1.2 Neutron diffraction study on the crystal and magnetic structures of arc-melted $PrFe_{10.5}Mo_{1.1}C_{0.4}$ alloy

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Abstract. The crystal and magnetic structures, especially the site occupation of C atoms in the crystal cell of arc-melted $PrFe_{10.5}Mo_{1.1}C_{0.4}$ permanent magnetic alloy have been determined by means of powder neutron diffraction study. Reitveld analysis of the neutron diffraction data indicates that Mo atoms prefers the 8i site, about 76 percent of the C atoms occupy the 8i substitution site, and the other part of C atoms enter the 2b interstitial site. The exact molecular formula should be $Pr(Fe_{10.575}Mo_{1.250}C_{0.175})C_{0.055}$. It seems that the site occupation of C atoms in the $ThMn_{12}$-type carbides depends not only on the kind of substitutional transition metals, but also on the components of the substitutional transition metals in the compounds. There are still amount of work to make systematic studies on the relations between them. The effect of C atoms on the magnetic properties is also discussed.

Key words: Neutron diffraction, Arc-melting, Permanent magnetic alloy

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 into the structure using gas-solid-phase reaction, and the nitrides and carbides have been regarded as promising candidates for permanent magnetic applications[1-3]. However the nitrides and carbides produced by gas-solid-phase reaction are metastable phase and will decompose at temperatures higher than 700°C in general, which limited the applications of these materials. While carbides prepared by melting method are more stable at high temperatures[4,5].

Zhang et al[6] and Hu et al [7] published results of neutron diffraction experiments on arc-melted $YFe_{11}TiC_{0.3}$ and $ErFe_{11}TiC_{0.25}$ alloys. They found that all of the carbon atoms occupy 8i site, not the interstitial 2b site. Mao[4,5] et al reported that most of the carbon atoms of arc-melted $YFe_{10.5}Mo_{1.5}C_x$ occupy the 2b site, only a small amount of carbon atoms enter 8i site. And Dong [8] et al found that in the arc-melted $YFe_{10}V_2C_{0.5}$ alloy almost half of the carbon atoms occupy 2b site, half enter 8i site. It is our conclusion that the site occupation of C atoms in the carbides prepared by arc-melting depends on the substituional transition metals, and it was found that the magnetic effect of carbon atoms introduced by arc-melting is different from that of the nitrogen and carbon atoms introduced by gas-solid-phase reaction[9].

The site occupation of carbon atoms is the key to understand the effect of carbon atoms on the magnetic properties, but how the site occupations depend on the substituional transition metals is still unknown. To study the site occupation of carbon atoms in this type of compounds and how the carbon atoms in the crystal cell affect the intrinsic magnetic properties is very important to magnetism and is very meaningful to improve the permanent magnetic properties.

In this paper, the crystal and magnetic structures, especially the site occupation of carbon atoms in the crystal cell of $PrFe_{10.5}Mo_{1.1}C_{0.4}$ permanent magnetic alloy are determined by powder neutron diffraction, and the effect of carbon atoms on the magnetic properties is discussed.

2. Experimental Method

$PrFe_{10.5}Mo_{1.1}C_{0.4}$ alloy was prepared by arc melting 99.5% pure materials in a purified argon atmosphere. The carbon was introduced as Fe-C alloy. X-ray diffraction and magnetic measurements determined the structure and the magnetic properties.

The neutron diffraction measurement were performed on the powder diffractometer using the heavy-water research reactor(HWRR) at China Institute of Atomic Energy in Beijing. The data were collected by scanning the samples from 7° to 87.2°in steps of 0.10° at room temperature with a neutron wavelength of 1.159Å. The diffraction pattern was analyzed by means of the Rietveld profile technique.

The refinement strategy consisted of refining the first with overall scale and temperature parameter. Then refined with cell constants, half width parameters(U,V,W only) and counter Zero point. Once good starting values were obtained for the above parameters, the atomic position parameters and then the occupation parameters were refined. Finally the rare-earth and the three transition-metal moments were varied to minimize the R factors. When more than two kinds of atoms occupy the same site, the occupation parameters can not be determined directly. So when refined the occupation parameters of carbon, molybdenum and iron atoms on the 8i site, considered the occupations of molybdenum in similar $R(Fe,Mo)_{12}$ samples we have studied and noticed that there is 5.08 percent of α-Fe phase in the sample, we restrained the occupation parameter of molybdenum to 2.5 after calculation.

3. Results and discussion

The neutron diffraction refinement results of $PrFe_{10.5}Mo_{1.1}C_{0.4}$ sample are listed in Table 1. And the neutron diffraction pattern of $PrFe_{10.5}Mo_{1.1}C_{0.4}$ at room temperature is showed in figure 1.

