Influence of atomic order on magnetic properties of Fe–Si alloys

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Abstract

The present work attempts to assess the influence of the order phenomenon on the soft magnetic properties of Fe–Si alloys in a wide concentration range. Alloys with Si content up to 7.6 wt% were prepared. Heat treatments including annealing at different temperatures ranging from 500 up to 1100 \( {^{\circ}}\)C and water quench were carried out in order to develop different states of order. \(^{57}\)Fe Mössbauer spectroscopy was used to evaluate and quantify the degree of order. Concerning magnetic properties, total losses and permeability were measured. Results show that magnetic properties are mainly influenced by two factors: stresses induced during quenching and atomic order phenomena. The former deteriorates them as the quenching temperature is increased while the latter becomes more appreciable as the Si content is higher.

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1. Introduction

Steel sheet made of 6.5 wt\% Si constitutes a very suitable material to be applied in electrical devices, such as transformers and motor cores due to its enhanced magnetic properties, which can allow worldwide large-scale energy savings. Nevertheless, the well-known poor ductility of Si-steel above 3 wt\% Si has hindered mass production by conventional hot and cold rolling schedules. The cause of this embrittlement has been usually associated to atomic ordering enhancement which digress the solution of Si atoms into the Fe matrix from the random distribution. Two kinds of order phenomena take place: short-range and long-range order, both of them having a strengthening effect on the mechanical properties of the alloys [1]. However, literature shows different conclusions about the type and degree of order present in the alloys. TEM studies have shown a random solution up to 5 wt\% Si, a mixture of B2 + D0\(_3\) (\(\text{FeSi} + \text{Fe}_2\text{Si}\)) in the 5–7 wt\% Si range and D0\(_3\) order above the latter amount [2], while neutron diffraction and Mössbauer spectroscopy indicate the presence of Fe\(_{13}\)Si and D0\(_3\) even below 5 wt\% Si [3]. In addition, order has been reported as a non-negligible...
parameter for the magnetic properties with controversial results as well about its influence on them [4–7]. This work attempts to characterize the order present in Fe–Si alloys up to 7.6 wt% Si subjected to different ordering heat treatment and attempts to relate the order degree with the magnetic properties, such as power losses and maximum permeability.

2. Experimental work

Five Fe–Si alloys with nominal Si concentrations up to 7.6 wt% were laboratory cast. Table 1 shows their chemical composition. Slices with 60 x 20 x 1.5 mm³ were spark-eroded from the ingots for subsequent heat treatments. Annealing for 2h at different temperatures (500, 700, 900, 1100 °C) in a protective atmosphere followed by water quench was applied in order to develop different degrees of order on the samples. In addition slow cooling from 1100 °C was carried out for all the alloys.

Magnetic measurements were performed in a specially designed mini-single strip tester [8]. This miniature SST allows accurate power loss measurements (±5%, calibrated accuracy IEC 404-3). ⁵⁷Fe Mössbauer spectra were obtained in transmission geometry at room temperature. Calibrations were periodically taken with pure Fe foil.

3. Results and discussion

Microstructure and grain size measurements show no big differences between the samples with different heat treatments and Si contents. Results show large equiaxial grains with sizes of 1.2–1.5 mm.

Fig. 1 shows the total power losses and the maximum permeability of the Fe–Si alloys after annealing at several temperatures for 2h followed by quenching. The results for the slow-cooled samples after annealing at 1100 °C during the same time are also shown. The first feature is that losses generally increase with the annealing temperature. In principle, that can be related to the internal stresses induced during high-temperature quenching, which have a detrimental effect on the losses. In addition, an unlike evolution of magnetic properties below and above 5 wt% is noticeable. For the former case, losses of the samples quenched from 1100 and 900 °C have the same value and then decrease with temperature at a high rate. For the latter case (FeSi-6 and higher) there is a continuous decrease of the losses with temperature, but at a slower rate. Concerning the permeability, it follows the same trend: the higher the T is, the lower the permeability. However, a transition can be noticed between 700 and 900 °C, especially for FeSi-6.

