Defect evolution during annealing of deformed FeSi alloys studied by positron annihilation spectroscopy

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High silicon steel is widely used in electrical appliances. Alloying iron with silicon improves its magnetic performance. A silicon content up to 6.5 wt. % gives excellent magnetic properties such as high saturation magnetization, near zero magnetostriction and low iron loss in high frequencies. Their workability is greatly reduced by the appearance of ordered structures, namely B2 and $D0_3$, as soon as the Si content becomes higher than 3.5 wt. %. This limits the mass production by conventional rolling to this maximum percentage of Si. In this work a series of FeSi (7.5 wt. % Si) samples with different degrees of deformation are investigated with positron annihilation spectroscopy and optical microscopy (OM). The influence of annealing on the concentration of defects of different deformed FeSi alloys has been investigated by positron annihilation lifetime spectroscopy and Doppler broadening of the annihilation radiation. OM is used to investigate the microstructure of deformed samples before and after annealing. The values of the *S* parameter present a decrease for all studied FeSi alloys with the increase of the annealing temperature, being attributed to a decrease of the concentration of defects. A sudden increase of the *S*-parameter value at 600 °C was observed for all samples, which could be related to the change of the ordering of the FeSi alloys at that temperature. At 700 °C, the values of the *S* parameter decreased drastically and starting from 900 °C, they became constant. The microstructures of the alloys, investigated by OM, show that recrystallization is completed at 900 °C and the samples are mainly free of defects, which is in agreement with the positron annihilation lifetime data. © *2011 American Institute of Physics*. [doi:10.1063/1.3535424]

The development of advanced techniques for materials processing requires a corresponding improvement of the materials characterization techniques and development of new ones. Positron annihilation spectroscopy (PAS) is considered to be one of the very important and promising techniques in characterizing materials.[1] The method relies on the propensity of positrons to become localized at open-volume regions of a solid and the emission of annihilation gamma rays that escape the system without any interaction. These gamma quanta hold information about the electronic environment around the annihilation site.[1]

The binary FeSi phase diagram[2] is shown in Fig. 1. According to the diagram, annealing of the FeSi (7.5 wt. % Si) could lead to the change of ordering from ($B2 + D0_3$) at room temperature to $D0_3$ at temperatures above T_c(∼650°C), which is known as order–disorder temperature. Further increase of the annealing temperature changes the order from $D0_3$ to B2 and above 900 °C the alloy is with fully disordered structure type A2. Therefore in the temperature interval between room temperature and 730 °C the size of $D0_3$ ordered zones increases with the increase in the annealing temperature.[3]

The defect structure in ordered alloys is well known to be much more complicated than in pure metals. The predominant type of crystal latice defect depends on the different structures (B2, $D0_3$) in these materials. Also, the deformation such as rolling induces many dislocations and vacancies, especially the vacancies which are not easy detectable with the classical characterization methods. For this reason powerful techniques should be used to distinguish between those different defects.[4–6]

The material was prepared by melting of ultralow carbon steel together with a given amount of Fe-75%Si master alloy in an open-air induction furnace in order to obtain the required chemical composition presented in Table I. More details about the material preparation can be found in Ref. 7.

After hot rolling and annealing the three samples with different cold reduction were prepared and used in this study as follows: sample 1 with 2.3% thickness reduction, sample 2 with 4.1% thickness reduction, and sample 3 with 18.5% thickness reduction. The samples used for the positron annihilation measurements were annealed isochronally for 1 h in a

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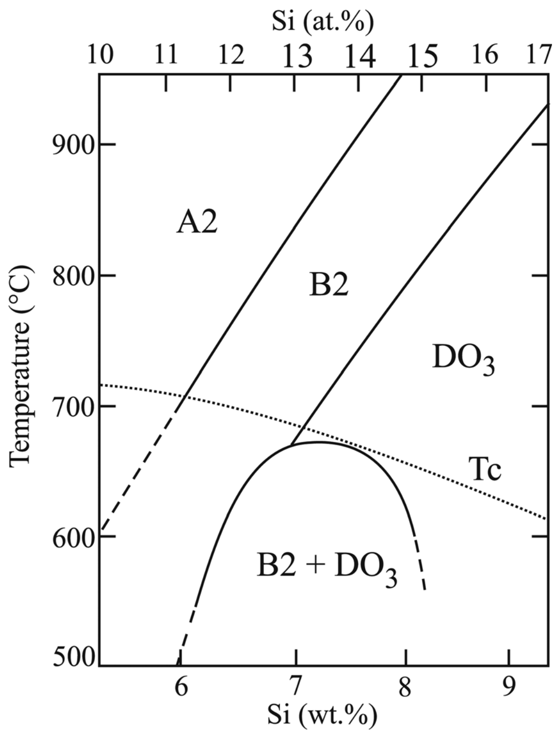

FIG. 1. Part of the binary FeSi diagram (Ref. 2).

tube furnace under vacuum at different temperatures ranging from 100 to 900 °C and air cooled in the tube.

