; Magnetoelastic; Y–Fe–V composites

1. Introduction

$REFe_{12-y}M_y$ (RE is rare earth element or Y and M is V, Ti, Mo, etc.) compounds with simple tetragonal $ThMn_{12}$-type structure are interesting because of their rather high Curie temperature (for instance, 532 K in $YFe_{10}V_2$) and magnetization level [1]. In addition, it is well known that the magnetic properties of these compounds are considerably affected by insertion of interstitial small atoms (e.g. H, N and C), so that some of them (particularly, $NdFe_{10.5}V_{1.5}$) exhibit intrinsic ferromagnetic properties better than those of $Nd_2Fe_{14}B$, the archetype of permanent magnet materials. In this respect, much attention has been paid on the effect of interstitial atoms on the crystallographic structure and magnetic properties of these compounds [1–13]. The purpose of this research is to study the influence of H and N interstitial atoms on the magnetoelastic properties of $YFe_{10}V_2$ compound, which have been rather less considered until now. We made this by measuring magnetostriction and thermal expansion of the polymer-bonded $YFe_{10}V_2Z_x$ (Z = N, H) powders.

The first effect of the insertion of interstitial atoms into $REFe_{12-y}M_y$ compounds is an expansion of their crystallographic unit cell (about 1% and 3% in volume upon insertion of H and N, respectively) [10,11]. Moreover, previous results show that the interstitial atoms occupy 2b crystallographic sites in the $ThMn_{12}$ structure. From a different viewpoint, the second-order crystal field parameter (A_2^o), and accordingly the magnetic anisotropy, is oppositely influenced by donor and acceptor interstitial elements. For example, insertion of the $H^+(N^-)$ enforces the negative (positive) contributions to A_2^o [4,5], so that the planar (axial) magnetic anisotropy is strengthened in the $REFe_{12-y}M_y$ compounds whenever RE is a rare earth element with negative Stevens factor (i.e. $\alpha_j<0$), such as Nd. But, in the case of the $YFe_{10}V_2$ compound where the Y^{+3} is non-magnetic and Fe-sublattice anisotropy favors *c*-axis as easy magnetization direction, insertion of H^+ (C^- or N^-) does not modify the easy axis, but weakens the anisotropy [1,11–13]. Briefly, most of magnetic properties of $REFe_{12-y}M_y$ compounds are affected by insertion of

*Corresponding author. Tel.: +98 511 8793912; fax: +98 511 8796416.
E-mail address: alinejad@ferdowsi.um.ac.ir (M.R. Alinejad).

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interstitial atoms. Hence, we expect sensible changes of the magnetoelastic interactions by introducing the interstitial atoms within the $YFe_{10}V_2$ compound. Considering the non-magnetic nature of Y ions, we may assume their contribution as negligible in the following results and attribute them to the effect of H and N on the magnetostrictive strains of Fe sublattice in this family of compounds.

2. Experimental details

$YFe_{10}V_2$ ingots were prepared by high frequency melting of the constituting elements under purified-argon atmosphere. Then, as-cast ingots were enveloped within tantalum foils and annealed for 24 h at 960 °C in a 133×10^{-9} mbar evacuated furnace. After complete removing of the outer oxidized layers by sand paper, the alloys were crushed into powders in alcohol to a size less than 200 μm. The hydrogenation and nitrogenation treatments of powders have been executed in an autoclave for 48 h duration under $P = 6$ and 20 bar pressures and $T = 250$ and 400 °C temperatures, respectively. Phase purity of the powders was assisted by X-ray diffraction (XRD).

For magnetoelastic effects experiments, the homogeneous and isotropic disk-shaped samples (with 8 mm in diameter, 2 mm thickness) were prepared by embedding powders into epoxy resin with the weight proportion of epoxy with respect to alloy powder of 5:100, and then, the mixtures were compacted using 2 GPa pressure. The epoxy resin was fully dried by post-annealing of the compact mixtures at 150 °C for 2 h. No creep has appeared during magnetostriction measurements.

