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Journal of Alloys and Compounds 458 (2008) 91–95

Journal of ALLOYS AND COMPOUNDS

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# Influence of hydrogenation on structure and magnetic properties of $HoFe_{11-x}Co_xTi$

N. Tajabor [a], M.R. Alinejad [a], L. Motevalizadeh [a,b,*], D. Fruchart [c], E.K. Hlil [c], D. Gignoux [c], S. Miraglia [c]

[a] *Department of Physics, Ferdowsi University of Mashhad, Mashhad 91775-1436, Iran*
[b] *Azad University of Mashhad, Mashhad, Iran*
[c] *Néel Institut, CNRS, BP 166, 38042 Grenoble Cedex 9, France*

Received 23 March 2007; received in revised form 10 April 2007; accepted 16 April 2007
Available online 19 April 2007

**Abstract**

Magnetic properties and crystal structure of the hydrides of ferromagnetic compounds $HoFe_{11-x}Co_xTi$ ($x = 1, 2, 4, 6, 7, 11$) are investigated. The crystal structure was determined by X-ray diffraction (XRD) analysis and the magnetization was measured in applied magnetic fields up to 10 T and at temperatures ranging from 5 K to room temperature. Results show that the crystal structure of the hydrides is the same as for parent compounds but with a moderate unit cell increase. Other properties such as saturation magnetization are affected by H insertion within the lattice. The effect of hydrogenation on magnetic anisotropy energy leads to disappearance of the FOMPs observed in the parent compounds.


*Keywords:* Magnetic properties; Intermetallic compounds; Metal hydrides; $ThMn_{12}$ structure

## 1. Introduction

The intermetallic compounds with formula $RFe_{11-x}M_x$ in which M = Ti, Cr, V, or Mo crystallize in a tetragonal structure with space group *I*4/*mmm* ($ThMn_{12}$ type structure) with two formula units per unit cell. Two main interests of these series of 1:12 type compounds as potential hard magnets are the high Fe content thus favoring a high magnetization level and a relatively high Curie temperature, in particular for the Ti containing compounds.

Insertion of light elements such as H, C, and N in the $RFe_{11-x}M_x$ compounds was found to induce marked changes on the magnetic properties. During the past 10 years, large efforts were devoted to the 1:12 type hydrides, more particularly on the characterization of $RFe_{11}TiH$ systems [1–5]. Parallel, the effect of Co substitution for Fe on the magnetic properties of $RFe_{11-x}M_x$ series were studied as well [6–12]. In particular the influence of Co substitution for Fe was investigated in the here-concerned $HoFe_{11-x}Co_xTi$ compounds [13]. Co substitution for Fe in these compounds leads to decrease the lattice parameters and has a beneficial effect in increasing the Curie temperature. The aim of this paper is to study the influence of hydrogenation on the crystal structure and the magnetic properties of $HoFe_{11-x}Co_xTi$.

## 2. Experimental

The parent compounds were prepared by arc melting good purity elements (≥3 N) Ho, Fe, Co, Ti under high purity (4N5) Ar atmosphere. To achieve homogeneity of the ingots, the samples were annealed at 1050 °C for 12 days in evacuated silica ampoules.

The hydrides were prepared by inserting ground powder samples in stainless steel autoclaves filled with pure hydrogen gas (5N5) at 20 bar pressure thus maintained at 200 °C for 20 h. The hydrogen content was determined by gravimetric method and the lattice parameters were derived from X-ray diffraction patterns on powdered samples, recorded at room temperature with Cu(Kα) radiation graphite monochromatised in backscattering Bragg–Brentano geometry, the record step being of 0.05°. The lattice parameter refinements were carried out by using a profile refinement analysis.

* Corresponding author at: Department of Physics, Ferdowsi University of Mashhad, Mashhad 91775-1436, Iran. Tel.: +98 511 8795162; fax: +98 511 8795560.
*E-mail addresses:* lmotevali@science1.um.ac.ir, lmotevali@yahoo.com (L. Motevalizadeh).