Positron annihilation lifetime (PALT) measurements were performed at room temperature using a fast–fast lifetime spectrometer.[5] Each spectrum contained more than 10^6 counts and several spectra were accumulated for each measuring point in order to ensure the reproducibility of the data. The Doppler broadening of the 511 keV annihilation line was measured using a high purity germanium detector with a resolution of 1.2 keV at the 514 keV line of ^{85}Sr. The results were analyzed in terms of the so-called S parameter, which is defined as the area under the central part of the annihilation photopeak divided by the total area, and it is a measure of the electron momentum distribution at the annihilation site.[1,5] The vacancy-type defects act as trapping sites for positrons and annihilation with low energy valence electrons at these defects results in a narrowing of the photopeak corresponding to an increase in the S parameter.

To obtain the optical micrographics the standard procedure for sample preparation by grinding and polishing was used, finishing in the last step with polishing with OPS (colloidal silica suspension) in ethanol. The microstructure was revealed by etching with 4% solution of nitric acid in alcohol (Nital), which is commonly used etchant for carbon and low alloyed steels.

It is known that cold rolling induces many defects such as dislocations and vacancies in the crystal lattices of metals and alloys which make their structure thermodynamically unstable,

TABLE I. Chemical composition (wt. %) For the Fe-Si alloys.

Element	% C	% Si	% Al	% Mn	% P	% S
wt. %	0.001	7.5	0.001	0.03	0.007	0.006

TABLE II. PALT measurement data for the 18.54% thickness reduction Fe7.5 Si alloy (sample 3).

Sample	τ_1(ps)	τ_2(ps)	τ_3(ps)	I_2	I_3
Deformed sample 3 (18.5% thickness reduction)	75 ± 3	166 ± 4	0	90 ± 4	0
Annealed at 600 °C	81	160	264 ± 31	65.4 ± 3	10.2 ± 2
Annealed at 650 °C	84 ± 3.7	165	230 ± 22	37 ± 2	12.3 ± 2
Annealed at 900 °C	107				

i.e., with over equilibrium defects concentration. These defects have a structure that is significantly different from the structure of the "normal" crystal lattice and they could be detected and quantified via the positron annihilation lifetime. Table II represents the positron annihilation lifetime data for the FeSi alloy deformed to 18.54% thickness reduction (sample 3) and after annealing for 1 h at different temperatures. For the deformed sample, the positron lifetime spectra could be fitted by two distinct lifetime components (τ_1 and τ_2) with relative intensities I_1 and I_2, respectively. The lifetime for positrons annihilated in defects (τ_2) is found to be around 160 ps, but the two state trapping model[8] could not be applied in the way it should be. This is probably due to the rather complex defect structure present in the material. It is important to take into consideration that the super dislocations of the $D0_3$ phase have been detected in alloys with Si contents above 6.5 wt. %,[9,10] which could affect the analysis of the highly deformed FeSi alloys. It is known that the value of the lifetime of positrons trapped in dislocations is close or slightly below the vacancy lifetime.[11,12] For this reason, it is easy to believe that the 160 ps positron annihilation lifetime value is related to vacancies trapped in the stress field around a dislocation line or in vacancies on a dislocation line. The PALT data suggested that no change of the defect type as a function of annealing time is observed at temperatures below 600 °C. Significant amounts of thermal vacancy clustering in FeSi alloys are created at the annealing temperature 600 °C, and 650 °C as shown in Table II. For the annealed sample at 600 °C, a third lifetime with a value of 264 ps with intensity of 10.2% is observed, which according to Nagai *et al.*[13] could be related to vacancy clustering in the FeSi alloy.

At 650 °C τ_3 becomes 230 ps and its intensity increased to be 12.3%. This longer lifetime component (τ_3) of 230 ps corresponds to a cluster of four vacancies cluster in the FeSi alloy.[13] After the annealing of samples at 900 °C for 1 h, only one lifetime component exists with a value of 107 ps, which is similar to that of the well-annealed pure Fe. This means that vacancies trapped in the stress field around a dislocation line (τ_2) and vacancy clusters (τ_3) are annealed out with the increase of the annealing temperature and the samples became mainly free of defects [concentrations in the samples are less than the lower detection limit of ($\sim 10^{-6}$) of the positron annihilation technique].[13]

The dependence of the S parameter on the isochronal annealing temperature for different deformed FeSi samples is shown in Fig. 2. In the temperature range (20–500 °C) the S parameter slightly decreases. A peak which appears at 600 °C is due to vacancy clustering (detected by the PALT measurements) which can be related to ordering effects ($B2 + D0_3 \rightarrow D0_3$). Viala *et al.*[14] observed FeSi alloys with a 6.5 wt. % Si formation of a

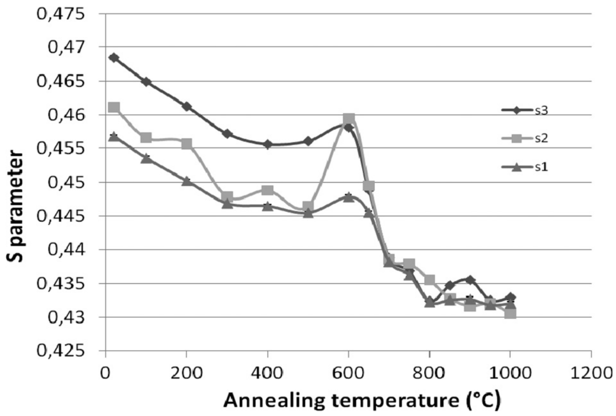

FIG. 2. The annihilation line shape parameter (S) as a function of the isochronal annealing temperature of differently deformed FeSi samples [s1 with 2.3% deformation, s2 with 4.12% deformation, and s3 with 18.54% deformation (annealing time 1 h)].