Magnetostriction and thermal expansion are measured by standard strain gage in applied fields up to 1.5 T, and in the temperature range of 77–320 K. The accuracy of these measurements is better than 2×10^{-6}. By measuring magnetostriction parallel (λ_t) and normal (λ_n) to the field direction, the anisotropic ($\Delta\lambda = \lambda_t - \lambda_n$), and volume ($\Delta V/V = \lambda_t + 2\lambda_n$) magnetostrictions were deduced. Thermal expansion coefficients (α) and their average in 80–300 K temperature interval were obtained by calculating slopes of the experimental curves and the corresponding linear fits.

3. Results and discussion

XRD patterns of the prepared samples confirm the tetragonal structure for $YFe_{10}V_2$ with $ThMn_{12}$ symmetry as major phase beside tiny traces of α-Fe as a minor phase. After nitrogenation, increase of the percentage of minor phase is negligible which was expected from low temperature of nitrogenation (400 °C) [6]. But, the XRD pattern shows that the crystallites are partially decomposes after nitrogenation so that only three broadened peaks of the major phase have been appeared in the XRD pattern of the nitrogenated compound. As shown in Table 1, refined lattice parameters of the major phase are consistent with the literature [10–12]. Also, it is clearly seen that the lattice parameters, and accordingly the unit cell volume, increase after insertion of the interstitial H and N atoms. It should be noticed that the lattice parameter refinements of the nitrogenated compound are not so accurate due to low number of the Brag's peaks. From comparison of the volume expansion with the literature and by weighting the samples before and after the gas–solid reactions, the amount of absorbed interstitial elements by metal matrix was estimated to be $x(H) = 1.0$ and $x(N) = 2.1$.

Table 1
Lattice parameters of $YFe_{10}V_2Z_x$ (Z = N, H) and expansion of the unit cell volume after hydrogenation and nitrogenation

Compound	a (Å)	c (Å)	V (Å^3)	$\Delta V/V$ (%)	α (10^{-5} K^{-1})	
					80 K	300 K
$YFe_{10}V_2$	8.4953	4.7734	344.490	–	1.39	2.96
$YFe_{10}V_2N_x$	8.5543	4.7733	349.291	1.4	1.03	2.91
$YFe_{10}V_2H_x$	8.5097	4.7800	346.143	0.48	1.14	2.69

Thermal expansion coefficients at 80 and 300 K have also been given.

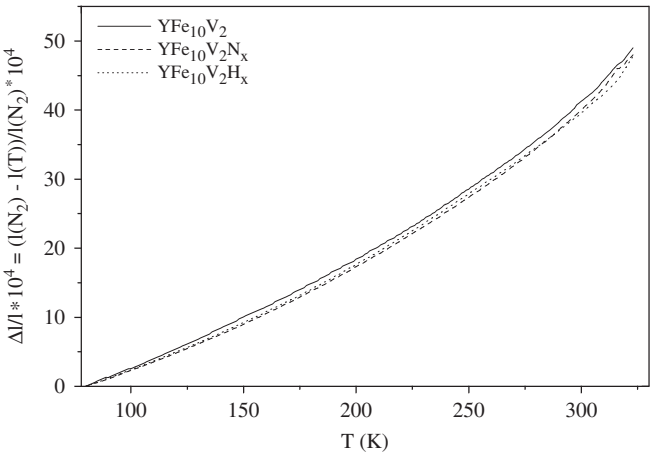

Fig. 1. Thermal expansion of $YFe_{10}V_2Z_x$ (Z = N, H) composites.

The magnitude of the thermal expansion coefficients (α), deduced from slopes of the experimental curves of the thermal expansion at typical temperatures of 80 and 300 K, is given in Table 1. Experimental curves of the thermal expansion are shown in Fig. 1. As clearly seen from this figure, thermal expansion curves of the Y-based compounds exhibit similar thermal variations, although their average thermal expansion coefficients changes about 10% upon insertion of interstitial atoms. Considering that the magnetic contribution to the thermal expansion of a polycrystalline sample originates from isotropic strictions, this similarity indicates that the H and N insertions do not affect considerably the isotropic magnetoelastic interactions in the $YFe_{10}V_2$ compound.

