Letter

Magnetic properties of $Ce_2Fe_{17-x}Si_x$ compounds

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(Received October 28, 1993)

Abstract

We have studied the magnetic properties of $Ce_2Fe_{17-x}Si_x$ with $x=1$, 2 and 3. The substitution of silicon for iron leads to a strong enhancement of the Curie temperature and a reduction of the unit-cell volume.

1. Introduction

The discovery that interstitial solid solutions of the R_2Fe_{17} compounds with N or H could increase the Curie temperature, has led to numerous studies of these materials for potential applications as permanent magnet materials. Also other variations such as substitutional solid solutions, or a combination of both, might lead to a further enhancement of the ordering temperature of these compounds [1]. Substitution of silicon for iron in R_2Fe_{17} compounds strongly increases the Curie temperature, whereas the unit-cell volume decreases with increasing silicon content. This increase in ordering temperature is rather peculiar. Replacement of ferromagnetic iron by diamagnetic silicon and the lattice contraction upon silicon substitution, would normally be expected to lead to a lowering in the Curie temperature. Conventional arguments suggest that silicon would occupy the 6c(4f) sites, which would reduce the negative Fe–Fe exchange interactions [2, 3] and thus lead to an enhancement of the Curie temperature. However, a recent neutron diffraction and Mössbauer effect study of the $Nd_2Fe_{17-x}Si_x$ solid solutions by Long *et al.* [4], indicates that silicon preferentially occupies the 18*h* site with the largest number of rare-earth near neighbours. Their investigation suggests that the increase in Curie temperature is closely related to the lattice expansion in the 9*d*–18*h* plane of the unit-cell. In fact this makes the $R_2Fe_{17-x}Si_x$ interesting also from a fundamental point of view. Our investigation forms part of a series of crystal-chemical studies in which we study the magnetic properties of cerium-based ferromagnetic intermetallic compounds.

2. Experimental details

The samples were prepared by arc melting from starting materials of at least 99.9% purity in an argon atmosphere. After arc melting, the samples were wrapped in tantalum foil and were vacuum annealed in quartz tubes at 900 °C for two weeks. The samples were investigated by powder X-ray diffraction. The magnetic measurements were performed on a SQUID magnetometer and a home-built Faraday balance.

3. Results and discussion

X-ray diffraction of $Ce_2Fe_{17-x}Si_x$ showed the prepared samples to be approximately single phase, having the rhombohedral Th_2Zn_{17} structure. The samples contained a few percent of elemental iron as an impurity phase in some cases. The lattice constants for compounds of varying silicon contents were derived from the diffraction diagrams. Substitution of silicon for iron leads to a contraction of the unit-cell in the basal plane and to a slight expansion in the *c*-axis [6], which is pronounced for silicon content $x=1.0$ (Table 1).

The magnetic isotherms at 5.0 K are shown in Fig. 1, from which the saturation magnetisation M_s was derived, as shown in Fig. 2. The introduction of silicon in Ce_2Fe_{17} has led to the disappearance of the metamagnetic transition as found in Ce_2Fe_{17} [1]. Values of the saturation magnetisation were obtained by extrapolating the $M(1/B)$ curves to $1/B=0$. From a comparison of the saturation magnetisation of $Y_2Fe_{17-x}Si_x$ as reported in refs. 3 and 9, it can be concluded that the valency of cerium is likely to be tetravalent. The decrease of the Fe moments are because of magnetic dilution and the contraction of the unit-cell volume.

TABLE 1. Lattice parameters of $Ce_2Fe_{17-x}Si_x$

Si content x	a axis (Å)	c axis (Å)	V (Å^3)
0[a]	8.491	12.409	774.79
0.2	8.486	12.412	774.01
0.4	8.484	12.408	773.46
0.6	8.478	12.420	773.12
0.8	8.477	12.429	773.12
1	8.456	12.472	769.8
2	8.457	12.428	769.8
3	8.441	12.424	766.55

[a]According to ref. 5.

SSDI 0925-8388(93)01076-G

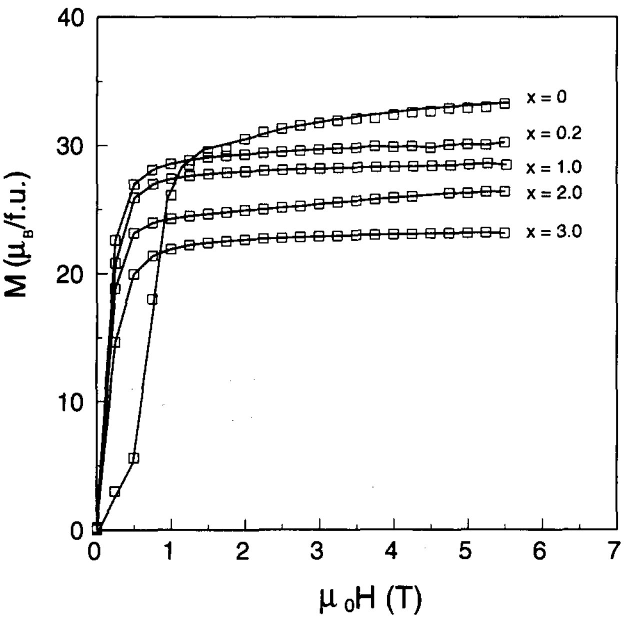

Fig. 1. Magnetic isotherms of $Ce_2Fe_{17-x}Si_x$ at 5.0 K.

