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Magnetic properties and ^{57}Fe Mössbauer effect investigation in $U_{1-x}Y_xFe_{10}Si_2$ solid solutions

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Abstract

The solid solutions $U_{1-x}Y_xFe_{10}Si_2$ have been studied by means of X-ray diffraction, ^{57}Fe Mössbauer effect and magnetic investigations. In the intermediate concentration range, a redistribution of the iron and silicon atoms over non-equivalent positions was observed despite the fact that both ternaries have similar occupancies in all positions. Such a redistribution could be a reason for the slight non-linearity observed in the concentration dependences of lattice parameters and magnetic properties. The results of a hyperfine field study were used to estimate the uranium magnetic moment in $UFe_{10}Si_2$, which is about $0.5\mu_B$ and coupled ferromagnetically with the iron sublattice magnetic moment.

1. Introduction

The $RFe_{10}Si_2$ intermetallic compounds belong to a relatively new class of magnetic materials $RFe_{12-x}M_x$ (R≡rare earth metal or Y; M≡Si, Ti, V, Cr, Mo, W, Re; $1 \leq x \leq 3$). These compounds are characterized by high iron and low R content, a large Curie temperature and a magnetic moment and a considerable uniaxial magnetocrystalline anisotropy of the iron sublattice. One representative of this family, namely $SmFe_{11}Ti$, is really considered as a hard magnetic material for permanent magnets. The others are also very interesting magnetic substances with competitive exchange interactions and contributions to magnetocrystalline anisotropy [1–5].

$UFe_{10}Si_2$ is an actinide representative of this class. The compound has $T_c = 653$ K, magnetic moment $M = (16.4\text{–}20)\mu_B$ per formula unit (according to the different references; it depends on applied field and the presence of extraneous free iron in the samples) and uniaxial magnetic anisotropy [6–8]. The main question, as usual for uranium magnetics, is the magnetic state of the uranium atoms. In present work, we used a traditional method of

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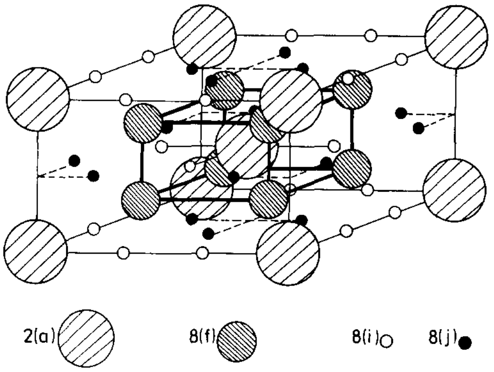


Fig. 1. Crystal structure of the $ThMn_{12}$ type.

non-magnetic dissolution of one of the sublattices, the uranium sublattice in this case, in order to obtain information on the contribution of this sublattice to the magnetic properties of the compound. Yttrium, being a classical non-magnetic analogue for the rare earth metals, forms the compound $YFe_{10}Si_2$ with the same crystal structure and $T_c = 558$ K and $M = 18.2\mu_B$ per formula unit [7, 9].

The tetragonal crystal structure of both compounds belongs to the $ThMn_{12}$ type (space group *I4/mmm*). The unit cell contains two formula units and is presented in Fig. 1. The uranium or yttrium atoms occupy positions of the 2(a) type, and iron and silicon are distributed over the 8(f), 8(i) and 8(j) sites with a considerable preference in the distribution.

The comparison of the magnetocrystalline anisotropy of these two compounds and the conclusion about the magnetic state of uranium have been given in ref. 7. However, in the case of uranium, trivalent yttrium cannot be considered as a simple non-magnetic substituent, as for most of normal trivalent rare earth metals, because of the non-predictable electronic state of uranium, which can vary within a wide range under different environmental conditions. The substitution of yttrium for uranium can influence the state of the iron sublattice through 5f–3d hybridization in uranium-containing compounds and cause a variation in the distribution of the iron atoms over different crystallographic sites. In such a case it would be useful to study a systematic change of the structure and magnetic properties upon substitution of uranium for yttrium. For this reason, we prepared the solid solutions $U_{1-x}Y_xFe_{10}Si_2$ ($0 \leq x \leq 1$ through steps of 0.2).