Fig. 2 shows typical isothermal curves of the anisotropic magnetostriction of the studied samples. It is clear that the anisotropic magnetostriction of all samples is positive.

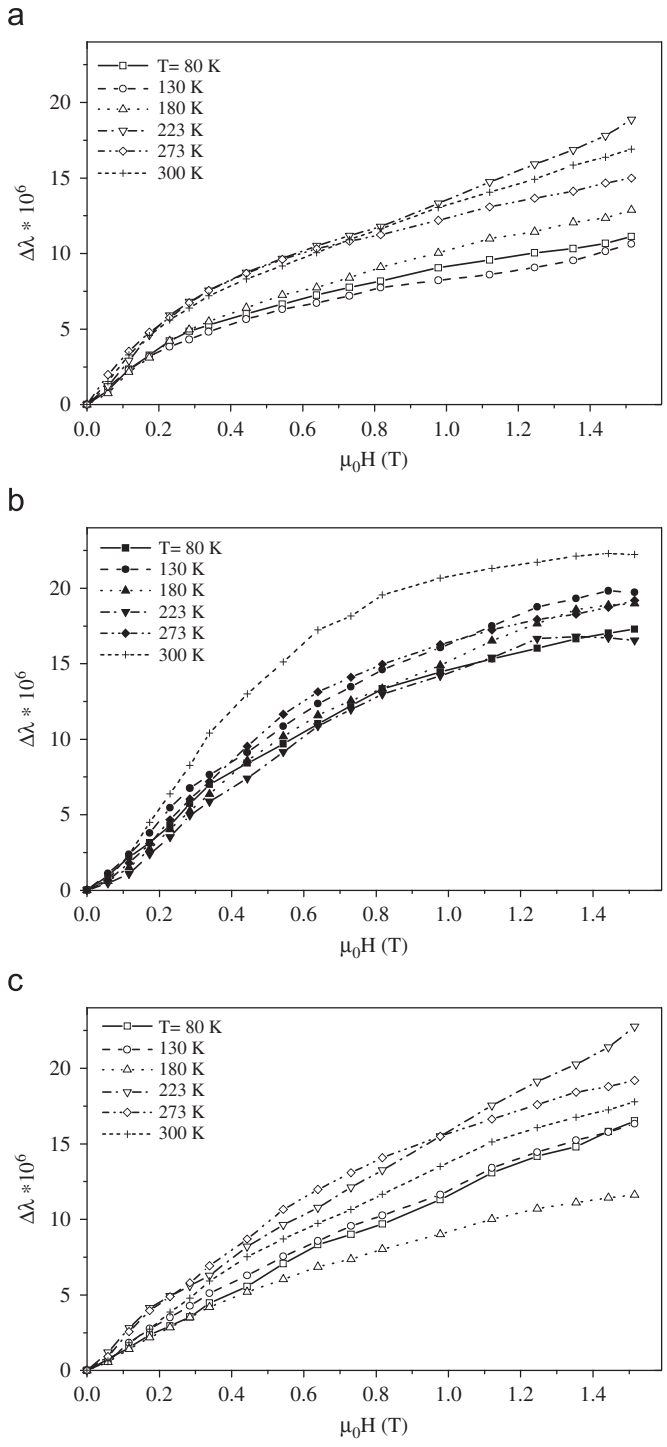

Fig. 2. Anisotropic magnetostriction of $YFe_{10}V_2Z_x$ (Z = N, H) compositions ($\Delta\lambda(80\,K) \equiv \Delta\lambda(N_2)$). (a) $YFe_{10}V_2$, (b) $YFe_{10}V_2N_x$ and (c) $YFe_{10}V_2H_x$.

