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Si/Al influence on $Fe_{70}Al_{30-x}Si_x$ alloys

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Abstract The main aim of this work is to study the influence of silicon/aluminium ratio in the structural and magnetic properties of ordered $Fe_{70}Al_{30-x}Si_x$ alloys by means of X-ray diffraction, Mössbauer spectroscopy and magnetic (SQUID) measurements.

Keywords Fe intermetallics · Mossbauer spectroscopy · Magnetic inhomogeneities

1 Introduction

Around the $Fe_{75}Al_{25}$ content, the phase diagram of Fe-Al alloys presents a transition from B2 to D03 structure (decreasing temperature). Within this range, the magnetic behaviour is complex, particularly around 30 at% Al. Indeed, with decreasing temperature the magnetic behaviour of $Fe_{70}Al_{30}$ goes from ferromagnetic through superparamagnetic-like behaviour to end in a spin-glass like state [1]. Small angle neutron scattering experiments in alloys around 30 at% Al show magnetic clusters that evolve with temperature [2, 3]. Si, like Al, is a non-magnetic element and it has only one p electron more than Al, so Si is a very appropriate adding element for studying the complex magnetic behaviour of Fe-Al alloys. It is also worth noting that Fe-Si and Fe-Al alloys have the same ordered (D03 and B2) structures [4]. The main

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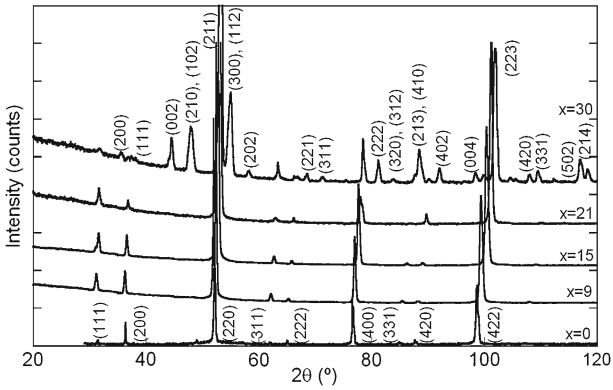

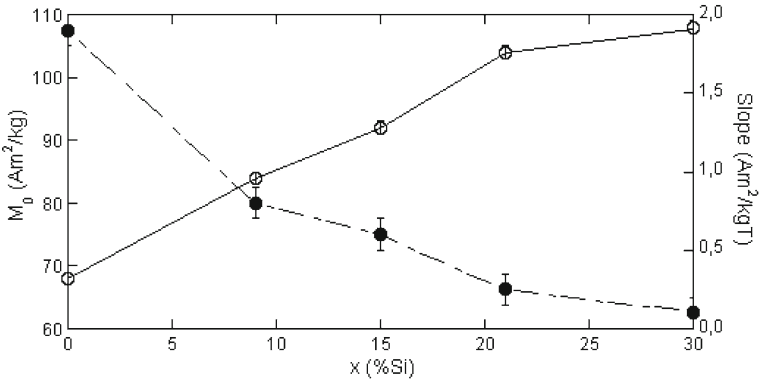

Fig. 1 XRD of $Fe_{70}Al_{30-x}Si_x$ alloys. x = 0–30 alloys show cubic phases. $D8_8$ is also present in x = 30

Fig. 2 M_0 (empty circles) and slope (filled circles) curves of $Fe_{70}Al_{30-x}Si_x$ alloys at RT (see text)

aim of this work is to study the influence of silicon/aluminium ratio in the structural and magnetic properties of ordered $Fe_{70}Al_{30-x}Si_x$ alloys.

2 Experimental

Induction melting obtained samples were powered by mechanical crushing. To obtain large domains of ordered structures, they were annealed at 1223 K, then cooled down to 1023 K and kept at this temperature for 3 h; after they were cooled down to 823 K and kept there for 1 week (in order to obtain great amount of D03 structure). The samples were studied by X-ray diffraction (XRD), magnetic measurements and Mössbauer spectroscopy. NORMOS program [5] was used to fit the spectra obtained in transmission geometry with ^{57}Co-Rh.

3 Results and discussion

In the XRD of $Fe_{70}Si_{30}$ (see Fig. 1) there is one structure that is not present in the other samples: the hexagonal $D8_8$, but even the minimum Al content makes this structure disappear. The long-range order parameter is very close to 1 and