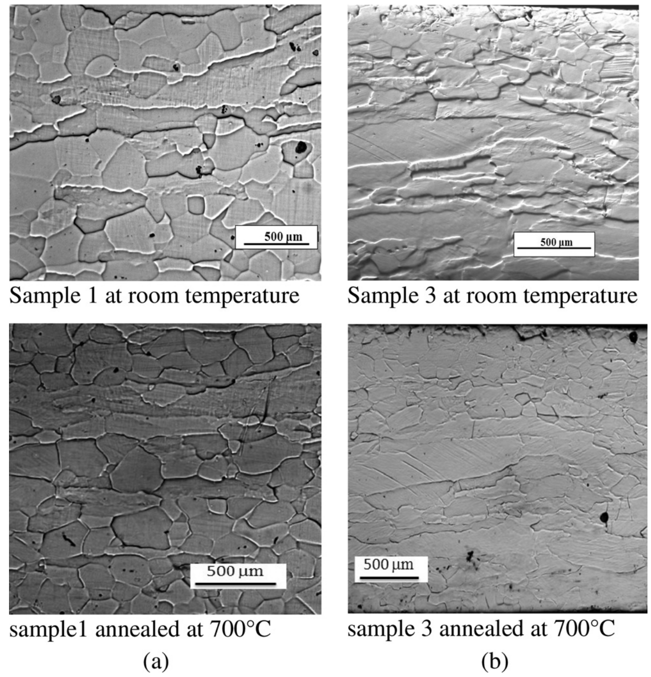

FIG. 3. Light optical microstructures (OM) for the FeSi alloy: (a) sample 1 deformed to a thickness reduction of 2.3% and annealed at 700 °C for 1 h and (b) sample 3 deformed to a thickness reduction of 18.54% and annealed at 700 °C for 1 h.

$D0_3$ superlattice through nucleation within the existing B2 domains. This $D0_3$ is formed at an annealing temperature ~650 °C. Also Swann *et al.*[2] identified the $D0_3$ precipitates in the B2 matrix of the 6.4% Si steel when the samples were held at 600 °C for 24 h. These previous results show that at nearly 600 °C, the $D0_3$ domains are formed in the B2 matrix of high Si electrical steels with 6.5–7.5% Si. After that, a significant decrease of the value of the S parameter is observed, which indicates that defects are annealed out after the heat treatment at temperatures higher than 650 °C. Starting from the annealing temperature of 850 °C, the S parameter became almost constant.

Figure 3 shows the microstructure of the FeSi alloy after deformation by rolling to a thickness reduction of 2.3% (sample 1) and 18.54% (sample 3) and after annealing of the deformed samples at 700 °C for 1 h. The optical microscope images show that the heavily deformed sample (sample 3 with 18.54% thickness reduction) has a higher amount of shear bands within pancake grains elongated along the rolling direction than the low deformed one (sample 1 with 2.3% thickness reduction). Microcracks could be observed in the micrographs of the heavy deformed sample [see Fig. 3(b)]. After annealing at 700 °C, the microstructure of the samples remains unchanged and the shear bands in both samples are the same before and after the annealing, which means that samples 1 and 3 are not recrystallized at all [see Figs. 3(a) and 3(b)]. The results from the optical microscopy reveal that the observed changes in the annihilation line shape parameter (S) should be associated with recovery but not with recrystallization processes.

In this paper the recovery and recrystallization of a cold deformed Fe7.5% Si alloy were studied by PAS and optical microscopy (OM). The PAS is proved to be a very sensitive technique that can detect small structural changes in the material even when the microstructural changes are not observed by OM method.

The PAS data suggested the following. The lifetime for positrons annihilated in defects (τ_2) is found to be around 160 ps in the deformed samples in the temperature range 20–500 °C, which is believed to be related to vacancies trapped in the stress field around a dislocation line or in vacancies on a dislocation line.

No change of the defect type with annealing is observed before 600 °C. At 600 °C, vacancy clustering in the FeSi alloys is observed.

The dependence of the S parameter on the isochronal annealing temperature shows that in the temperature range (20–500 °C) the S parameter slightly decreases and a peak appears at 600 °C. This peak is due to vacancy clustering that can be related to ordering effects ($B2 + D0_3 \rightarrow D0_3$), which is also detected by the PALT measurements. After that a significant decrease of the value of the S parameter can be observed, which indicates that defects are annealed out after the annealing at temperatures higher than 600 °C.

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