The present paper contains the results of magnetic measurements as well as of Mössbauer effect investigations of the component distribution and the hyperfine interactions in this system.

2. Experimental details

The $U_{1-x}Y_xFe_{10}Si_2$ alloys were obtained by melting components (uranium and yttrium purity, 99.9%; iron and silicon, 99.99%) in an arc furnace under an argon protective atmosphere and annealing at 900 °C for two weeks. The X-ray and thermomagnetic phase analysis revealed a single-phase state for all uranium-containing samples. The compound with $x=1$, $YFe_{10}Si_2$, had about 5% of extraneous α-Fe phase, which did not disappear during homogenization. This fact is in agreement with ref. 9, where it was concluded to be impossible to prepare single-phase samples of $YFe_{10}Si_2$ as well as these silicides with rare earth metals.

The Mössbauer measurements were carried out using a conventional constant acceleration spectrometer (POLON) with ^{57}Co in a chromium matrix as the source from 14 to 640 K. The velocity scale was calibrated using an α-Fe absorber at room temperature.

The Curie temperatures were determined by d.c. susceptibility measurements.

3. Results and discussion

In Fig. 2, the concentration dependences of the lattice parameters a and c are presented. The crystal structure of the $ThMn_{12}$ type is preserved for the entire range of concentrations. The smooth monotonic increase in both lattice parameters with increasing yttrium content reflects the fact that the yttrium atomic radius is larger than that of uranium. The lattice expansion is isotropic; the c/a ratio is nearly constant for the compounds investigated. A slight negative deviation from the Vegard law is typical for such solid solutions and can be connected with some redistribution of the iron and silicon atoms during U–Y substitution, as shown below by Mössbauer effect measurements.

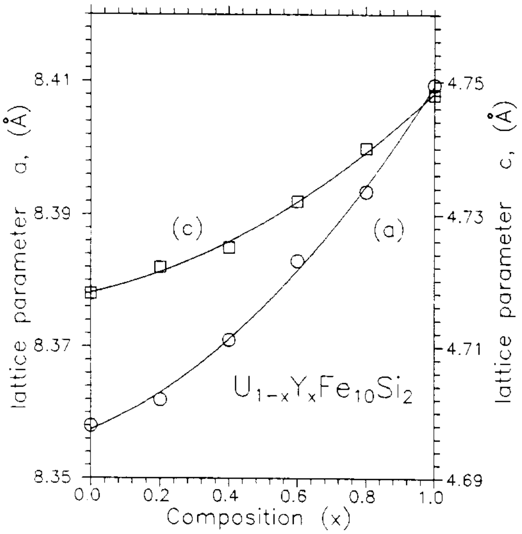
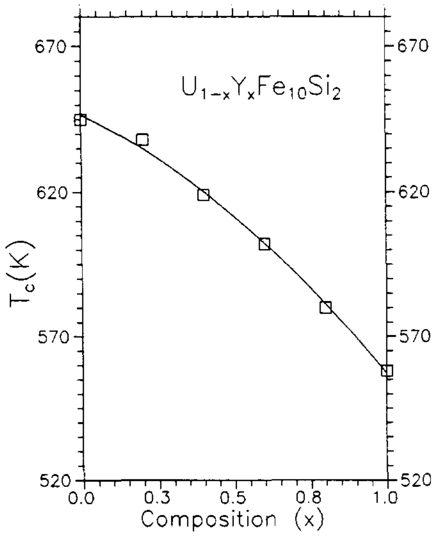


Fig. 2. Concentration dependences of the lattice parameters.

Figure 3 presents the concentration dependence of the Curie temperature. The monotonic decrease in T_c with increasing yttrium content can be explained by the magnetic state of uranium and the decrease in the considerable U–Fe exchange interaction contribution to the T_c value of the compounds.

