

## Effect of Si on the magnetic properties of the Fe<sub>70</sub>Al<sub>30</sub> alloy

G.A. Pérez Alcázar<sup>a,\*</sup>, Ligia. E. Zamora<sup>a</sup>, J.D. Betancur-Ríos<sup>a</sup>, J.A. Tabares<sup>a</sup>,  
J.M. Greneche<sup>b</sup>, J.M. González<sup>c</sup>

<sup>a</sup>Departamento de Física, Universidad del Valle, A. A. 25360, Cali, Colombia

<sup>b</sup>Laboratoire de Physique de l'Etat Condensé, UMR CNRS 6087, Université du Maine, 72085 Le Mans, Cedex 9, France

<sup>c</sup>Instituto de Ciencia de Materiales de Madrid-CSIC, Cantoblanco, 28049 Madrid, Spain

### Abstract

(Fe<sub>70</sub>Al<sub>30</sub>)<sub>100-x</sub>Si<sub>x</sub> ( $x = 0, 5, 10$  and  $20$ ) alloys were obtained by mechanical milling for durations of 12, 24 and 36 h. They were studied by X-ray diffraction (XRD), Mössbauer spectrometry and magnetic measurements at room temperature. The XRD experiments show that all the samples have the Fe-BCC phase and for  $x = 20$  an additional Fe<sub>0.34</sub>Si<sub>0.66</sub> phase appears. The obtained Mössbauer spectra show a decreasing broad magnetic component and increasing broad paramagnetic component when the Si concentration increases. The spectra were fitted by using a hyperfine field distribution and a quadrupolar splitting distribution, respectively. The broad spectra show the disordered character of the samples. When the milling time increases the mean HF increases. These results suggest that Si acts as magnetic dilutor and that the milling time increases the magnetism. All the samples present magnetic hysteresis cycles in which the coercive field decreases with the Si increase, improving in this way the semi-soft character of the alloy.

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### 1. Introduction

Mechanical alloying (MA) has been used to produce a large variety of interesting powdered materials, for commercial as well as academic sectors, due their technological applications and the scientific interest, mainly in the area of nanomaterials [1,2]. In this preparation technique, the reactions occur longer from the equilibrium conditions, and with this high grain refinement while amorphization of the products can be achieved. Concerning the Fe–Al alloys, the system studied in this work, they are cheap soft magnetic materials which has been used in electric power transformers and additionally they are very interesting materials from the experimental and theoretical point of view [3–5]. Apiñaniz et al. [6] evaluated the evolution of the magnetic properties of the Fe<sub>70</sub>Al<sub>30</sub> alloy for different milling times: they found that

the magnetic behavior changes and the system tends to be disordered. Otherwise, the Fe–Si system has been studied for a long time due to its strong use as soft magnetic material, in motors, transformers, and flux multipliers and in high-frequency electronic devices (GHz). The nanostructured powders are studied in order to improve their magnetic properties and to compete thus with amorphous materials [7]. Several Fe–Si alloys have been prepared by MA looking for the amorphization of the system; however until 100 h milling the amorphization was not completely achieved [8]. In 1997, Tie et al. [9] studied the Fe<sub>0.6</sub>Si<sub>0.4</sub> system prepared by MA for different milling times, and they found the Fe<sub>5</sub>Si<sub>3</sub> phase, showing that a high-temperature phase can be obtained by MA at room temperature (RT).

Motivated by the applications of these two systems, we study the magnetic properties of Fe–Al, adding Si system prepared by MA for different milling times. In this work the results obtained using X-ray diffraction (XRD), magnetometry and Mössbauer spectrometry are presented and discussed.

\*Corresponding author. Tel.: +57 2 3212385; fax: +57 2 3393237.

E-mail address: [geperez@univalle.edu.co](mailto:geperez@univalle.edu.co) (G.A. Pérez Alcázar).

## 2. Experimental procedure

The sample series  $(\text{Fe}_{70}\text{Al}_{30})_{100-x}\text{Si}_x$  ( $x = 0, 5, 10$  and  $20$ ) were prepared by MA with high purity Fe, Al and Si parent powders, in a Fritsch-Pulverisette 5 planetary mill (vacuum of  $4.5 \times 10^{-2}$  Torr and a frequency of 280 rpm). Milling times of 12, 24 and 36 h and ball mass to powder mass ratio of 15:1 were considered. The Mössbauer measurements were performed using a conventional Mössbauer spectrometer (MS) at RT, with a 25 mCi  $\gamma$ -source of  $^{57}\text{Co}/\text{Rh}$ . The spectra were fitted using the MOSFIT program [10] while the isomer shift values are referred that of the  $\alpha$ -Fe at RT. The XRD patterns of the samples were performed at RT using the  $\text{Cu}/\text{K}\alpha$  radiation and they were refined by the Maud program [11], allowing the structural phases, the lattice parameters and the grain sizes to be determined. Hysteresis cycles were performed by using a conventional VSM.

