

Magnetic properties of $\text{RFe}_{11.35}\text{Nb}_{0.65}\text{C}_y$ (R identical to Y, Gd, Tb, Dy, Ho, or Er)

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1995 J. Phys.: Condens. Matter 7 5365

(<http://iopscience.iop.org/0953-8984/7/27/021>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.151

The article was downloaded on 12/05/2010 at 21:39

Please note that [terms and conditions apply](#).

Magnetic properties of $RFe_{11.35}Nb_{0.65}C_y$ ($R \equiv Y, Gd, Tb, Dy, Ho, \text{ or } Er$)

Kai-Ying Wang[†], Yi-Zhong Wang[‡], Bo-Ping Hu[‡], Wu-Yan Lai[†] and Zhen-Xi Wang[‡]

[†] Magnetism Laboratory, Institute of Physics, Chinese Academy of Sciences, PO Box 603, Beijing 100080, People's Republic of China

[‡] San Huan Research Laboratory, Chinese Academy of Sciences, PO Box 603, Beijing 100080, People's Republic of China

Received 9 December 1994, in final form 21 February 1995

Abstract. A new series of the carbides $RFe_{11.35}Nb_{0.65}C_y$ ($R \equiv Y, Gd, Tb, Dy, Ho \text{ or } Er$) ($y \simeq 1.0$) with the $ThMn_{12}$ structure was synthesized by gas–solid-phase reaction at 773 K. The unit-cell volumes of the carbides increase upon carbonation by nearly 4.0% across the series. The average enhancement of the Curie temperature is about 160 K. The highest Curie temperature is 733 K for the carbide $GdFe_{11.35}Nb_{0.65}C_y$. The saturation magnetization of the Fe sublattice is $22.5\mu_B \text{ FU}^{-1}$ at 4.2 K and $20.7\mu_B \text{ FU}^{-1}$ at room temperature. The maximum value of the anisotropy field found at room temperature for $DyFe_{11.35}Nb_{0.65}C_y$ is about 8 T.

1. Introduction

Ternary rare-earth compounds based on the tetragonal $ThMn_{12}$ -type structure are fairly common. These compounds have the chemical composition $RFe_{12-x}M_x$ ($1.0 < x < 4.0$), where M represents elements such as Al, Ti, V, Cr, Mo, W, Si or Re [1]. It has been found that the magnetic properties of these intermetallic compounds $RFe_{12-x}M_x$ are drastically modified by nitrogenation or carbonation [2–5]. The interstitial nitrogen and carbon atoms not only have the effect of increasing the Curie temperature and saturation magnetization but also give rise to a profound change in the magnetocrystalline anisotropy.

In a previous investigation [6] we pointed out that the $ThMn_{12}$ -type rare-earth iron-rich compounds could also be stabilized by a third stabilizing element Nb according to the diagonal position relationship in the element periodic table [7]. We synthesized a new series of $ThMn_{12}$ -type compounds $RFe_{12-x}Nb_x$ for which the value of x is in the range $0.5 < x < 0.8$ and investigated the magnetic properties of the compounds $RFe_{11.35}Nb_{0.65}$ and their nitrides [8]. However, it is worth studying the effect of interstitial carbon atoms on the magnetic properties of the compounds $RFe_{11.35}Nb_{0.65}$. In this paper, the structure and the intrinsic magnetic properties of the carbides $RFe_{11.35}Nb_{0.65}C_y$ ($R \equiv Y, Gd, Tb, Dy, Ho \text{ or } Er$) are reported.

2. Experimental details

The samples $RFe_{11.35}Nb_{0.65}$ ($R \equiv Y, Gd, Tb, Dy, Ho \text{ or } Er$) were prepared by arc melting 99.9% pure constituent elements in an argon atmosphere. The ingots were turned over

several times to ensure homogeneity. The carbides were synthesized by heat treatment of the pulverized powder (particle diameter, less than $20\ \mu\text{m}$) in acetylene at about $773\ \text{K}$ for 3 h. In order to avoid the hydrogen being absorbed into the compound, the sample space was pumped before cooling. The phase composition and purity were identified by x-ray diffraction and a thermomagnetic scan at a field of $0.04\ \text{T}$. The latter method was also used to determine the Curie temperature. The saturation magnetizations were derived from the magnetization curves on free powder in the field up to $6\ \text{T}$ with an extracting-sample magnetometer. The values of the anisotropy fields were derived from the magnetization curves both parallel and perpendicular to the aligned direction on oriented samples and the singular point detection (SPD) technique at room temperature. The carbon content, estimated with the Perkin-Elmer 2400 CHN element analyser, is about 1 carbon atom per formula for all the samples.