The neutron diffraction refinement results indicates that molybdenum atoms prefers the 8i site, about 76 percent of the carbon atoms occupy the 8i site, and the other part of carbon atoms enter the 2b interstitial site, the exact molecular formula should be $Pr(Fe_{10.575}Mo_{1.250}C_{0.175})C_{0.055}$, this means that about 43% carbon atoms lost in procedure of arc-melting. And there is about 5.08% α-Fe phase in the sample. The results are different from the site occupations of carbon atoms in arc-melted $RFe_{11}TiC_x$, $YFe_{10}V_2C_{0.5}$ and $YFe_{10.5}Mo_{1.5}C_x$ alloys. It seems that the site occupation of carbon atoms in the $ThMn_{12}$-type carbides depends not only on the kind of substitutional transition metals, but also on the components of the substitutional transition metals in the compounds. There are still amount of

Table 1. Neutron diffraction refinement results for $PrFe_{10.5}Mo_{1.1}C_{0.4}$

Atoms	Site	x	y	z	Occ.	$M_x(\mu_B)$	$M_y(\mu_B)$	$M_z(\mu_B)$
Pr	2a	0.0000	0.00	0.00	2.00	1.43	0.00	0.00
Fe	8i	0.3595	0.00	0.00	5.15	1.56	0.00	0.00
Mo	8i	*0.3595*	*0.00*	*0.00*	*2.50*	0.00	0.00	0.00
C	8i	0.3595	0.00	0.00	0.35	0.00	0.00	0.00
Fe	8j	0.2736	0.50	0.00	8.00	1.49	0.00	0.00
Fe	8f	0.2500	0.25	0.25	8.00	1.49	0.00	0.00
C	2b	0.0000	0.00	0.50	0.11	0.00	0.00	0.00

a=b=8.581, c=4.777(Å); R_P=6.99, R_{WP}=7.62, R_N=2.98, R_M=6.39(%), α-Fe content: 5.08%

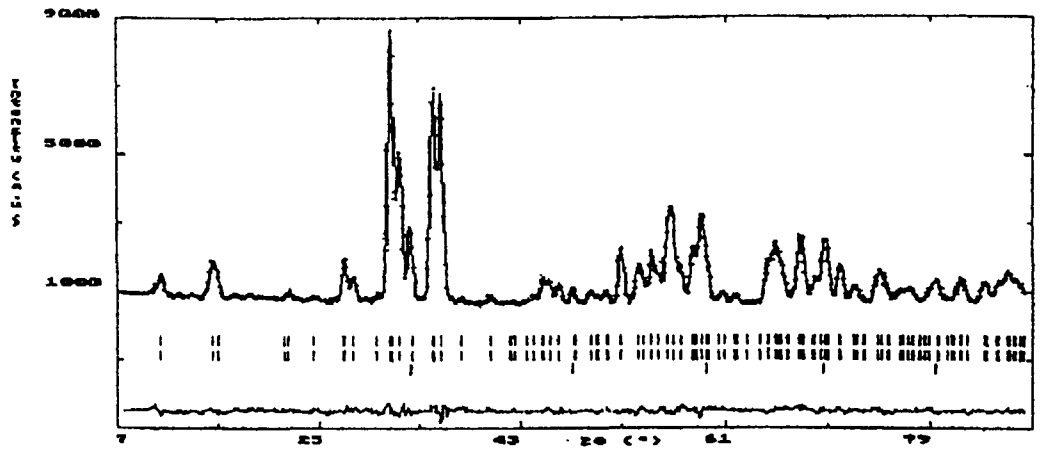

Fig. 1. The neutron diffraction pattern of $PrFe_{9.5}Mo_{1.5}C_{0.4}$ at room tperature

work to make systematic studies on the relations between them.

Yang[9] et al. found that in the $RTiFe_{10.75}C_{0.25}$ samples introducing substitutional carbon atoms by arc-melting have no effect on Curie temperature, but the saturation magnetization is increased and the unit cell volume is decreased slightly, the planar anisotropy for rare-earth ions with negative second-order Stevens constant α_J such as Nd^{3+} and Pr^{3+} is increased and the axial anisotropy for those with positive α_J such as Sm^{3+} is strengthened. While interstitial carbon or nitrogen atoms introduced into the $R(Fe,M)_{12}$-type alloys by gas-solid-phase reaction increase unit cell volume, enhance the Fe-Fe exchange reaction, so increase the Curie temperature and saturation magnetization. Meanwhile the interstitial carbon or nitrogen atoms will increase the axial anisotropy for rare-earth ions with negative second-order Stevens constant α_J such as Nd^{3+} and Pr^{3+} [10].

Because only a small quantity of carbon atom enter the 2b interstitial site, the general effect of C atoms on the magnetic properties should be as this: increasing the planar magnetic anisotropy of rare-earth sublattice, thus maintaining the easy-planar magnetic structure, meanwhile slightly increasing the Curie temperature and the saturation magnetization. The neutron diffraction result shows that the atom moments M_x for Pr, Fe(8i), Fe(8j) and Fe(8f) ions are 1.43, 1.56, 1.49 and 1.49μ_B respectively, which are somewhat smaller than expected.

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