## 3. Results and discussion

The XRD patterns for samples with  $x = 5, 10$  and  $20$  (not showed here), milled during 36 h present the BCC phase reported for melted binary  $\text{Fe}_{70}\text{Al}_{30}$  alloy [3]. The lattice parameter decreases from 2.900 Å for  $x = 5$  to 2.842 Å for  $x = 20$  and the grain size changes from 96 to 214 nm. The change of lattice parameter is a consequence of the substitution of Fe and Al atoms by Si atoms which present low atomic size. For  $x = 20$  the pattern presents

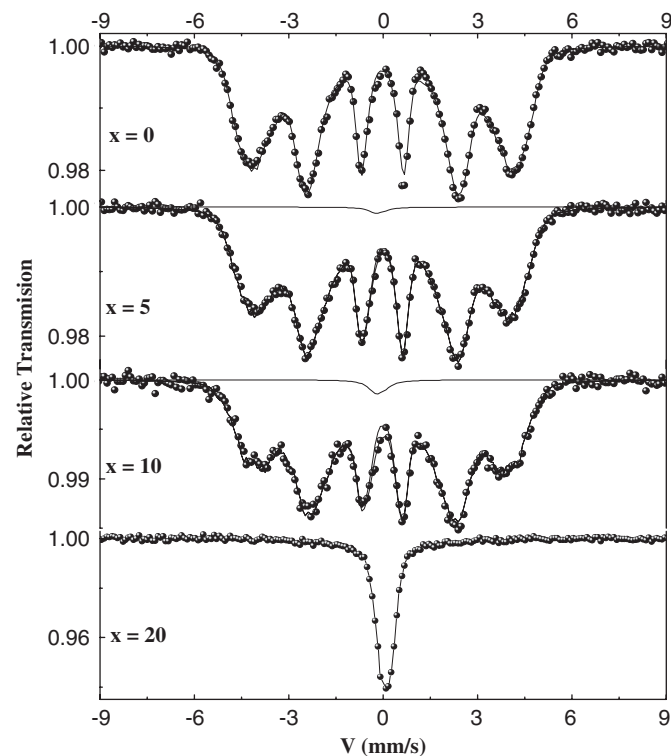


Fig. 1. Mössbauer spectra for samples milled for 36 h and with different Si contents.

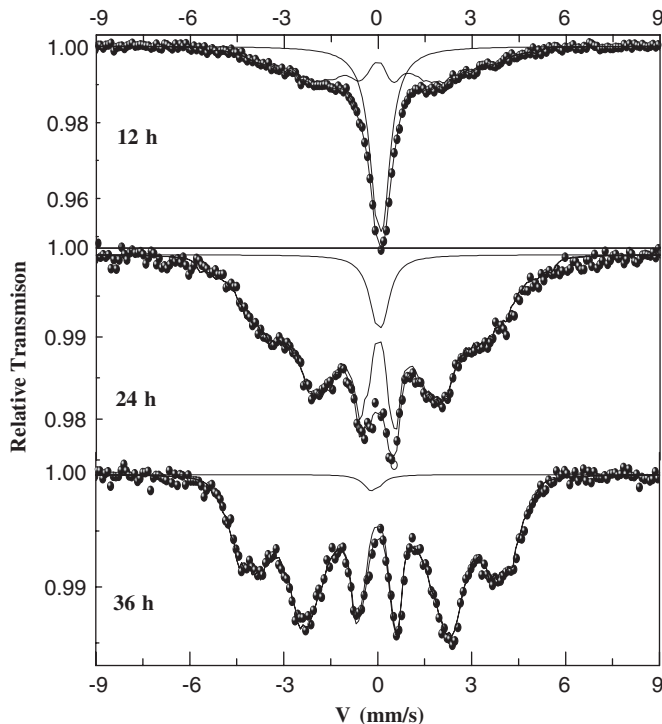


Fig. 2. Mössbauer spectra for  $x = 10$  and different milling times.