A possible explanation for those changes in magnetic properties can be found in the order present in the material, which can modify the domain wall mobility [5], as well as the electrical resistivity [4].

Mössbauer spectroscopy is well known to be an ordersensitive technique due to the strong dependence of the hyperfine parameters on the Fe atoms surroundings. As the environments of Fe atoms change with the degree of order, this can be evaluated with an adequate analysis of the Mössbauer spectra, which is something not trivial in the case of diluted Fe alloys [9]. For this reason, an appropriate program has been developed to fit the spectra. The program introduces 64 Lorentzian-shape sextets corresponding to different configurations for the Fe atoms in the first and second neighborhoods: (x,y), x being the number of Si atoms on the first and y the number of Si atoms on the second neighborhood, respectively. In addition, the site occupancies of the lattice are included as fitting parameters, controlling the

<table>
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<tr>
<th>Sample</th>
<th>Si wt%</th>
<th>Si at%</th>
<th>Al wt%</th>
<th>C wt%</th>
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<tbody>
<tr>
<td>FeSi-3</td>
<td>3.3</td>
<td>6.39</td>
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<td>0.004</td>
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<tr>
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<td>3.8</td>
<td>7.18</td>
<td>0.09</td>
<td>0.03</td>
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<td>FeSi-5</td>
<td>5</td>
<td>9.35</td>
<td>0.09</td>
<td>0.002</td>
</tr>
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<td>FeSi-6</td>
<td>5.6</td>
<td>10.56</td>
<td>0.04</td>
<td>0.001</td>
</tr>
<tr>
<td>FeSi-8</td>
<td>7.6</td>
<td>14.01</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>
areas of the different spectra. From them it is possible to deduce the long-range order parameters following the Bragg–Williams model. The details of the program will be published elsewhere [10]. Three spectra of FeSi-6 with different heat treatments are shown in Fig. 2, with their corresponding components. Fig. 3 gives the long-range order parameters (LROP) of the Fe$_{15}$Si, D0$_3$ and B2 structures for the different samples. Concerning the influence of the Si content, the lower Si-alloys (FeSi-3 and FeSi-4) shows low D0$_3$ and B2 LROPs and a predominant presence of Fe$_{15}$Si order. On the other hand, FeSi-5 has an increased D0$_3$ LROP, while FeSi-6 shows an enhanced B2 LROP. FeSi-8 shows predominant presence of D0$_3$ and B2 order and very low value for the Fe$_{15}$Si LROP. It can be concluded that there is always a tendency for ordering in the FeSi alloys, even for the lower Si contents, since no spectra could correctly be fitted with the assumption of random distribution of the atoms. In addition a tendency for large interatomic Si–Si distances is detected, since order changes from Fe$_{15}$Si towards Fe$_3$Si and B2 as the Si content is increased. About the influence of the quench temperature on the order, two remarks can be made: there is a slight change on FeSi-3, FeSi-4 and FeSi-5, pointing to higher degrees of D0$_3$ and B2 order with lower quench temperatures. Fe-6 is the alloys showing the larger changes in order: as the temperature is increased, D0$_3$ order grows at expenses of reducing B2 order. FeSi-8 only shows a small change at 1100°C pointing to a higher degree of D0$_3$ order.

Finally, concerning the influence of order on the magnetic properties it does not seem not easy to discern between the influence of order and the influence of stresses induced during quenching. Although up to 5 wt% the differences in degree of order are very slight, the increase of losses occurring at 900°C might be related to the increased LROPs. On the other hand, the suppression and substitution of D0$_3$ order for B2 one seems to have a beneficial effect for the losses as well agreeing with former works [6].

4. Conclusions

The magnetic properties of several Fe–Si alloys were largely changed by means of thermal ordering treatments. Two effects were clearly detected: internal stresses produced by quenching, which deteriorate them and changes in the degree of order, which does not have large effect, except for high Si content. In this sense, FeSi-6 showed that substitution of D0$_3$ order by B2 seems to have a beneficial effect on the magnetic...
properties. In the other alloys, order seems to be detrimental for them. The reason might be due to the interactions of the ordered structures with the movement of the domain walls.

References