Structure and magnetic properties of a solution of carbon in $YFe_{10.5}Mo_{1.5}$ prepared by arc melting

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Abstract. The structural and magnetic properties of a solution of carbon in YFe$_{10.5}$Mo$_{1.5}$ prepared by arc melting have been studied by means of magnetic measurements and the neutron powder diffraction technique. It was found that the carbides retain the ThMn$_{12}$-type structure but with an increase of unit-cell volume. The neutron diffraction results indicate that most of the carbon atoms occupy the interstitial 2b sites and the others occupy the substitutional 8i sites. According to the magnetic measurements, both the Curie temperature and the saturation magnetization are found to increase with the carbon concentration x in the YFe$_{10.5}$Mo$_{1.5}$C$_x$ series, and the magnetocrystalline anisotropy is also affected by the carbon atoms.

1. Introduction

It has been found that the magnetic properties of the ThMn$_{12}$-type compounds can be drastically improved by introducing nitrogen or carbon atoms [1–4]. These non-metallic atoms are introduced by gas–solid-phase reaction and occupy the interstitial 2b sites in ThMn$_{12}$-type structure. As a direct effect of the volume expansion caused by interstitial modifications, the Fe–Fe interatomic distances increase, which enhances the exchange interactions, and consequently the saturation magnetization and Curie temperature increase enormously. Simultaneously, the chemical bonding effects also enhance the Fe–Fe exchange interactions [5]. It was found that adding interstitial atoms slightly decreases the magnetic anisotropy of the iron sublattice [6]. However, the most important contribution to the magnetic anisotropy comes from the rare-earth sublattice. Theoretical studies indicate that the second-order crystal-field parameter A_{20} for the rare-earth site is changed from negative for the host compounds to positive for the interstitially modified nitrides [7, 8]. Experimentally, it was found that the rare-earth ions which possess a negative second Stevens factor α_J, such as Pr, Nd, Tb, Dy, and Ho, exhibit an easy axis upon nitrogenation.

Owing to their excellent intrinsic magnetic properties, the nitrides and carbides of the ThMn$_{12}$-type compounds have been regarded as promising candidates for permanent magnet applications. Unfortunately the nitrides or carbides prepared by gas–solid-phase reaction are metastable compounds and they will decompose at temperatures higher than 700 °C in general. Thus conventional high-temperature powder metallurgy is not applicable to these materials.

Obviously, carbides prepared by the melting method are more stable at high temperatures, since usually they will have been treated at high temperature (800–1100 °C)

for a long time (about one week). However, it is difficult to synthesize single-phase carbides with $ThMn_{12}$-type structure by the melting method, especially when the carbon content is larger than 0.3.

R   Fe   Fe   Fe   X
2a   8i   8j   8f   2b
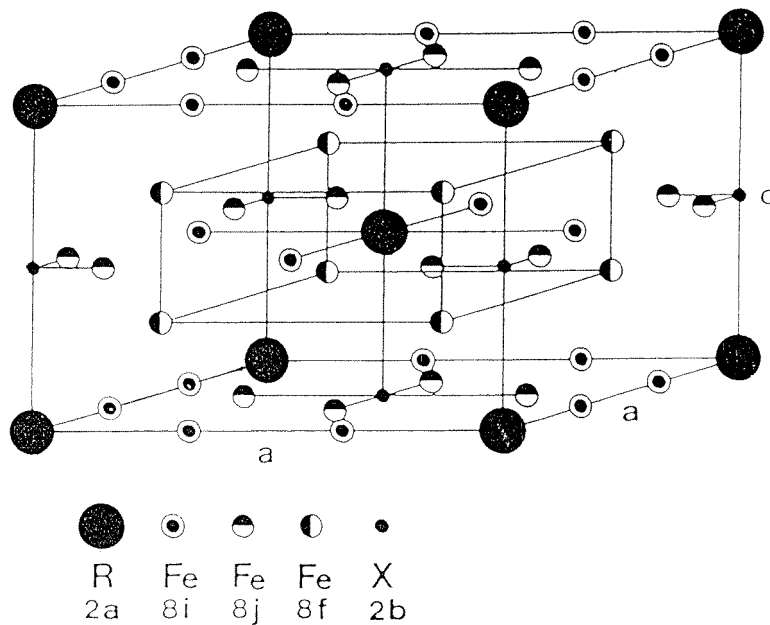
Figure 1. A diagram of the unit cell of the $R(Fe, M)_{12}$ compounds having the $ThMn_{12}$-type body-centred-tetragonal structure.