In Fig. 4, the Mössbauer spectra of the studied compounds are presented. As in the earlier papers [10, 11], the experimental spectra were approximated by three sextets resulting from three non-equivalent positions of the iron atoms. The sextet with largest hyperfine field H_{hf} is related to iron atoms located on the 8(i) sites. The sextet with the lowest value of H_{hf} corresponds to iron atoms on the 8(f) sites as proposed in ref. 10. The Fe–Fe distance d_{Fe-Fe} for the 8(f) sites is shorter than that for the 8(j) sites, whereas the numbers of the nearest iron neighbours for both positions are almost equal. Therefore, the main factor influencing H_{hf} is d_{Fe-Fe}.

The dispersion of hyperfine fields connected with the different numbers of silicon atoms in the closest regions of the ^{57}Fe Mössbauer nuclei was taken into account by different linewidths of the external and internal lines ($\Gamma_{1-6} > \Gamma_{2-5} > \Gamma_{3-4}$) when the experimental spectra were fitted. The results of the fitting are indicated in Fig. 4 by full lines and reveal a good approximation



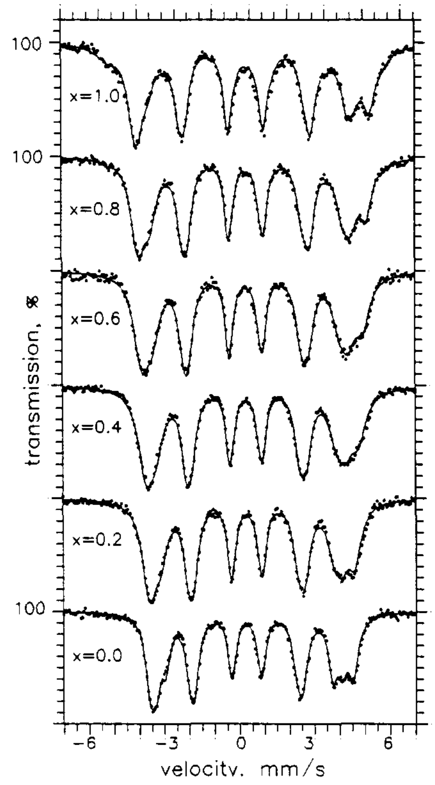


Fig. 3. Concentration dependence of the Curie temperature.

Fig. 4. Mössbauer spectra of $U_{1-x}Y_xFe_{10}Si_2$ solid solutions for different x at 14 K.

to the experimental data. As follows from Fig. 4, the substitution of uranium for yttrium results in a change in the line intensity for the sextets corresponding to the different iron sublattices. Even for a low yttrium concentration ($x=0.2$), a decrease in the iron occupation could be seen for the 8(j) sites with simultaneous increase in the 8(f) occupation factor. Such a tendency is observed up to $x=0.6$. Further substitution leads to some return to the initial (corresponding to low x) distribution of the iron atoms over 8(f), 8(i) and 8(j) positions.

In Fig. 5, the occupation factors for all sites are shown *vs.* the yttrium concentration. The average number of the nearest neighbours and the average Fe–Fe distance for each position are calculated by using the results of the fitting of the Mössbauer spectra. They are presented in Fig. 6.

The hyperfine fields H_{hf} for all sites of the unit cell increase with increasing yttrium content (Fig. 7(a)). The H_{hf} values for 8(i) and 8(f) positions increase linearly, whereas for the 8(j) sites the curve $H_{hf}(x)$ displays a small change in its slope. This is apparently connected with the decrease in $d_{Fe-Fe}(x)$ for 8(j) positions when x is approaching the 0.4–0.6 region (Fig. 6(b)). In the same concentration range, a similar dependence of the isomer shift is observed for the nuclei located on the 8(f) sites (Fig. 7(b)). This also probably results from the size effect.

The increase in the average H_{hf} value (Fig. 8) means an increase in the average iron moment, which can lead to enhancement of the Fe–Fe exchange interaction and, consequently, to an increase in the Curie temperature with increasing yttrium content. Therefore, the opposite behaviour of the $T_c(x)$