3. Results and discussion

Compared with their parent alloys [8], the carbides $\text{RFe}_{11.35}\text{Nb}_{0.65}\text{C}_y$ retain the ThMn_{12} -type structure (space group, $I4/mmm$), but all the peaks shift to smaller-angle positions. Typical x-ray diffraction patterns for Er alloy and its carbide are shown in figure 1.

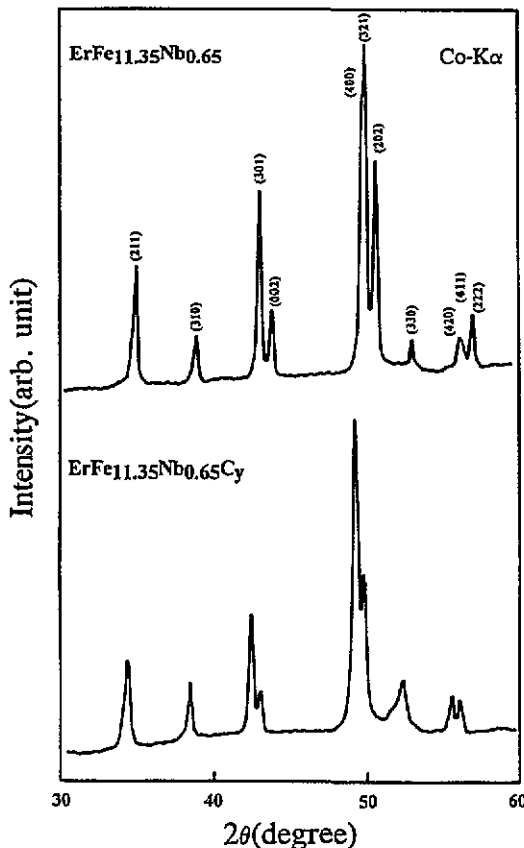


Figure 1. X-ray diffraction patterns of $\text{ErFe}_{11.35}\text{Nb}_{0.65}$ and $\text{ErFe}_{11.35}\text{Nb}_{0.65}\text{C}_y$.

The least-squares method was used to calculate the lattice parameters. The values of a and c and the unit-cell volumes V , together with relative volume expansion $\Delta V/V\%$, are listed in table 1 and plotted in figure 2. It can be seen that the lattice parameters a and c and the unit-cell volumes V for both the hosts and their carbides decrease with increasing atomic number of the rare-earth R atom owing to lanthanide contraction. Considering the crystallographic relationship between the tetragonal $ThMn_{12}$ -type structure of $RFe_{11.35}Nb_{0.65}$ and the fact that the unit-cell volume expands by nearly 4.0%, it is reasonable to be assumed that the $2b$ interstitial vacancies will become occupied as proved for $NdFe_{10}Mo_2N_{0.5}$ by neutron diffraction experiments [9].

Table 1. Structural data and magnetic properties of $RFe_{11.35}Nb_{0.65}$ and $RFe_{11.35}Nb_{0.65}C_y$.

Compound	a (nm)	c (nm)	V (nm ³)	$\Delta V/V$	T_c (K)	M_s (μ_B f.u. ⁻¹)		B_A (T)	EMD
						4.2 K	293 K		
$YFe_{11.35}Nb_{0.65}$	0.8511	0.4790	0.3470		526	21.3	18.0	2.1	c axis
$YFe_{11.35}Nb_{0.65}C$	0.8589	0.4884	0.3600	3.7	685	22.5	20.7	<0.5	c axis
$GdFe_{11.35}Nb_{0.65}$	0.8543	0.4804	0.3510		597	15.0	14.2	3.6	c axis
$GdFe_{11.35}Nb_{0.65}C$	0.8573	0.4949	0.3689	3.6	733	17.5	17.1	0.62	c axis
$TbFe_{11.35}Nb_{0.65}$	0.8532	0.4798	0.3493		556	13.1	13.4	—	Plane
$TbFe_{11.35}Nb_{0.65}C$	0.8566	0.4938	0.3623	3.6	713	13.4	16.6	7.4	c axis
$DyFe_{11.35}Nb_{0.65}$	0.8521	0.4800	0.3485		536	12.2	13.3	<0.5	c axis
$DyFe_{11.35}Nb_{0.65}C$	0.8565	0.4935	0.3639	4.2	699	12.6	14.1	8.0	c axis
$HoFe_{11.35}Nb_{0.65}$	0.8518	0.4798	0.3481		520	11.0	12.8	2.5	c axis
$HoFe_{11.35}Nb_{0.65}C$	0.8586	0.4902	0.3614	3.7	686	14.7	17.1	4.9	c axis
$ErFe_{11.35}Nb_{0.65}$	0.8486	0.4794	0.3452		507	11.7	15.2	2.7	c axis
$ErFe_{11.35}Nb_{0.65}C$	0.8580	0.4878	0.3590	3.9	673	15.3	17.8	—	Plane