Yang *et al* studied the effects of carbon on the structural and magnetic properties of $RTiFe_{10.75}C_{0.25}$ [9]. It was found that introducing the carbon atoms via arc melting has no effect on the Curie temperature, but the saturation magnetization is increased and the unit-cell volume is decreased slightly with the addition of carbon. They also reported that carbon atoms increase the planar anisotropy for rare-earth ions with negative second-order Stevens constants α_J, such as Nd^{3+} and Dy^{3+}, and strengthen the axial anisotropy for those with positive α_J, such as Sm^{3+}. The effect of carbon atoms introduced by arc melting is different from that of the interstitial nitrogen or carbon atoms introduced by gas–solid-phase reaction. Zhang *et al* [10] and Hu *et al* [11] published results of neutron diffraction experiments carried out on $YFe_{11}TiC_{0.3}$ and $ErFe_{11}TiC_{0.25}$ prepared by arc melting. They found that all of the carbon atoms occupy 8i sites, not the interstitial 2b sites. Figure 1 is a diagram of the unit cell of $R(Fe, M)_{12}$ compounds having the $ThMn_{12}$-type body-centred-tetragonal structure. Hu *et al* also studied the structure of $YFe_{10.5}Mo_{1.5}C_{0.3}$ and found that it is different from that of $R(Fe, Ti)_{12}C_x$ compounds, though both carbides were prepared by arc melting. They reported that only about 40% of the carbon atoms occupy 8i sites and the others occupy 2b sites in $YFe_{10.5}Mo_{1.5}C_{0.3}$.

Since a proportion of the carbon atoms occupy the 2b interstitial sites in the $RFe_{12-x}Mo_xC_y$ compounds, the interstitial modification becomes significant when the carbon content is sufficiently large [12]. It is imperative to study the structure of $RFe_{12-x}Mo_xC_y$ with a high carbon content.

In this paper, the structure and magnetic properties of $YFe_{10.5}Mo_{1.5}C_x$ ($x = 0$–0.6) are reported.

2. Experimental method

YFe$_{10.5}$Mo$_{1.5}$C$_x$ ($x = 0$, 0.2, 0.3, 0.4, 0.6) compounds were prepared by arc melting 99.5%-pure materials in a purified argon atmosphere. The carbon was introduced as Fe–C alloy. X-ray diffraction was used to determine the structure. The powder sample, of cylindrical shape, was aligned in a 17 kOe field in epoxy resin. Magnetic measurements were performed with a field of up to 20 kOe over a temperature range from 300 K to 1000 K by a vibrating-sample magnetometer (VSM). The neutron diffraction measurements were performed on the powder diffractometer using the heavy-water research reactor at the Institute of Atomic Energy in Beijing. The data were collected by scanning the samples from 7.05° to 88.0° in steps of 0.15° at room temperature, with a neutron wavelength of 1.159 Å. The diffraction patterns were analysed by means of the Rietveld profile technique.

Table 1. The lattice parameters a and c, unit-cell volume V, Curie temperature T_C, saturation magnetization σ_s, and magnetic anisotropy field H_A of YFe$_{10.5}$Mo$_{1.5}$C$_x$.