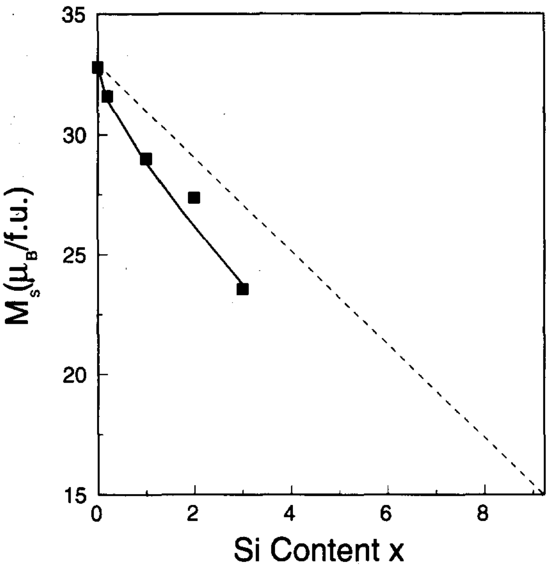

Fig. 2. Saturation magnetisation M_s as a function of silicon content x. The dotted line indicates the decrease of M_s due to magnetic dilution, the solid curve is a guide to the eye.

The magnetisation $\sigma(T)$ as a function of temperature of field-cooled free particle samples is shown in Fig. 3. At low Si content $x(x<1)$ there is still a cusp-like anomaly at 210 K [7] probably associated with a helimagnetic structure [8] of the iron moments, as in the pure Ce_2Fe_{17} compound [8]. The Curie temperatures as derived from the data in Fig. 3 are shown in Fig. 4. The Curie temperatures raise sharply and tend to saturate as the solubility limit seems to be reached [6]. The Curie temperature of the $Ce_2Fe_{17-x}Si_x$ compounds are enhanced, but to a lesser extent compared with, *e.g.* the nitrogenated compounds. Further research is needed to clarify the mechanism underlying the increase in Curie temperature of the $R_2Fe_{17-x}Si_x$ compounds.

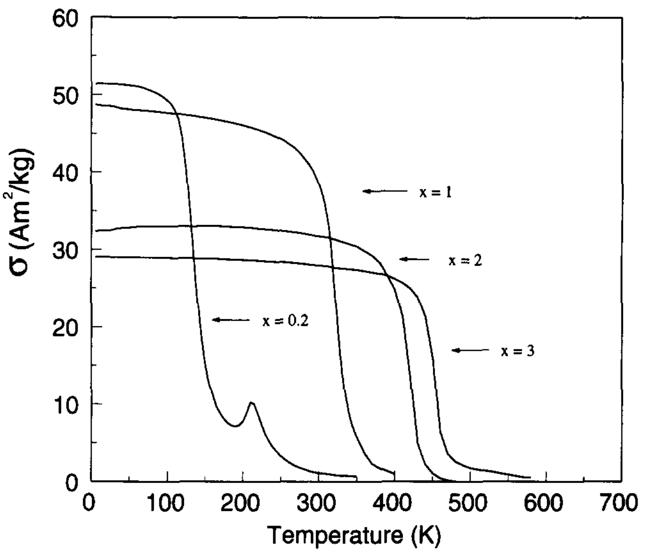

Fig. 3. Temperature dependence of the magnetisation $\sigma(T)$ measured at $\mu_0H=0.1$ T for field cooled free particle samples.

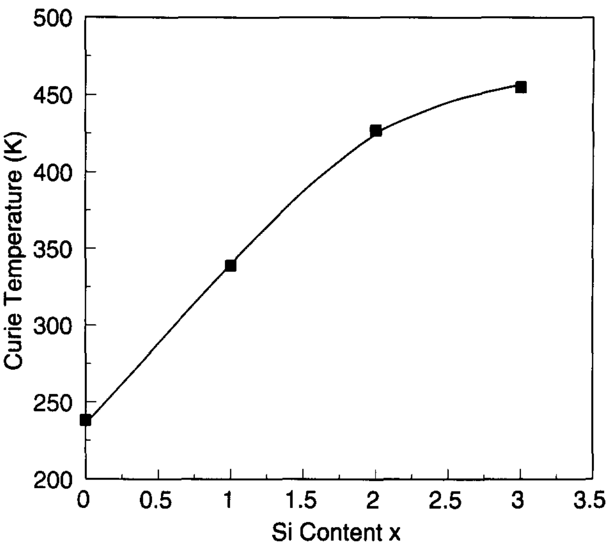

Fig. 4. Curie temperatures as a function of silicon content x. The solid curve is a guide to the eye.

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