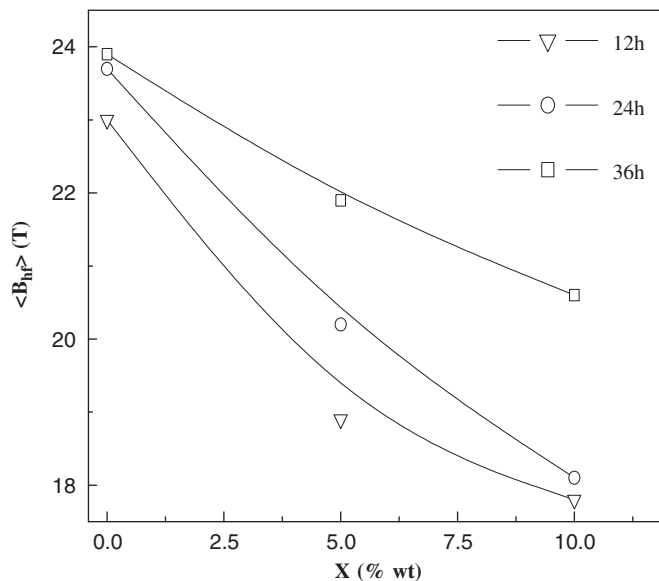


Fig. 3. Mean hyperfine field vs. Si content for samples milled for different durations.

two additional small peaks, one at low angles and other between the first two peaks of the BCC phase. These additional peaks correspond to the  $\text{Fe}_{0.34}\text{Si}_{0.66}$  cubic phase with a lattice parameter of 4.023 Å, and a grain size of 61.0 nm. The quantity of this phase is of 16%. This means that when Si is added it induces the precipitation of this fragile phase. XRD patterns for other samples present a similar behavior.

Table 1

Mössbauer parameters, saturation magnetization and coercive fields obtained from the fit of the spectra and the hysteresis cycles for samples of the  $(\text{Fe}_{70}\text{Al}_{30})_{100-x}\text{Si}_x$  system

Sample		$\langle \text{IS} \rangle$ (mm/s)	$\Gamma$ (mm/s)	$2\varepsilon$ (mm/s)	$\langle B_{\text{hf}} \rangle$ (T)	Area (%)	$M_s$ (emu/g)	$H_c$ (Oe)
$x = 0$	12 h	0.19		−0.12	23.0	100	162	20
	24 h	0.18		−0.12	23.7	100	152	17
	36 h	0.18		−0.02	23.9	100	164	18
$x = 5$	12 h	0.24	0.30		0	6	112	5
		0.18		−0.03	18.9	94		
	24 h	0.06	0.30		0	2	127	4
		0.10		−0.04	20.2	98		
	36 h	−0.06	0.30		0	1	145	4
0.09		−0.04		21.9	99			
$x = 10$	12 h	0.20	0.32		0	44	76	N
		0.11		−0.02	17.8	56		
	24 h	0.17	0.33		0	7	107	N
		0.11		−0.03	18.1	93		
	36 h	−0.02	0.30		0	3	125	N
0.10		−0.04		21.0	97			

N: Not possible to be measured with the VSM used.

Fig. 1 shows the Mössbauer spectra of the previous samples. The spectrum for  $x = 0$  was fitted with a hyperfine field distribution (HFD) and those with  $x = 5$  and 10 with a HFD and an (increasing) paramagnetic site. The spectrum for  $x = 20$  was fitted with a quadrupolar splitting distribution. This type of fit shows the disordered character of the samples. Also it is noted that when Si substitutes the Fe and Al atoms, the lattice contracts and the ferromagnetic behavior reported for the binary system [3] decreases. These results show the dilution of the magnetic properties of the system induced by Si atoms. The HFD for  $x = 0$ , not showed here, shows three most probable Fe sites at 30.4, 27.4 and 16.0 T which correspond to configurations with (7Fe + 1Al), (6Fe + 2Al) and (4Fe + 4Al) atoms as nearest neighbors (nn), respectively [3]. When Si atoms are added into the lattice these sites are displaced to lower fields and some of them appear as paramagnetic, explaining in this way the increasing paramagnetic contribution of the spectra.

Fig. 2 shows the Mössbauer spectra and their fits for  $x = 10$  at different milling times. It can be noted that the intensity of the paramagnetic line decreases, whereas the magnetic character increases while the milling times increases. Fig. 3 resumes the effect in the mean hyperfine field (MHF) of the milling time and the Si content in the  $\text{Fe}_{70}\text{Al}_{30}$  system. It can be observed that the MHF increases when the milling time increases, except for  $x = 20$ , for which it is zero. In the same way the MHF decreases with the increases of the Si content in the  $\text{Fe}_{70}\text{Al}_{30}$  system. The increase of the MHF with the milling time is a consequence of the increasing of the disorder. Table 1 shows the Mössbauer parameters obtained from the fits of all previous spectra. The  $x = 20$  spectra (not showed here) were all fitted with quadrupolar splitting distributions showing the paramagnetic character of these samples.

Finally the hysteresis cycles for samples milled during 36 h (not showed here) are typical of soft materials and in Table 1 the obtained saturation magnetization  $M_s$  and coercive field  $H_c$  values are reported for all the samples. It can be noted that the increase of the Si content decreases the  $M_s$  values. However,  $M_s$  increases and  $H_c$  decreases when the milling time increases, improving thus the soft-magnetic character. The N in this table means that it was not possible to be measured with the VSM used.

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