	a (Å)	c (Å)	V (Å^3)	T_C (K)	σ_s (emu g^{-1})	H_A (kOe)
YFe$_{10.5}$Mo$_{1.5}$	8.540	4.791	349.4	433	78.66	9.33
YFe$_{10.5}$Mo$_{1.5}$C$_{0.2}$	8.546	4.792	350.0	460	87.41	9.69
YFe$_{10.5}$Mo$_{1.5}$C$_{0.3}$	8.555	4.793	350.8	465	90.41	8.61
YFe$_{10.5}$Mo$_{1.5}$C$_{0.4}$	8.560	4.794	351.3	480	95.95	8.05
YFe$_{10.5}$Mo$_{1.5}$C$_{0.6}$	8.584	4.800	353.7	488	101.74	6.81
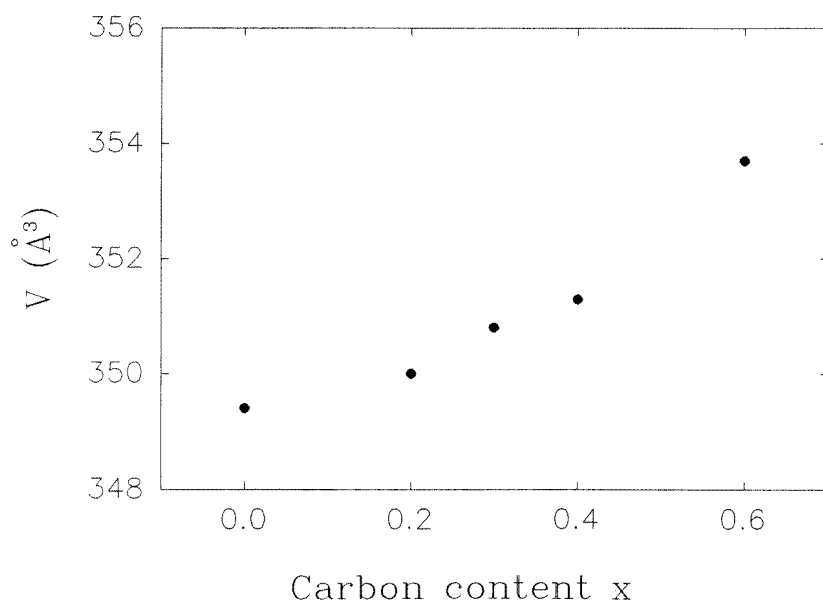
Figure 2. The unit-cell volume (V) of YFe$_{10.5}$Mo$_{1.5}$C$_x$ versus the carbon content.

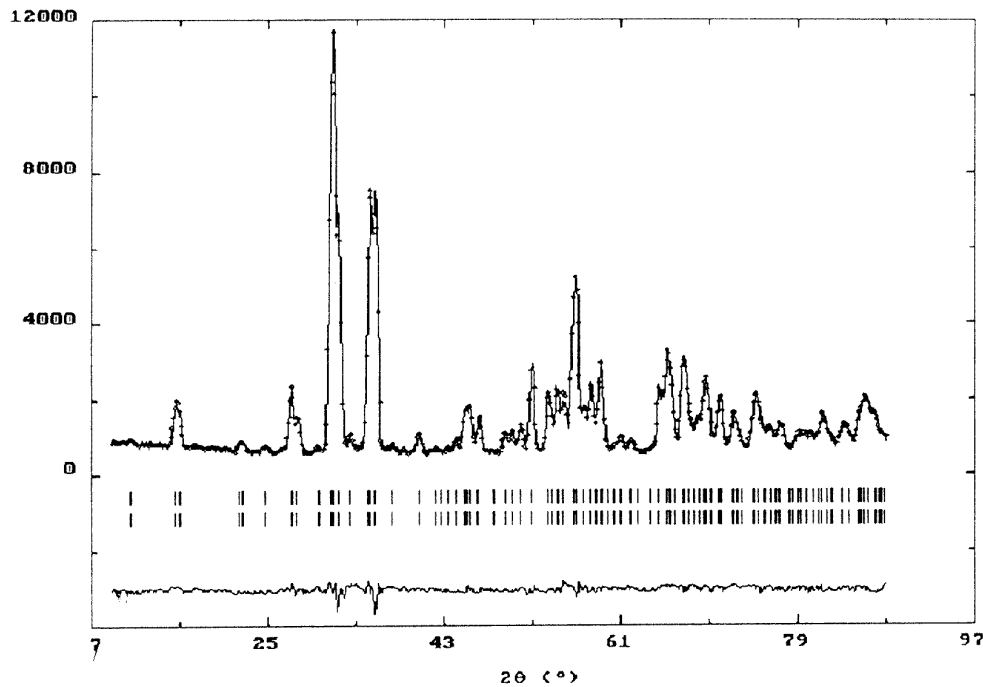**Figure 3.** The neutron diffraction pattern of $YFe_{10.5}Mo_{1.5}C_{0.6}$ at room temperature.