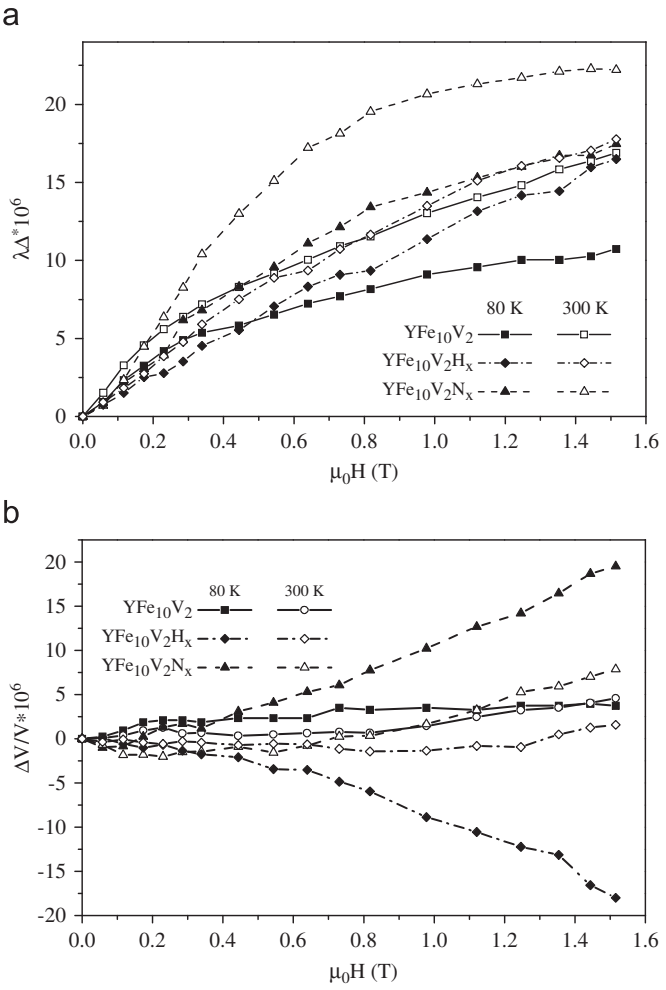

Fig. 3. Influence of H and N insertion on the anisotropic and volume magnetostriction of $YFe_{10}V_2Z_x$ (Z = N, H) compositions.

The anisotropic and volume magnetostrictions of the studied compounds at two typical 80 and 300 K temperatures are compared in Fig. 3. The main characteristics of these results are the followings:

i. For the compound without inserted elements, the anisotropic magnetostriction curves exhibit a rather pronounced negative curvature in the low field region and then tend toward a linear variation at higher fields (Fig. 2a). These variations are similar to those previously reported for polycrystalline samples of the $YFe_{10}V_2$ alloy [14,15]. However, the data of the present study are about 30% larger than in the previous study.
ii. The sign of low field curvature of $\Delta\lambda$ isothermal curves, that is negative for the original sample (Fig. 2a), changes to positive in the nitride (Fig. 2b) and tends to zero in the hydride samples (Fig. 2c).
iii. $\Delta\lambda$ curves of the nitride sample (Fig. 2b) tend to saturate in presence of applied fields below 1.5 T, the effect being more evident at higher temperatures.
iv. $\Delta\lambda$ increases as temperature is enhanced from 80 to 300 K, the effect being the largest in the starting alloy and in the nitride.
v. $\Delta\lambda$ increases after insertion of H or N, the effect being the largest in the nitride.
vi. $\Delta V/V$ variations are of parabolic or almost linear type.

