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Mössbauer spectroscopy study of the influence of Si addition to ordered and disordered Fe$_{60}$Al$_{40}$ alloys

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Abstract. We have used Mössbauer spectroscopy to study the influence of Si addition to Fe$_{60}$Al$_{40}$ composition without changing the iron content of the alloy. The study has been performed in ordered and mechanically deformed samples. This work shows that the B2 structure present in ordered Fe$_{60}$Al$_{40}$ alloy does not change with Si introduction in the studied ternary alloys. However, the binary Fe$_{60}$Si$_{40}$ alloy shows D03 and B20 structures. Mechanical deformation of the ordered samples induces a clear change of the Mössbauer spectra of binary alloys with the presence of new sextets or variation of the previous ones; however, the Mössbauer spectra of the studied ternary alloys are very similar to the ordered ones.

1. Introduction
The magnetic behavior of FeAl alloys has been a subject of several theoretical and experimental studies because they often display unique magnetic properties. Hernando et al. [1] showed that a paramagnetic to ferromagnetic transition linked to an order-disorder transition between ordered B2 structure (paramagnetic) and disordered A2 structure (ferromagnetic) can be observed after mechanical deformation in Fe$_{60}$Al$_{40}$. In Fe$_{70}$Al$_{30}$ an enhancement of the magnetism of the intermetallic alloy after mechanical deformation has been observed too [2].

In this work we study the influence of Si addition to Fe$_{60}$Al$_{40}$ binary alloy in ordered and mechanically deformed samples by means of Mössbauer spectroscopy and X-ray diffraction.

2. Experimental
Five Fe$_{60}$Al$_{1-x}$Si$_x$ alloys with x= 0, 12, 20, 28 and 40 were prepared by induction melting in alumina crucibles and cast into ingots under a 40 kPa pressure of high purity argon. Iron, Aluminum and Silicon with a purity of 99.99, 99.99 and 99.999%, respectively were used. To obtain the ordered samples, they were annealed for 2 h at 1043 K, then they were kept at 843 K for 10 h and afterwards they were cooled down to room temperature at a rate of 3 K/min. The mechanical deformation has been performed crushing the ordered samples to obtain homogeneous powders with a diameter of about 10 μm. $^{57}$Fe Mössbauer spectroscopy measurements were carried out at room temperature in transmission geometry using a conventional spectrometer with a $^{57}$Co-Rh source. The measured spectra were fitted using the NORMOS program developed by Brand et al. [3]. All the isomer shift values presented in this work are relative to metallic iron at room temperature. XRD measurements were carried out in a Philips PW 1710 powder diffractometer using CuKα radiation.
3. Results and discussion

From the structural point of view the largest difference in the binary phase diagrams of Fe rich FeAl(Si) system corresponds to alloys with composition 60 at.%Fe. Indeed, the alloy Fe_{60}Al_{40} belongs to a region of the phase diagram where the equilibrium structure corresponds to the B2 structure; however, Fe_{60}Si_{40} alloy is located in a biphase zone of B20 and D03 structures. The B20 structure (\(\varepsilon\)-FeSi) is of distorted NaCl type. It consists of a simple cubic Bravais lattice having four Fe atoms and four Si atoms in the unit cell, with P2\(_1\)3 spatial group, where all Fe positions are equivalent. Fe local symmetry is trigonal, with a unique Si atom as nearest neighbor, located at 2.28 Å, three Si atoms as second nearest neighbors located at 2.35 Å, and three Si atoms as third nearest neighbors located at 2.52 Å [4]. In such a structure all Fe and Si atoms are equivalent and each atom is surrounded by 7 atoms of different kind (the distances up to the third nearest neighborhood are very similar).

![X-ray diffraction patterns of the ordered samples.](image)

3.1. Ordered Fe_{60}Al_{40-x}Si\(_x\) alloys

Figure 1 shows the XRD pattern of the alloy series Fe_{60}Al_{40-x}Si\(_x\). The pattern of binary alloys correspond to phase diagram equilibrium ones, that is to say, Fe_{60}Si_{40} presents a mixture of phases D03 and B20. On the other hand, Fe_{60}Al_{40} alloy shows a perfect B2 structure. As far as ternary alloys are concerned Figure 1 shows that Al addition causes the disappearance of the B20 structure. Indeed, no characteristic reflection corresponding to B20 structure is observed. Neither can any characteristic (111) reflection of the D03 structure be observed in the diffractograms of the alloys containing Si. Therefore, all the studied ternary alloys present the B2 structure, alike in Fe_{60}Al_{40} alloy, with no hints of the D03 structure. So, the minimum Al addition in this study, 12 at. %Al, is enough to destroy completely the phases present in the binary Fe_{60}Si_{40} alloy. Figure 1 shows clearly that the lattice parameter increases with Al addition. It increases quite linearly from 2.82 Å (12 at. %Al) to 2.90 Å (40 at. %Al).
Figure 2 shows the Mössbauer spectra of the ordered alloy series studied. The alloys that present B2 phase at room temperature in the XRD do not present any sextet at all. Indeed, Fe$_{60}$Al$_{40}$ alloy presents a unique singlet. On the other hand, Fe$_{60}$Si$_{40}$ alloy, which presents D0$_3$ characteristic reflections, has been fitted by means of five sub-spectra (see figure 2 a, bottom). Four of the five sub-spectra are sextets corresponding to the D0$_3$ structure (two sextets for 8Fe position, and one for positions 4Fe and 3Fe). The fifth sub-spectrum is a doublet corresponding to the B20 structure. It is worth noting that this results show the expected pattern of FeAl binary alloys, where the D0$_3$ structure gives sextets in Mössbauer spectra taken at room temperature and the B2 structure does not give any sextet [5]. From the area relative to the sub-spectra it is concluded that 58 % of the sample has D0$_3$ structure and 42 % B20 one. Moreover, analyzing the relative areas of the sub-spectra corresponding to the D0$_3$ structure by means of the binomial distribution of atoms in it, we obtain that such structure is highly ordered, only a 4% of disorder is found on it.

