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Defect Study on Deformed Fe–Si Alloys by Positron Annihilation Techniques

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A set of Fe–Si samples with different Si content, varying from 0 to 7.6 wt.%, was investigated using positron annihilation spectroscopy. Different deformations were performed on these samples and the lineshape parameters of the annihilation line were measured in order to investigate the defects in the alloys. An exponential decay of the *S*-parameter in function of the time can be observed after deformation of the samples. The decay constant increases with increasing Si content. During the measurements of the samples with increasing Si content, the initiation of *DO*₃ structure can be seen above 5 wt.% of Si, as well as the decoration by impurities.

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1. Introduction

Fe–Si alloys combine low magnetostriction with high saturation magnetization, which makes these materials especially suitable for the construction of transformers. Despite the improvement of the magnetic properties with increasing Si content, the concentration of this element is limited to about 4 wt.% in the commercial products. This limit is due to the drastic reduction in ductility for compositions above about 4 wt.% of Si which causes problems in the rolling process used in sheet production [1]. Interest in the improvement of these commercial products has resulted in great efforts towards the development of new processing routes for the production of Fe–Si alloys with 5.5–6.0 wt.% of Si. Besides the Si

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content, the magnetic properties such as coercive force, saturation magnetization, and power loss are strongly influenced by the structural state, grain size, and defect density.

The rolling process on industrial level includes a hot rolling followed by a cold rolling. The cold rolling cannot be executed after a certain time ($\sim$ days) due to a loss of ductility [2]. This process is still not understood. In this work, deformed Fe–Si samples with different Si content were observed with positron annihilation spectroscopy.

2. Experimental details

Several Fe–Si alloys with compositions from 2.3 to 7.6 wt.% Si were prepared by casting at a laboratory scale. Disks with a thickness of 2.5 mm were cut by spark erosion from a bar of raw material (diameter of 12.5 mm) and annealed at 1050°C for 2 h in an induction furnace under vacuum. The samples were slowly cooled (2°C/min) in the furnace to obtain a defect-free state. In order to remove the thin oxide layer, all the samples were chemically polished in a mixture of 6 vol% HF + 25 vol% H₂O₂ + 69 vol% H₂O and carefully washed in pure ethanol. Uniaxial deformations of different thickness reduction were performed at room temperature by pressing samples between two steel blocks under an hydraulic press. Commercial ultra-low carbon (ULC) steel and two very pure Fe samples (3N8 and 4N8) were cut and annealed in the same way in order to compare the results. In addition a set of pure Si and graphite was in our disposition. All measurements were started less than 35 min after deformation. A detail of the chemical composition of the samples can be found in Table I.

TABLE I

Chemical composition of the Fe–Si alloys.

Sample	Si wt.%	C wt.%	Al wt.%	Mn wt.%	N wt.%	S wt.%	Cr wt.%
ULC steel	< 0.005	0.0046	0.0348		0.006		
Alloy 1	2.3	0.005	0.003	0.041	0.03	0.006	0.035
Alloy 2	3.66	0.002	0.009	0.036	0.011	0.006	0.03
Alloy 3	5.64	0.001	0.044	0.13	0.002	0.007	
Alloy 4	7.6	0.002	0.12	0.005	0.005	0.005	0.029

The Doppler broadening (DB) of the 511 keV annihilation line has been measured with a Canberra high purity germanium detector with a resolution of 1.2 keV at the 514 keV line of ⁸⁵Sr. Coincidence Doppler broadening (CDB) measurements were performed on a setup consisting of two Canberra high purity germanium detectors coupled to two digital signal processor (DSP) units. The final resolution is 0.92 keV at the 514 keV line of ⁸⁵Sr.

3. Results and discussion

3.1. Doppler broadening measurements

Different samples were deformed as explained above. The lineshape of the annihilation line was characterized by the well-known S - and W -parameters, representing the annihilation of the positron, respectively, with low energy conduction electrons and high energy core electrons. A first effect which was observed is an exponential (time) decay of the S -parameter (Fig. 1). The same effect was visible in the W -parameter but as a “growth”.

Fig. 1. Evolution of S_{norm} and W_{norm} after a deformation of 4.5% (the curves have been shifted in height for a better visibility).

The fitting has been done by a simple exponential decay of the first order (with $L = S$ or W):

$$L_{\text{norm}} = e^{-t_1/\tau} \quad \text{with} \quad L_{\text{norm}} = \frac{L(t_1) - L(\infty)}{L(0) - L(\infty)}. \quad (1)$$

In Eq. (1), t_1 represents the measuring time. $t_{1,\text{start}}$ represents the beginning of the measurement and it is different from the time t_0 when the deformation is performed ($t_{1,\text{start}} - t_0 = 35$ min). The values obtained after the fitting are presented in Table II.

TABLE II
Decay constants for S_{norm} and W_{norm} after a deformation of 4.5%.

Si-content	$\tau(S_{\text{norm}})$ (in hours)	$\tau(W_{\text{norm}})$ (in hours)
3.66 wt. %	89.3 ± 0.9	83.2 ± 0.8
5.64 wt. %	97.5 ± 2.4	91.8 ± 2.1
7.6 wt. %	153.4 ± 2.1	156.7 ± 1.4

The decay constant increases with increasing Si content. This result could be related to the fact that the sheets cannot be rolled after an amount of time [2] where they might have reached a state similar to the saturation state of the S - and W -parameter.

The evolution of the lineshape parameters can be presented in a SW -plot (Fig. 2).

Fig. 2. SW -evolution for different samples (DB).

Comparing the deformed ULC and Fe-Si alloys with the pure undeformed iron samples (Fe(3N8) and Fe(4N8)), one can see an increase in the S -parameter due to the defects introduced after deformation. The purity of the samples can be compared in Fig. 2b, c, where the different Fe samples (ULC, Fe(3N8), and Fe(4N8)) lie on the same line but for a more pure sample, the S decreases and W increases. The Fe(3N8) has been measured twice to check if the measurements are reproducible or not. The geometry and count rate used in all the measurements were the same, giving reliable results.

3.2. Coincidence Doppler broadening measurements

The same set of samples was prepared and measured after the saturation state was reached ($t_1 = \infty$). The SW correlation is shown in Fig. 3.

Fig. 3. SW -evolution for different samples (CDB).

The deformed iron samples (ULC and Fe(4N8)) and undeformed iron (Fe(4N8)) appear on one straight line (dot line) in accordance to previous measurements. The initiation of the DO_3 structure is around 6 wt.% Si [3–5]. The evolution observed for the different samples can be an indication for that initiation (solid line), where for increasing Si content the S -parameter increases and W -parameter decreases till a value between alloy 3 (5.64 wt.% Si) and alloy 4 (7.6 wt.% Si) and then evolves in the opposite way. The low values in W for Si and C indicate the lowering of the W -parameter of the alloys compared to Fe(4N8). The two dotted lines evolve in the direction of C probably due to the decoration by C. Other elements were not taken into account as they do not affect the overall evolution [6].

4. Conclusion

A decaying effect of the S -parameter was observed after deformation of the Fe–Si samples. The decay constant increases with increasing Si content. The initiation of the DO_3 structure as well as the decoration by impurities were observed by the use of positron annihilation spectroscopy.

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