Table 2. Neutron diffraction refinement results for $YFe_{10.5}Mo_{1.5}C_{0.6}$.

Atoms	Site	x	y	z	Occupancy	μ_z (μ_B)
Y	(2a)	0	0	0	2	0
Fe	(8i)	0.358 41(28)	0	0	4.744(208)	1.66(17)
Fe	(8j)	0.277 24(30)	0.5000	0	8	1.80(17)
Fe	(8f)	0.2500	0.2500	0.2500	8	2.14(15)
Mo	(8i)	0.358 41(28)	0	0	3.200(208)	0
C	(8i)	0.358 41(28)	0	0	0.056(208)	0
C	(2b)	0	0	0.5000	0.679(38)	0

$R_p = 7.11\%$, $R_B = 4.27\%$, $R_m = 7.43\%$

3. Results and discussion

3.1. Crystal structure

According to the x-ray diffraction studies and thermomagnetic analysis, all of the samples retain the $ThMn_{12}$-type tetragonal structure, and no α-Fe phase is observed. The lattice parameters are listed in table 1. The cell volume increases with the carbon content (see figure 2), which is different from the case for $R(Fe, Ti)_{12}C_x$ [9]. These results suggest that a significant proportion of the carbon atoms are introduced into the interstitial sites. The

neutron powder diffraction measurements were carried out on $YFe_{10.5}Mo_{1.5}C_{0.6}$. A perfect neutron diffraction pattern was obtained, and it is shown in figure 3. This also indicates that the sample is single phase. The refinement results are listed in table 2. The data show that carbon atoms occupy about 34% of the 2b sites, whereas iron atoms occupy 8j, 8i, and 8f sites, and Mo atoms occupy only the 8i sites. They also suggest that a small proportion of the carbon atoms occupy 8i sites. The total carbon content is only 0.37, and is less than the stoichiometric amount (0.60), which is due to the evaporation during the arc-melting process. The result, that about 92.4% of carbon atoms introduced by the arc-melting method occupy the interstitial 2b sites in the $YFe_{10.5}Mo_{1.5}C_x$, which is different to the results of Hu *et al*, leads to obvious effects on the magnetic properties of the carbides of $Y(Fe, Mo)_{12}$ prepared by arc melting.
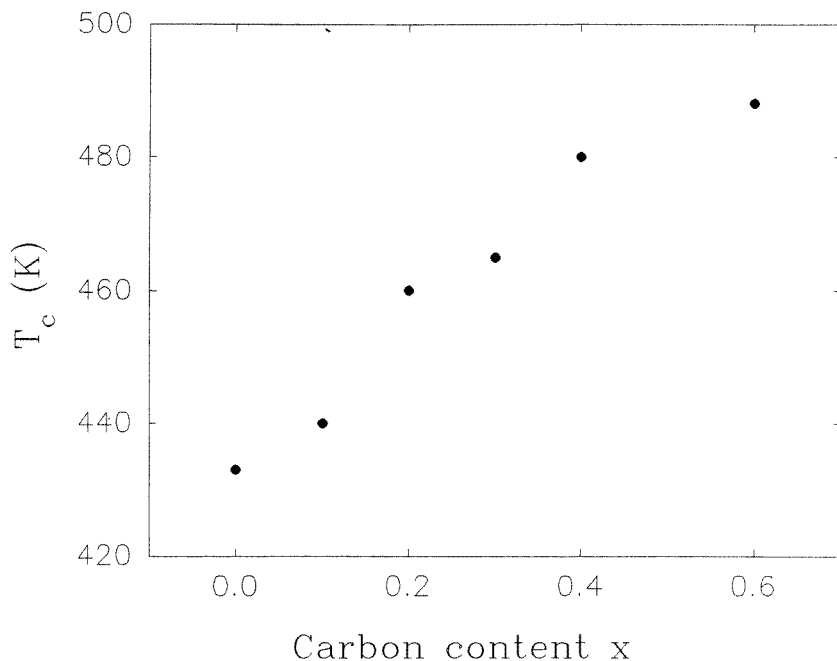
Figure 4. The Curie temperature (T_C) of $YFe_{10.5}Mo_{1.5}C_x$ versus the carbon content.