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doi:10.1016/j.jallcom.2007.04.213

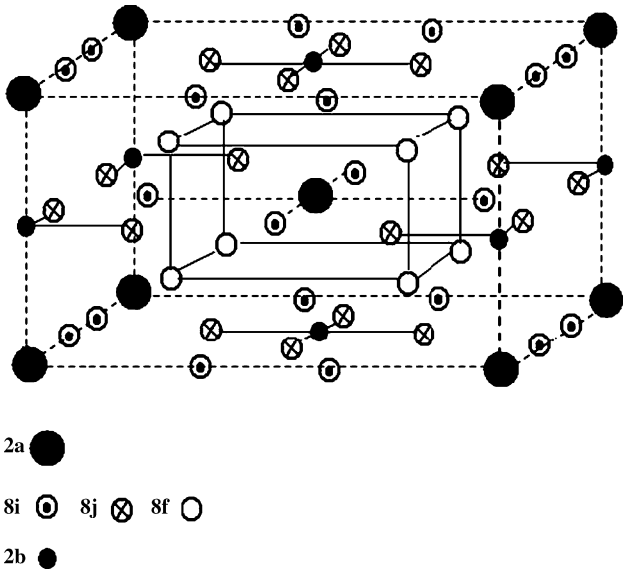


Fig. 1. Schematic representation of the $RTM_{12}X$ unit cell (R refers to rare-earth, TM to transition metal and X to interstitial atoms). R atoms occupy 2a, TM atoms occupy 8i, 8j and 8f and X atoms occupy 2b sites.

Magnetization measurements performed between 5 and 300 K and up to 10 T fields were undertaken using an extraction type magnetometer. The saturation magnetizations were deduced plotting $M$ versus $1/H^2$ [14].

## 3. Results and discussion

### 3.1. Cell parameter analysis

It is well known that in $ThMn_{12}$ type of structure the Ho atoms occupy the origin and the center of the tetragonal cell (2*a*). The Fe atoms totally or partly occupy one or more of the positions 8i (*x*,0,0), 8j (*x*,0,1/2) and 8f (1/4,1/4,1/4) as shown in Fig. 1. Because of a most favorable heat of formation of Fe–Ti than Ho–Ti binaries, the Ti atoms occupy preferentially the 8i sites [3]. Moreover since the atomic site of 8f has the smallest volume reference to those of 8i and 8j sites, the Co atoms for increasing Co content occupy 8f sites, however then they occupy 8j and 8i sites, respectively to form energetically favorable Co–Ho bonds [15]. Tomey et al. reported that hydrogen atoms occupy the octahedral 2b site in Er(Fe, M)$_{12}D_x$ [16]. But later reported results indicate that small amounts of hydrogen atoms could occupy either the 16l or the 32o sites [17].

Analysis of X-ray diffraction patterns show that all samples crystallize in the characteristic tetragonal $ThMn_{12}$ structure with space group *I4/mmm*. Results of the structural data are presented in Table 1 and displayed in Fig. 2(a–c). As can be seen, hydrogen insertion experimentally corresponding to ~1H/at per formula unit, leads to increase both the cell parameters, a being

Table 1
Crystal lattice parameters *a* and *c*, unit cell volume *V*, and saturation magnetization for the $HoFe_{11-x}Co_xTi$ ($x$ = 1, 2, 4, 6, 7, 11) compounds and their hydrides at room temperature