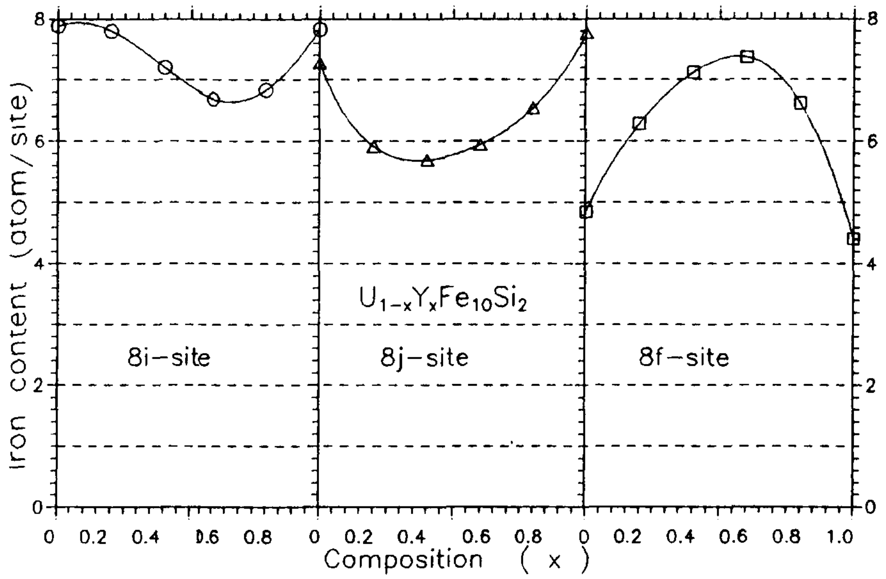


Fig. 5. Distribution of the iron atoms on different crystallographic positions *vs.* yttrium content x.

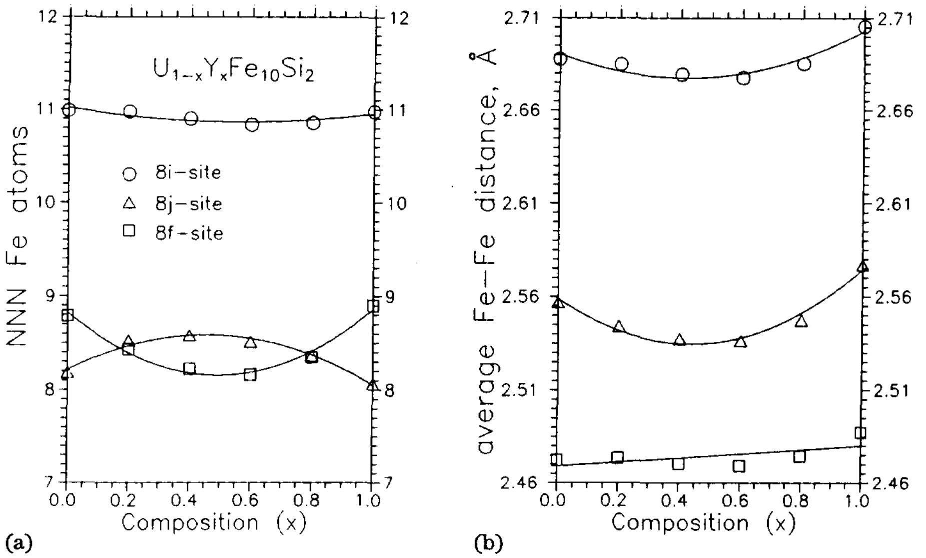


Fig. 6. Concentration dependences (a) of the number of nearest iron neighbours (NNN) and (b) of the average Fe–Fe distance for the iron atoms on different crystallographic positions.