The Curie temperatures of parent alloys and their carbides were determined from thermomagnetic curves at a field of 0.04 T. The values of T_c are listed in table 1 and shown in figure 3. The largest values of T_c are found for the gadolinium alloy and its carbide, since Gd^{3+} has the largest spin moment. After carbonation, the interstitial carbon atoms have a large effect in increasing the Curie temperature. The average increase in T_c is about 160 K. This may be attributed to the increase in Fe–Fe distance induced by crystalline expansion. The values of the increase in Curie temperature for carbides are about 40–50 K lower than those of the corresponding nitrides [8], but in accordance with those found for other 1:12 carbides [3].

The Fe–Fe and R–Fe exchange interactions can be deduced from the molecular-field approximation from the expression for the magnetic ordering temperature [10]. Using the same calculation procedure as in [8], we obtain the Fe–Fe exchange interaction coefficient $n_{FeFe} = 304\mu_0$ ($T_c = 685$ K) for $YFe_{11.35}Nb_{0.65}C$ or $n_{FeFe} = 294\mu_0$ ($T_c = 657$ K) for $LuFe_{11.35}Nb_{0.65}C$. Because of the effect of interstitial C atoms, the exchange interactions of the Fe sublattice in $RFe_{11.35}Nb_{0.65}$ ($R \equiv Y$ or Lu) is enhanced by about 37%, which is less than that found in the case of the corresponding nitrides [8]. Neglecting the R–R interactions and taking the Fe-sublattice ordering temperature $T_{Fe} = 657$ K for $LuFe_{11.35}Nb_{0.65}C$ and the average volume of the $RFe_{11.35}Nb_{0.65}C$ series as 0.1816 nm³, we obtain values of n_{RFe} in the range $(165-181)\mu_0$ and the average value is $\langle n_{RFe} \rangle = 176\mu_0$ (for heavy rare earths). This value is slightly larger than that of $\langle n_{RFe} \rangle = 160\mu_0$ for the series of parent compounds. This means that the R–Fe exchange interaction in $RFe_{11.35}Nb_{0.65}$ is slightly strengthened by interstitial C atoms, which is different from the weakening case by the interstitial N atoms

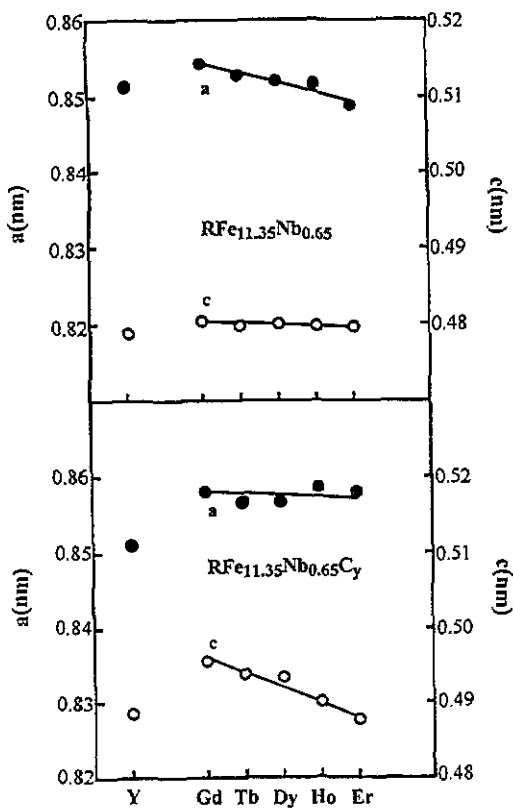


Figure 2. Lattice parameters a and c of $RFe_{11.35}Nb_{0.65}Cy$.