![Fitted Mössbauer spectra. a) Ordered samples. b) Mechanically deformed samples.](image-url)

The ternary alloys do not follow in a straightforward way neither of the Mössbauer patterns shown by the two binary alloys of this series. For a proper fit of the spectra of the ternary alloys it is necessary to use a doublet, instead of a singlet, like in Fe$_{60}$Al$_{40}$ alloy. The appearance of a doublet instead, is due to the different configurations of Al/Si atoms as nearest neighbors of Fe atoms. In the B2 structure the probability of finding a Fe atom surrounded by 8 Fe atoms as nearest neighbors is 16,7%; so a large number of Al/Si atoms surround Fe atoms in this structure. Moreover, each nearest neighbors configuration around one Fe atom gives place to a singlet, but taking into account that adding Si in these alloys the isomer shift increases, the singlets will have an isomer shift that depends on the particular configuration and therefore the peak of the spectrum is formed by singlets with an isomer shift that changes slightly between configurations. Due to the large number of configurations the Mössbauer peak becomes a doublet with not very large quadrupolar momentum. Table 1 shows clearly that increasing the Si content in the alloy the isomer shift increases but on the other hand the quadrupolar splitting decreases.
Table 1. Hyperfine parameters IS (isomer shift), \( \Delta \) (quadrupolar splitting) and Width (line width) obtained from the the Mössbauer spectra of the alloys Fe\(_{60}\)Al(Si)

| % Al | Width (mm/s) | IS (mm/s) | \( \Delta \) (mm/s) |
|------|-------------|-----------|-------------------|
| 12   | 0.42 ± 0.01 | 0.0989 ± 0.0005 | 0.281 ± 0.002 |
| 20   | 0.43 ± 0.01 | 0.0903 ± 0.0003 | 0.288 ± 0.001 |
| 28   | 0.48 ± 0.01 | 0.077 ± 0.001 | 0.295 ± 0.003 |
| 40   | 0.50 ± 0.01 | 0.074 ± 0.001 | |

3.2. Mechanically deformed Fe\(_{60}\)Al\(_{40-x}\)Si\(_{x}\) alloys

Figure 3 shows the Mössbauer spectra of the alloys after mechanical deformation. It is interesting to compare the results obtained after deformation with the ones obtained in the annealed samples. In the case of Fe\(_{60}\)Si\(_{40}\) alloy the width of the doublet obtained in the deformed alloy is very similar to the one obtained in the ordered one. However, there is a clear difference in the sextets corresponding to the D03 structure, the ones of the deformed sample are much wider than the ones of the ordered alloy. So, this result indicates clearly that at the deformation level presented in this work most of the deformation of the alloy occurs in the D03 structure. In the case, of the binary alloy Fe\(_{60}\)Al\(_{40}\), it is clear that the mechanical deformation induces the appearance of a sextet, indicating that mechanical deformation of the binary Fe\(_{60}\)Al\(_{40}\) induces the appearance of a magnetic contribution. This result is in agreement with the results reported in Reference 1 and there have been different studies about the causes of the induction (or enhancement) of magnetism in FeAl alloys [1, 6]. On the other hand, it is interesting to notice that unlike the binary alloys the ternary ones present very similar Mössbauer spectra to the ordered ones. Therefore, this result shows that mechanical deformation does not induce any sextet in the ternary alloys, indicating that in opposition to what happens in FeAl binary alloys mechanical deformation does not cause any magnetic contribution in the studied ternary alloys.

4. Conclusions

The Mössbauer study performed in this work shows clearly that in ordered alloys Al addition to Fe\(_{60}\)Si\(_{40}\) changes the structural order and destroys the magnetic contribution present in it. In mechanically deformed alloys the deformation produces a variation of the magnetic contribution present in ordered Fe\(_{60}\)Si\(_{40}\) alloy and gives place to a new magnetic contribution in Fe\(_{60}\)Al\(_{40}\) alloy. However, no magnetic contribution is observed after mechanical deformation in the studied ternary alloys.

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