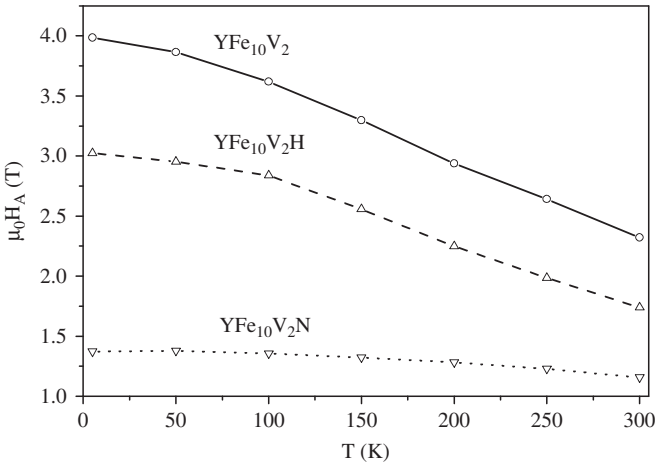

Fig. 4. The effect of H and N on the magnetic anisotropy field of $YFe_{10}V_2Z_x$ (Z = N, H) compositions. These values have been measured using singular point detection method. The required magnetization curves have been obtained using an extracting sample magnetometer along hard axis of the magnetization of previously aligned compounds.

vii. $\Delta V/V$ is rather weak except at 80 K in the nitride where it is positive and in hydride where it is negative.

The interpretation of all these behaviors is not simple on account of the different possible contributions. First of all, the striction of the resin has to be taken into account. Indeed, when the crystallites in a compound or in a composite have become single domains under an applied field, they can be considered as dipoles. In that case, it has been shown that dipolar forces tend to elongate the sample in the field direction. On account of the much smaller Young's modulus of the resin compared to that of the alloy, the elongation is expected to be larger in a composite than in the alloy [16,17]. This is in agreement with the larger value of the anisotropic magnetostriction in the composite than in the polycrystal as underlined in item (i).

In addition, domain's effect that is the largest contribution to the magnetostriction in cubic materials appears as different sign of low field curvatures of $\Delta\lambda$ (item (ii)). This effect occurs although spontaneous magnetization of all samples is along the tetragonal *c*-axis [11]. Indeed, we expect such an effect when the easy direction of magnetization is not uniaxial or is in the basal plane as in $YFe_{10.5}Mo_{1.5}N$ [10]. This effect indicates that different domain patterns form in the original, nitride and hydride samples.

Considering that, the unsaturated value of anisotropic magnetostriction increases when the anisotropy decreases; thermal effects (item (iv)) can be ascribed to the decrease of the anisotropy field between 80 and 300 K as it is shown in Fig. 4. As well, effect of H and N insertion on $\Delta\lambda$ (item (v)) is in agreement with the decrease of the anisotropy field when these elements are inserted (cf. Fig. 4). In addition, saturation trend of $\Delta\lambda$ of the nitride sample as quoted in item (iii) is consistent with this fact that the anisotropy field of this sample is below 1.5 T (Fig. 4).

Concerning the volume effect (item (vi)), on account of the increase of the Curie temperature after insertion in these type of materials, one expects a positive volume magnetostriction as it is occurred after nitrogenation. The negative volume magnetostriction observed in the hydride (item (vii)) shows that probably other contributions are responsible for this effect. In fact, as the applied field increases above a few tenth of tesla, the magnetostriction mainly originates from the magnetocrystalline anisotropy that mainly contributes to the anisotropic magnetostriction and from the dependence of magnetic interactions to the inter-atomic distance that is at first order isotropic and mainly contributes to the volume magnetostriction. So, the electrical field of H^+ ions and their effect on the magnetocrystalline anisotropy and inter-atomic distances should be considered on the top of these contributions.

4. Concluding remarks

The effect of insertion of H and N interstitial atoms on the magnetostriction and thermal expansion of $YFe_{10}V_2$ composition is studied. Results show that these insertions have not considerable effects on the isotropic magnetoelastic interactions. In addition, low field curvatures of the anisotropic magnetostriction curves change by these insertions. Also, anisotropic magnetostriction curves exhibit saturation behavior in presence of available fields in this research, only after nitrogenation. Finally, magnitude of the volume magnetostriction at 80 K considerably increases by these insertions, although its sign changes to negative by hydrogenation. These results are discussed based on the effect of H and N interstitial atoms on the microstructure and magnetocrystalline anisotropy of $YFe_{10}V_2$ composition.

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