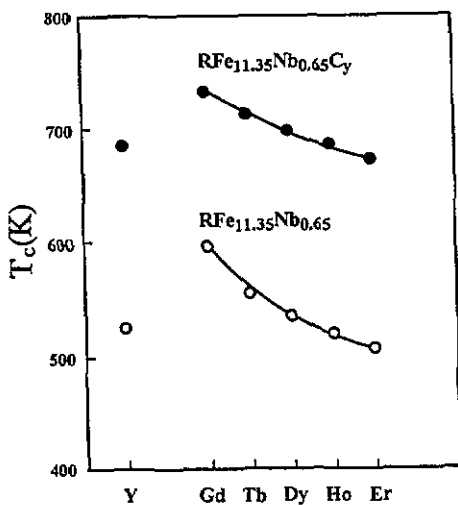


Figure 3. Curie temperatures of $RFe_{11.35}Nb_{0.65}$ and $RFe_{11.35}Nb_{0.65}Cy$.

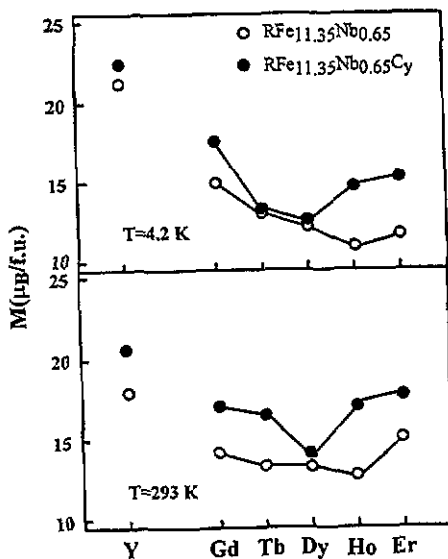


Figure 4. Saturation magnetizations of $RFe_{11.35}Nb_{0.65}$ and $RFe_{11.35}Nb_{0.65}Cy$.

(n_{RFe}) = $147\mu_0$) [8]. However, the strengthened Fe–Fe exchange interactions substantially contribute to the enhancement of the Curie temperature by carbonation or nitrogeation.

The saturation magnetizations of the $RFe_{11.35}Nb_{0.65}$ alloys and their carbides at 4.2 and 293 K are listed in table 1 and presented in figure 4. It can be seen that the saturation magnetizations increased after carbonation. From the saturation magnetization of the carbides $YFe_{11.35}Nb_{0.65}C_y$, it can be derived that the average moment of Fe atom is $1.98\mu_B$ at 4.2 K and $1.82\mu_B$ at 293 K. Compared with parent alloys, the relative increase in Fe atom moment is 5.6% which is lower than 7.3% for $YFe_{11}TiC_y$ [3] and 11.2% for $YFe_{11.35}Nb_{0.65}N_y$ [8]. For 1:12 carbides, the interstitial carbon atoms induced an increase in the magnetization of the Fe sublattice, but they have little effect on that of the Fe sublattice in 2:17 carbides [11, 12]. Although the structures of both the 1:12 and the 2:17 compounds originate from $CaCu_5$, the crystal symmetries of these compounds are different. This difference may be understood as arising from the different effects of interstitial carbon on the 3d band of 1:12 and 2:17 compounds.

The nature of the magnetocrystalline anisotropy of the carbides $RFe_{11.35}Nb_{0.65}C_y$ were determined from the aligned x-ray diffraction patterns, magnetization curves parallel and perpendicular to the aligned direction and the SPD technique at room temperature. Their easy-magnetization directions (EMDs) and the values of the magnetocrystalline anisotropy field are listed in table 1. It can be seen in table 1 that the values of the magnetocrystalline anisotropy field of Y, Gd alloys decrease after carbonation. This means that the interstitial carbon atoms weaken the magnetocrystalline anisotropy of the Fe sublattice. Similar phenomena have also been observed for $YFe_{11}TiC_y$ [3] and $YFe_{10}V_2N_y$ [2] and $YFe_{12-x}Mo_xN_y$ [13].