3.2. Magnetic properties

Table 1 lists the Curie temperatures (T_C), saturation magnetizations (σ_s), and magnetic anisotropy fields (H_A) of $YFe_{10.5}Mo_{1.5}C_x$ ($x = 0.0, 0.2, 0.3, 0.4,$ and 0.6). The arc-melting-introduced carbon atoms have the effect of increasing the Curie temperature and the saturation magnetization. The variation of T_C with the carbon content (x) is illustrated in figure 4. Since yttrium is non-magnetic, the moment of the yttrium compounds represents that of the Fe sublattice. The introduction of carbon increases the average magnetization of Fe atoms in the lattice. The variation of the average Fe magnetic moments as a function the carbon content (x) is illustrated in figure 5. The results were extrapolated to $x = 1$, and, at that point, the average Fe moment is about 1.65 μ_B at room temperature. The average Fe moment in the $YFe_{10.5}Mo_{1.5}N$ is 1.60 μ_B (represented by the solid dot in figure 5) [12].
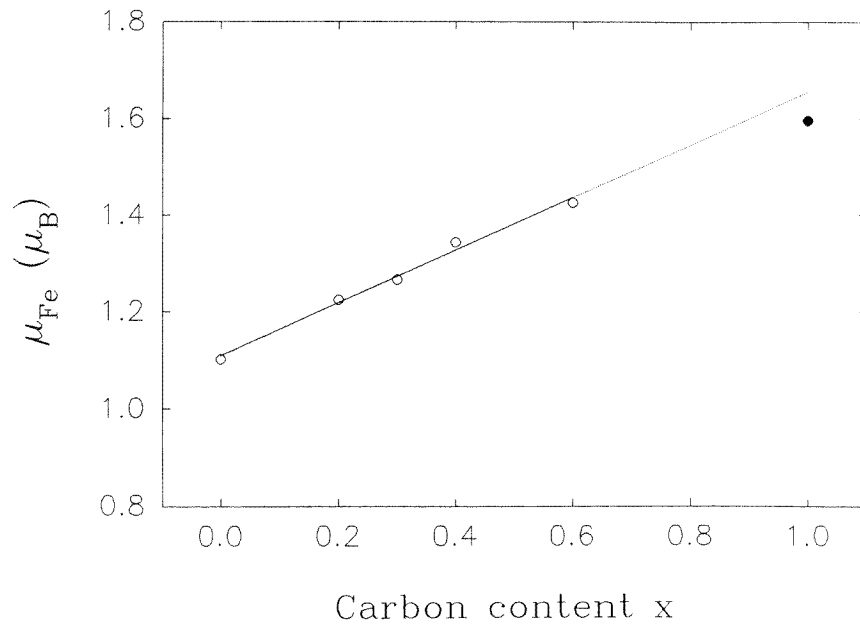
Figure 5. The average Fe moments of $YFe_{10.5}Mo_{1.5}C_x$ versus the carbon content (the solid dot indicates the average Fe moment of $YFe_{10.5}Mo_{1.5}N$).

This indicates that the arc-melting-introduced carbon atoms have a similar effect on the average Fe magnetic moments to that of nitrogen atoms.

The anisotropy field H_A was calculated by using the Sucksmith–Thompson method and the results are listed in table 1. Since the yttrium is non-magnetic, the magnetic anisotropy of $YFe_{10.5}Mo_{1.5}C_x$ manifests the effect of carbon on the magnetocrystalline anisotropy of the Fe sublattice. There is a slight trend towards decrease with the carbon content, which is similar to the effect of interstitial nitrogen [6].

4. Conclusions

In conclusion, most of the carbon atoms introduced by arc melting occupy the 2b sites in $Y(Fe, Mo)_{12}C_x$ compounds. The introduction of carbon has the effect of increasing both the Curie temperature and the saturation magnetization of $Y(Fe, Mo)_{12}$ compounds; these effects are similar to those of the nitrogen or carbon atoms introduced by gas–solid-phase reaction. This is of significance for technical applications, because of the better stability of these carbides.

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