| Compounds | $a$ (Å) | $c$ (Å) | $V$ (Å$^3$) | $\Delta V/V$ (%) | $M_s$ ($\mu_B/f \times u$) |
|---|---|---|---|---|---|
| $HoFe_{10}CoTi$ | 8.499 | 4.777 | 345.057 | – | 14.88 |
| $HoFe_{10}CoTiH$ | 8.524 | 4.783 | 347.526 | 0.716 | 15.71 |
| $HoFe_9Co_2Ti$ | 8.495 | 4.773 | 344.445 | – | 15.61 |
| $HoFe_9Co_2TiH$ | 8.511 | 4.780 | 346.249 | 0.524 | 15.77 |
| $HoFe_7Co_4Ti$ | 8.468 | 4.757 | 341.110 | – | 15.99 |
| $HoFe_7Co_4TiH$ | 8.478 | 4.763 | 342.348 | 0.363 | 15.97 |
| $HoFe_5Co_6Ti$ | 8.435 | 4.743 | 337.461 | – | 16.07 |
| $HoFe_5Co_6TiH$ | 8.462 | 4.748 | 339.983 | 0.747 | 15.38 |
| $HoFe_4Co_7Ti$ | 8.422 | 4.736 | 335.925 | – | 14.39 |
| $HoFe_4Co_7TiH$ | 8.440 | 4.742 | 337.789 | 0.555 | 13.87 |
| $HoCo_{11}Ti$ | 8.382 | 4.709 | 330.845 | – | 11.62 |
| $HoCo_{11}TiH$ | 8.395 | 4.714 | 332.224 | 0.412 | 11.31 |

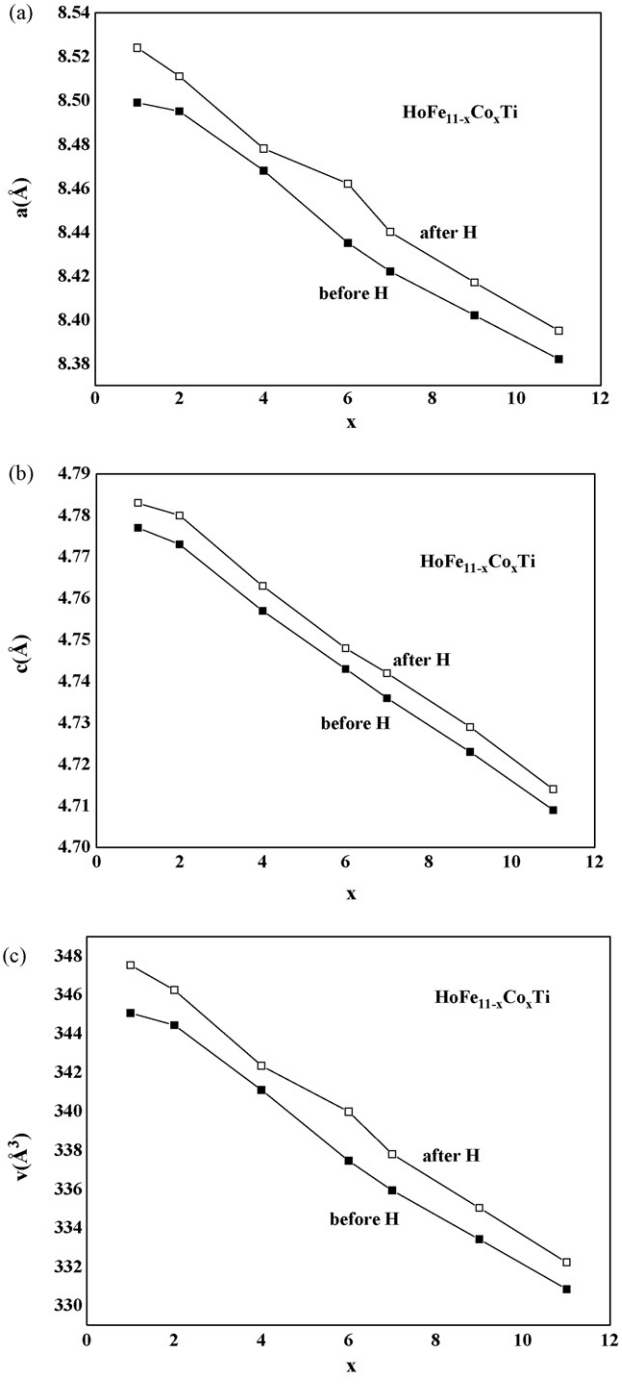


Fig. 2. (a–c) Lattice parameters (a and c) and unit-cell volume ($V$) for the parent and the hydride compounds vs. $x$.