For 1:12-type nitrides or carbides, the sign of A_{20} for the rare-earth sublattice will change from negative for the alloys to positive for the nitrides or carbides, while the absolute values increase. This change in A_{20} causes the sublattice of the rare-earth ions Tb^{3+} , Dy^{3+} and Ho^{3+} with a negative second-order Stevens coefficient $\alpha_J < 0$ to possess an easy c axis; however, the sublattice of Er^{3+} with a positive $\alpha_J > 0$ possesses an easy plane after carbonation [14]. At room temperature, the magnetocrystalline anisotropy field B_a of the Tb carbide is 7.4 T, and that of the Dy carbide is 8 T, which are lower than those of their corresponding nitrides [8]. The change in the magnetocrystalline anisotropy of Er carbide is also similar to that of Er nitride [8]. Using the exchange interaction and crystal-field model [15, 18], and taking the temperature dependence of the Fe-sublattice magnetization and anisotropy constant from $YFe_{11.35}Nb_{0.65}C$, exchange coefficient $n_{RFe} = 176\mu_0$ and second-order crystal-field coefficient $A_{20} = 80K a_0^{-2}$, we calculated the anisotropy field B_a at room temperature for $RFe_{11.35}Nb_{0.65}C$ ($R \equiv Tb, Dy$ or Ho). The values of B_a are 8.0 T, 6.0 T and 2.0 T for the Tb, Dy and Ho carbides, respectively, in reasonable agreement with the experimental data of 7.4 T, 8.0 T and 4.9 T. The value of $A_{20} = 80K a_0^{-2}$ here is only half that of $160K a_0^{-2}$ found for the $RFe_{11.35}Nb_{0.65}N_y$ ($R \equiv Tb$ or Dy) nitrides. This indicates that the interstitial C has less effect on the crystal-field interaction than the interstitial N does. A similar saturation has also been found for $Sm_2Fe_{17}C_y$ and $Sm_2Fe_{17}N_y$ [16].

4. Conclusions

In the present work, we synthesized a new series of carbides $RFe_{11.35}Nb_{0.65}C_y$ ($R \equiv Y, Gd, Tb, Dy, Ho$ or Er) by gas–solid–phase reaction at 773 K. The carbides retain the same $ThMn_{12}$ -type structure as their parent alloys, but with an expansion in the unit-cell volume

of nearly 4%. The Curie temperatures of the carbides increase by about 160 K on average. The carbonation weakens the anisotropy field of the Fe sublattice but has great effect on that of the rare-earth sublattice. The B_a of the carbide $\text{DyFe}_{11.35}\text{Nb}_{0.65}\text{C}_y$ reaches about 8 T. Using the exchange interaction and the crystal-field model, the second-order crystal-field coefficient A_{20} was estimated to be $80K a_0^{-2}$.

Acknowledgment

This work was partly supported by the State Key Laboratory for Advanced Metal Materials.

References

- [1] Buschow K H J 1992 *Rep. Prog. Phys.* **54** 1123
- [2] Wang Y Z, Hadjipanayis G C, Kim A, Sellmyer D J and Yelon W B 1992 *J. Magn. Magn. Mater.* **104-7** 1132
- [3] Hurley D P F and Coey J M D 1992 *J. Phys.: Condens. Matter* **4** 5573
- [4] Wang Y Z, Hadjipanayis G C 1991 *J. Appl. Phys.* **70** 6009
- [5] Gong W and Hadjipanayis G C 1992 *IEEE Trans. Magn.* **MAG-28** 2563
- [6] Wang Kai-Ying, Wang Yi-Zhong, Hu Bo-Ping, Lai Wu-Yan And Wang Zhen-Xi 1995 *J. Alloys Compounds* at press
- [7] Gilmore G N 1979 *A Modern Approach to Comprehensive Chemistry* 2nd edn (Stanley Thomas) p 179
- [8] Hu Bo-Ping, Wang Kai-Ying, Wang Yi-Zhong, Wang Zhen-Xi, Yan Qi-Wei, Zhang Pan-Lin and Sun Xiang-Dong 1995 *Phys. Rev. B* **51** 2905
- [9] Wang Y Z, Hadjipanayis G C, Tang Z X, Yelon W B, Papaefthymious V, Moukarika A and Sellmyer D J 1993 *J. Magn. Magn. Mater.* **119** 41
- [10] Belorizky E, Fremy M A, Gavigan J P, Givord D and Li H S 1987 *J. Appl. Phys.* **61** 3971
- [11] Hu Bo-Ping and Liu Gui-Chuan 1991 *Solid State Commun.* **79** 785
- [12] Zhong X P, Radwanski R J, De Boer F R, Jacobs J H and Buschow K H J 1990 *J. Magn. Magn. Mater.* **86** 333
- [13] Wang Yi-Zhong, Hu Bo-Ping, Song Lin, Wang Kai-Ying and Liu Gui-Chun 1994 *J. Phys.: Condens. Matter* **86** 7085
- [14] Li H S and Cadogan J M 1992 *J. Magn. Magn. Mater.* **109** L153
- [15] Cadogan J M, Gavigan J P, Givord D and Li H S 1988 *J. Phys. F: Met. Phys.* **18** 779
- [16] Li H S and Cadogan J M 1992 *Solid State Commun.* **82** 121