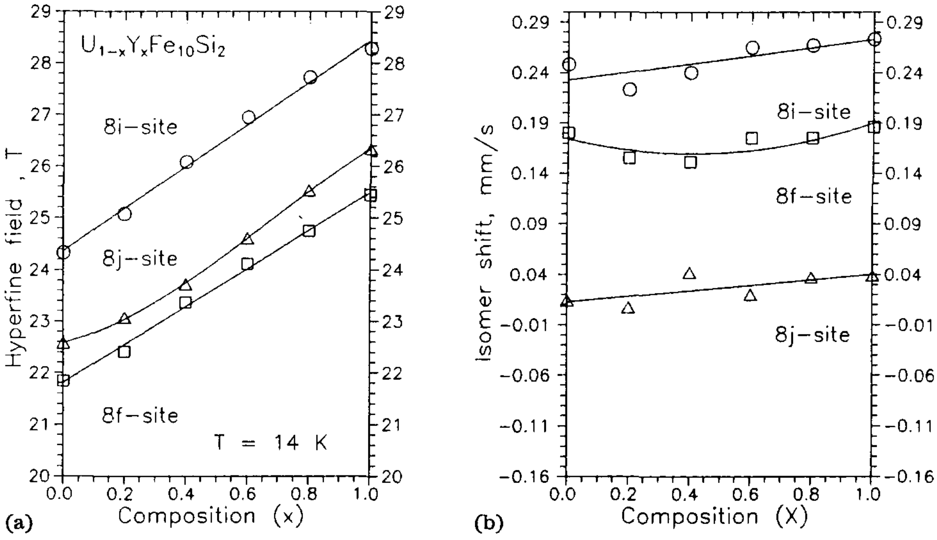


Fig. 7. Concentration dependences (a) of the hyperfine field and (b) of the isomer shift for the iron atoms on different crystallographic positions at $T = 14$ K.

dependence, shown in Fig. 3, could not be explained without taking into account a considerable change in the U–Fe exchange interaction contribution, accompanied by only a modest change in the iron sublattice contribution.

Assuming a field–moment conversion factor of 14.5 T/μ_B proposed in ref. 12, we presented in Fig. 8 the values of the resulting magnetic moment

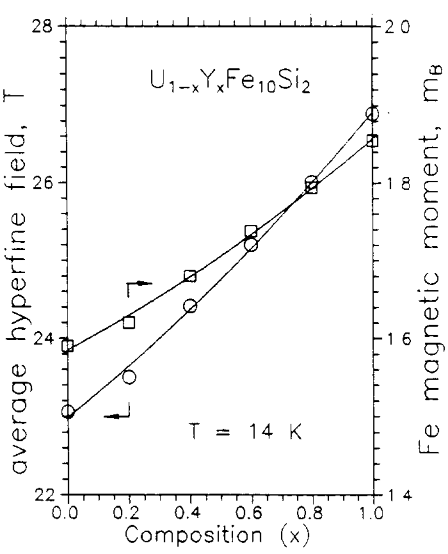


Fig. 8. Concentration dependences of the average hyperfine field and of the calculated resulting magnetic moment of iron sublattices at $T = 14$ K.

μ_{Fe} of the iron sublattices for the compounds studied. The value of μ_{Fe} for the compound without uranium ($x = 1$), $18.5\mu_B$, is in good agreement with the magnetometric result, $18.2\mu_B$ [7]. Therefore, we can use μ_{Fe} for $x = 0$, $15.9\mu_B$, to estimate the uranium magnetic moment μ_U in $UFe_{10}Si_2$ as the difference between the molecular magnetic moment M and μ_{Fe}. The most reasonable value of M, $16.4\mu_B$ [7], gives $\mu_U = 0.5\mu_B$. It may even be larger when other reported data for M are considered. In any case, we obtain a positive contribution of μ_U to M, and consequently, a ferromagnetic arrangement between uranium and iron sublattices. The conclusion about the magnetic state of the uranium sublattice in $UFe_{10}Si_2$ has been already derived from spontaneous magnetostriction [8] and magnetocrystalline anisotropy [7] investigations. Thus, the results presented may be taken as evidence for magnetic ordering of the uranium sublattice in $UFe_{10}Si_2$.

4. Conclusion

A continuous series of solid solutions forms between $UFe_{10}Si_2$ and $YFe_{10}Si_2$ intermetallic compounds for the whole range of the U–Y substitution. The lattice parameters and magnetic properties vary monotonically with yttrium concentration. In the intermediate concentration range, a redistribution of the iron and silicon atoms over the three non-equivalent crystallographic positions was observed despite the fact that both ternaries have a similar iron atom occupancy for these positions. Such a redistribution could be the reason for a slight non-linearity in concentration dependences of lattice parameters and magnetic properties.

The results of a hyperfine field study were used to estimate the magnetic moments of the uranium atoms in $UFe_{10}Si_2$. The uranium moments have values of about $0.5\mu_B$ and they are ordered ferromagnetically in relation to the iron sublattice magnetic moments.

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