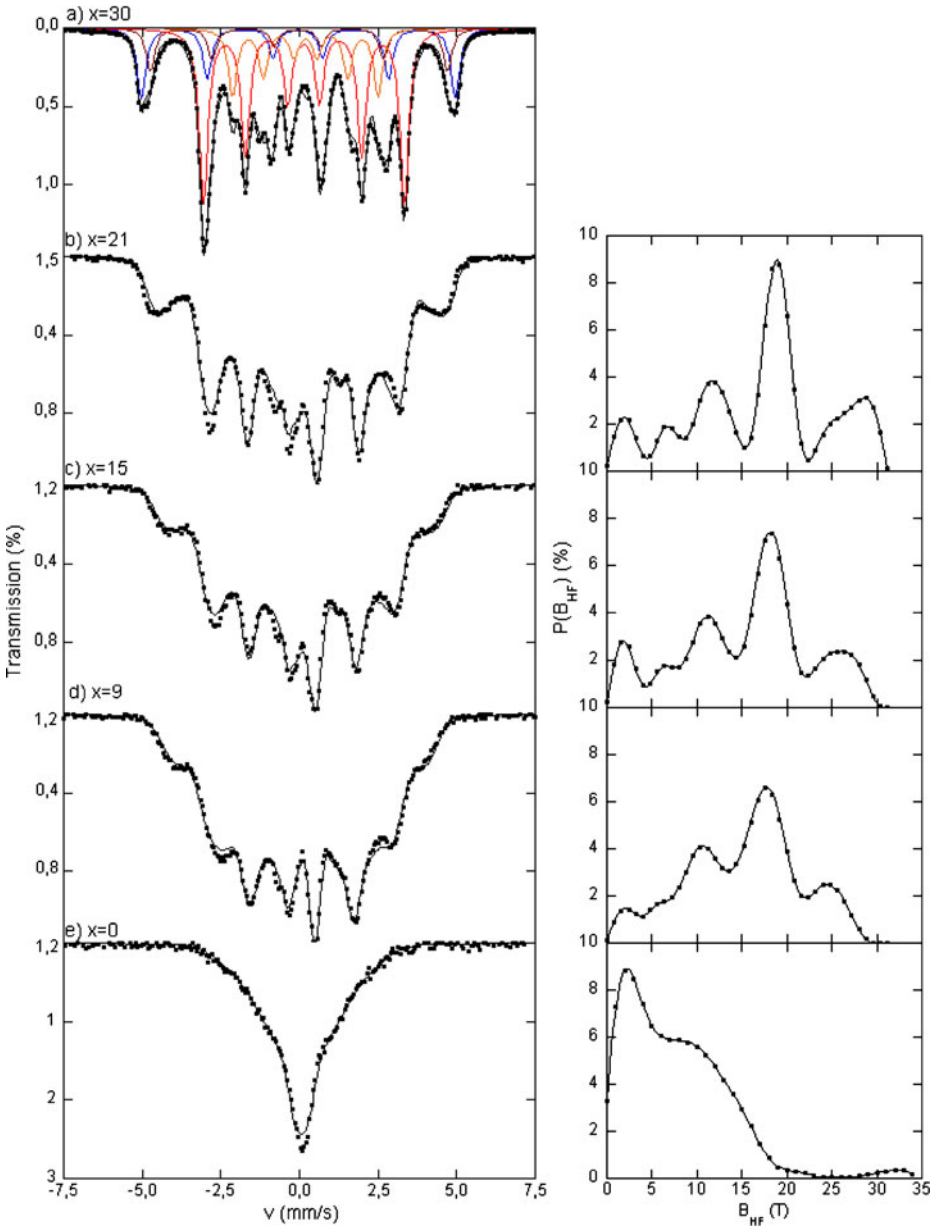

Fig. 3 Mössbauer spectra of $Fe_{70}Al_{30-x}Si_x$ alloys at RT and the corresponding P(Bhf)

the increase of Si/Al ratio makes the lattice parameter of D03 decrease linearly. However, by means of XRD it is very difficult to obtain the amount of D03 and B2 ordered structures because reflections are located at the same angles. Anyway, Fig. 1 shows a clear variation of the ratio of (111) and (200) peak intensities, indicating the presence of B2 structure in the samples with large Al content.

Figure 2 shows M_0, linear fit of the magnetization between 4T and 7T extrapolated to zero applied field, and its slope versus Si content. M_0 decreases with Al content, but the magnetization slope at high fields increases. The M(H) curve of $Fe_{70}Si_{30}$ alloy is the unique that saturates at high fields. It indicates that $Fe_{70}Si_{30}$ is the only alloy that is in a classical ferromagnetic state. The Mössbauer spectra of the binary alloys $Fe_{70}Si_{30}$ and $Fe_{70}Al_{30}$ are very different (see Fig. 3). The first one can be fitted using discrete sextets. Indeed, it has been fitted with two sextets corresponding $D8_8$ (Fe_5Si_3) structure and 4 to the D03 one. The fit of D03 structure indicates the amount of Fe in it is 73%, in good agreement with the fact that $D8_8$ structure needs an excess of Si atoms with respect to the nominal one.

However, the Mössbauer spectrum of $Fe_{70}Al_{30}$ sample, similar to the spectrum obtained by other authors [6, 7], can only be fitted by means of a continuous distribution of B_{hf} fields. Indeed, the fit by means of a hyperfine field distribution (see Fig. 3) shows that the larger contribution is centred at 2 T, and contributions of B_{hf} up to 20 T decrease monotonously. Even though, Mössbauer spectra of ternary alloys can be well fit with 4 discrete sextets (whose width increases in relation to $x = 30$ alloy) and one doublet, Fig. 3 shows the fit performed with a continuous distribution of B_{hf}. $P(B_{hf})$ indicates clearly that with Al content increase the B_{hf} of the observed bumps decreases, and they get wider and merge into two wide bumps located at 2T and 10T. Assuming that the width of each bump is only due to different Al/Si chemical configuration around Fe, one would expect the maximum width to occur at $x = 15$ (the minimum should be at $x = 0$ and $x = 30$). However, the width of the bumps increases with Al content and the maximum is observed in $x = 0$ alloy. Therefore, the change of the structural chemical surroundings of Fe positions by Al/Si substitution in these very ordered alloys is not the cause of the observed behaviour in Mössbauer spectra. Its origin must be on magnetic grounds. On the other hand, Fig. 2 shows that the introduction of Al produces a deviation from classical ferromagnetic behaviour, which increases with increasing Al content. So, these results clearly suggest the latter behaviours must be correlated. Moreover, Figs. 2 and 3 show that M_0 and the maximum B_{hf} decrease with Al content increase in the alloy.

4 Conclusions

Al addition makes $D8_8$ phase present in $Fe_{70}Si_{30}$ alloy disappear. Magnetic measurements and Mössbauer data show that Si addition to $x = 0$ alloy changes completely and monotonically the complex magnetism, related to magnetic inhomogeneities [2, 3], to a classic ferromagnetic behaviour. So, the influence of Si and Al in the magnetism of Fe in this alloy series is opposite.

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