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Structural and magnetic study of new YFe_2D_x compounds ($0<x\leq3.5$)

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Abstract

Deuterium absorption by YFe_2 at 408 K allows us to obtain several single phase YFe_2D_x deuterides for $1.2\leq x\leq2.9$. For $x=1.2$, 1.75 and 1.9, superstructure lines indicated structural deviations from the ideal C15 cubic structure. For $x=2.55$, 2.7 and 2.9 the deuterides were found to crystallize in the C15 type structure. For $x\geq3$ a rhombohedral distortion is observed. These deuterides were studied by neutron diffraction, bulk magnetic and ^{57}Fe Mössbauer experiments. The existence of deuterides with different crystallographic and magnetic structures explains the existence of two phase regions and several plateau pressures in the isotherms at 373 K and 408 K.

Keywords: Deuterides; Laves phases; X-ray and neutron diffraction; Magnetism; Mössbauer

1. Introduction

The study of thermodynamic, structural and magnetic properties of RFe_2 hydrides has raised interest since several years. The RFe_2H_x compounds (R=Tb, Fe, Er, Lu) prepared by hydrogen absorption at room temperature and for $1.2<x<3.2$ have the same cubic structure as the RFe_2 parent compound, with a continuous increase of the cell parameter [1]. For larger hydrogen content a rhombohedral distortion occurs. Therefore the plateaus, which are observed in the isotherms of the RFe_2 compounds in the range $1.2<x<3.2$ were not explained [2]. A hydrogen filling of interstitial sites (A2B2 and AB3) with different binding energies was proposed to explain this multi-plateau behaviour, but it was not clearly related to the evolution of the isotherms [3].

In this paper, the structural evolution of YFe_2D_x compounds prepared by deuterium absorption at 408 K is compared to the evolution of the P–C isotherms at 373, 408 and 473 K. Moreover, some deuterides presenting structural deviations from the ideal cubic C15 structure were studied by neutron diffraction as a function of temperature. The single phase deuterides were also characterized by bulk magnetic and Mössbauer measurements. These results are discussed in order to explain the multiplateau behaviour observed in the YFe_2–D_2 system.

2. Experimental

The preparation and characterization by X-ray diffraction and bulk magnetic measurements of YFe_2 and YFe_2D_x ($1<x<3.5$) compounds is described in [4]. The neutron diffraction patterns were registered on the D1B diffractometer ($\lambda=2.52$ Å) at the Institut Laue Langevin in Grenoble. The ^{57}Fe Mössbauer experiments were performed at 4.2 K and room temperature.

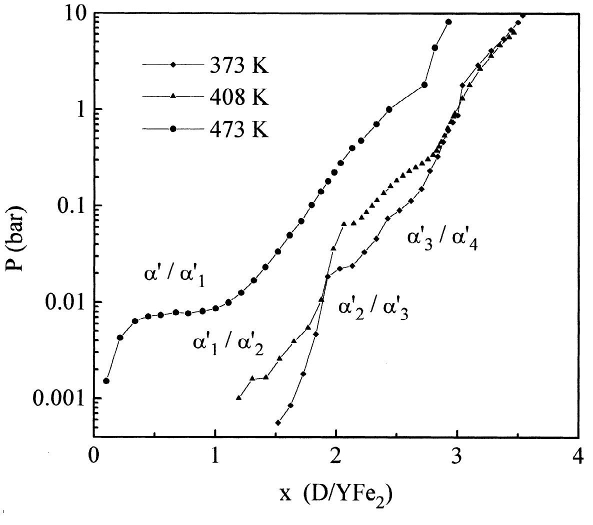

Fig. 1. Absorption isotherms of YFe_2.

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Table 1
X-ray diffraction and magnetic results obtained at room temperature for single phase deuterides

x (D f.u.$^{-1}$)	T_a (K)	Space group	Cell parameters		$\Delta V/V$ (%)	M_s (uem g^{-1}) (at 295 K)
			a (Å)	c (Å)		
0		*Fd-3m*	7.360 (1)		0	61 (1)
1.2	408	*I-4*	11.977 (2)	7.613 (1)	9.74	72 (1)
1.75	408	*I-43m*	15.34 (2)		13.2	71 (2)
1.9	408	cubic *P*	15.378 (2)		14.0	77 (1)
2.55	408	*Fd-3m*	7.783 (1)		18.2	63 (1)
2.7	408	*Fd-3m*	7.810 (1)		19.4	63 (2)
2.9	408	*Fd-3m*	7.828 (1)		20.3	40 (1)
3.5	298	*R-3m*	5.627 (1)	13.344 (1)	22.0	27 (1)

T_a is the absorption temperature and M_s the saturation magnetization.

3. Results and discussion

The pressure-composition absorption isotherms measured at 373, 408 and 473 K are displayed in Fig. 1. Several plateaus are observed in the same range of hydrogen content as in the absorption isotherms of $ErFe_2H_x$. When increasing the temperature the width of the plateaus decreases, and at 473 K no plateau is observed for $1.2<x<2.5$. The existence of different plateaus at 373 and 408 K suggests the existence of deuterides with different structures. At 473 K for $1<x<2.5$ the isotherm displays a continuous increase of the pressure, indicating a solid solution behaviour.

These results lead us to study the structural evolution of the deuterides as a function of both deuterium content and temperature. Deuterium absorption at 408 K allows to obtain single phase deuterides with different structures for $x<3.3$. Structural deviations from the cubic C15 structure were observed by X-ray diffraction at 300 K: a tetragonal distortion with $a'=a.\sqrt{5/2}$ and $c'=a$ was found for $YFe_2D_{1.2}$, whereas superstructure lines leading to a doubling of the cubic cell parameter were observed for $YFe_2D_{1.75}$ and $YFe_2D_{1.9}$. Deuterides with C15 structure (within the accuracy of our measurements) were obtained for $2.55\leq x\leq 2.9$. A rhombohedral distortion appears for $x>3$. The cell parameters (at 300 K) and the space groups of these deuterides are given in Table 1. The evolution of the cell volume as a function of D content (Fig. 2) indicates the existence of several two phase regions in agreement with the plateaus observed in the absorption isotherms at 373 and 408 K.

Concerning the influence of the temperature, in situ neutron diffraction experiments have shown that for $x=1.2$ and 1.9 D/f.u. the superstructure lines disappear for temperatures higher than 460 and 440 K, respectively. Above these temperatures neutron diffraction patterns of

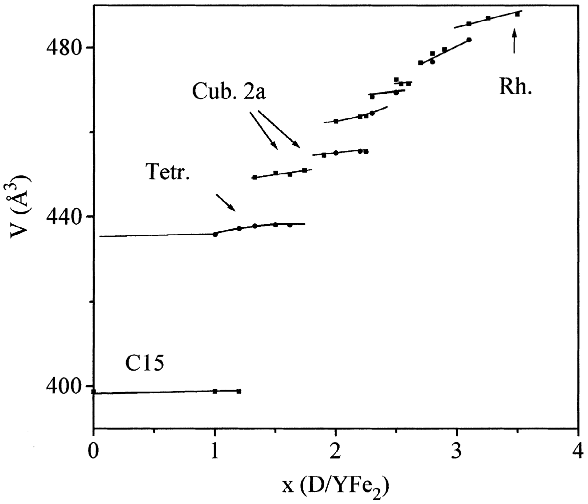

Fig. 2. Evolution of the cell volume (equivalent to C15) as a function of deuterium content for YFe_2D_x.

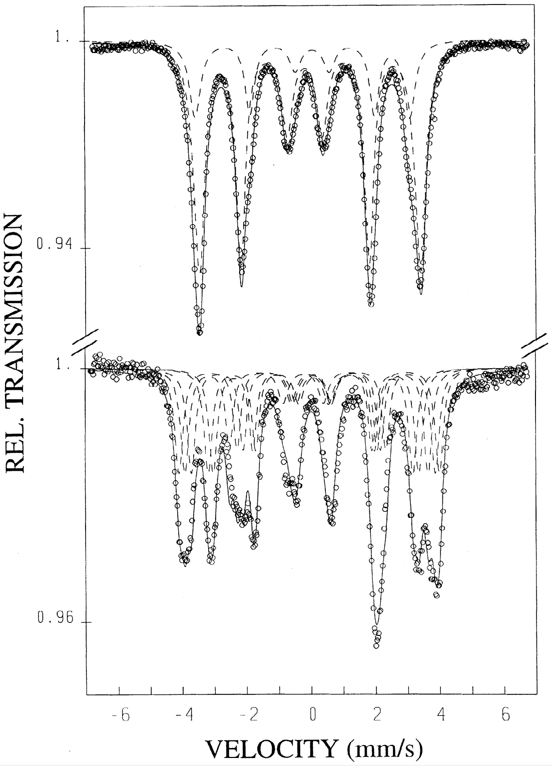

Fig. 3. ^{57}Fe Mössbauer spectra recorded at 4.2 K from YFe_2 (top) and $YFe_2D_{1.2}$ (bottom) (∘∘∘) experimental (- - - and — — — fit).

the C15 type structure were obtained and these structural transformations were found to be reversible. This allows us to explain the disappearance of plateaus for $1.2<x<2.5$ in the isotherm at 473 K, since at this temperature all the deuterides have the same C15 structure. In this case a progressive filling of A2B2 sites should occur, like in the YMn_2–H_2 system above the transition temperature [5].

All the YFe_2D_x deuterides were found to be ferromagnetic at room temperature. Their saturation magnetization increases from YFe_2 to $YFe_2D_{1.9}$, and decreases for larger deuterium content.

The Mössbauer spectra demonstrate the significant change of crystallographic and magnetic structure upon deuterium absorption. In agreement with the tetragonal structure of $YFe_2D_{1.2}$ five non equivalent Fe sites have to be taken into account to fit the spectrum (Fig. 3). Under this assumption the spectra with a deuterium content up to 1.9 D/f.u. can be correctly fitted. For larger deuterium amount the spectra increasingly broaden, no longer permitting an unambiguous interpretation. All these structural deviations could result from magnetostrictive properties induced by deuterium absorption.

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