Preannealing effect and soft magnetic properties of the nanocrystalline 
Fe$_7$Cu$_1$Nb$_3$Si$_{12}$B$_{10}$ alloy

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(Received: May 12, 1998 Accepted: August 26, 1998)

The magnetic susceptibility, its disaccommodation and microstructure of the nanocrystalline Fe$_7$Cu$_1$Nb$_3$Si$_{12}$B$_{10}$ alloy obtained by the crystallization of amorphous ribbons (after different preannealing) are investigated. It is stated that the preannealing leads to the increase of the magnetic susceptibility for the nanocrystalline samples. It is due to the narrowed size distribution of α-FeSi grains in these samples. However, the disaccommodation intensity of the nanocrystalline samples is independent of preannealing conditions.

Key words: magnetic susceptibility, disaccommodation.

1. Introduction

Fe-Cu-Nb-Si-B alloys with the nanocrystalline structure were developed by Yoshizawa and co-workers$^{1,2}$. Nanocrystalline materials which are obtained by partial crystallization of amorphous alloys consist of ultrafine crystalline grains (10-20 nm diameter) embedded in the remaining amorphous matrix. The presence of fine grains causes the suppression of the magnetocrystalline anisotropy and leads to superior soft magnetic properties$^{3}$. It is found$^{4,5}$ that the magnetic properties of nanocrystalline alloys can be affected by the structure of the as-quenched amorphous phase which depends on preparation conditions.

In this paper we present results of investigations of magnetic properties (susceptibility and its disaccommodation) and microstructure for the samples of the nanocrystalline Fe$_7$Cu$_1$Nb$_3$Si$_{12}$B$_{10}$ alloy subjected to the different preannealings.

2. Experimental Procedure

The amorphous alloy in the form of ribbons 22 μm thick and 10 mm wide was produced by the rapid quenching technique on a single roller. The magnetic susceptibility was measured by means of a completely automated set-up in the temperature range from 150 K up to 650 K. The amplitude and frequency of the magnetizing field were 0.16 A/m. and 2 KHz, respectively. From these results the isochronal disaccommodation curves were constructed such as

\[ \Delta \left( \frac{1}{\chi} \right) = \left( \frac{1}{\chi_2} \right) - \left( \frac{1}{\chi_1} \right) = f(T) \]  

where $1/\chi_1$ and $1/\chi_2$ are reciprocal magnetic susceptibilities at times $t_1=2$ s and $t_2 = 120$ s after demagnetization, respectively.

The microstructure of the samples was studied using Mössbauer spectroscopy and X-ray diffractometry. From the analysis of the transmission Mössbauer spectra taken at room temperature the phase composition, content of iron in the crystalline and amorphous phases and type of order in the crystalline phase were determined. Under the assumption that the average hyperfine field is proportional to the iron content in the amorphous phase, the iron content in the amorphous matrix and its volume fraction were estimated from the equations:

\[ Fe_{am} = \frac{74at\% \times <B_{am}>}{<B_{ar-q}>} \]  

and \[ V_{am} = \frac{R \times <B_{ar-q}>}{B_{am}} \]

where R is the relative area of subspectra corresponding to the amorphous matrix, $<B_{ar-q}>$ and $<B_{am}>$ are the average hyperfine fields for the as-quenched samples and amorphous matrix.

The short range order parameter ($\alpha_s$) of the crystalline α-FeSi phase was calculated from the equation:

\[ \alpha_s = \frac{n - n_0}{n_0} \]  

where $n_0$ is the average number of Si atoms in the neighbourhood of Fe atoms arranged randomly in α-FeSi lattice (obtained from binomial distribution) and n is the number of these atoms in the investigated samples (determined from Mössbauer spectra analysis). All investigations were carried out for the nanocrystalline samples of the Fe$_7$Cu$_1$Nb$_3$Si$_{12}$B$_{10}$ alloy obtained by the heat treatment (at 823 K for 10 s and 10 min) of preannealed amorphous ribbons (at 603 and 673 K).
Fig. 1 The temperature dependence of the magnetic susceptibility for the nanocrystalline Fe$_{77}$Cu$_{17}$Nb$_{5}$Si$_{13}$B$_{10}$ alloy (annealed at 823 K for 10 min) preannealed: at 603 K for 20 min and then 673 K for 1 h (■), at 603 K for 20 min (▲), at 673 K for 1 h (×) and without preannealing (○).

3. Results

Fig. 1 shows, as an example, the magnetic susceptibility ($\chi$) for the nanocrystalline samples of Fe$_{77}$Cu$_{17}$Nb$_{5}$Si$_{13}$B$_{10}$ alloy obtained by annealing at 823 K for 10 min. Before that treatment the samples were subjected to the different preannealings. It is seen that the nanocrystalline sample preannealed at 603 K for 20 min and then at 673 K for 1 h exhibits the highest value of the magnetic susceptibility equal to about 21000 at room temperature. However, the susceptibility of the nanocrystalline sample without preannealing reaches the value of about 7000 at room temperature.

The disaccommodation of the magnetic susceptibility ($\Delta(1/\chi)$) versus temperature is presented in Fig. 2.

One can notice that the isochronal disaccommodation curves exhibit the similar shape which does not depend on the preannealing conditions of the samples. In the temperature range from 175 K up to 475 K only temperature independent background is observed. The disaccommodation intensity rapidly increases above 550 K.

4. Discussion

From Mössbauer spectroscopy analysis it is found (Table 1) that the amount of the crystalline α-FeSi phase in the nanocrystalline samples almost does not depend on the preannealing conditions. Moreover, the iron content in that phase and amorphous matrix is nearly the same for all investigated samples. Furthermore, it is stated that in the crystalline α-FeSi phase only the short range order occurs (Table 1). It is seen from Fig. 1 that the preannealing of the amorphous ribbon at 603 K for 20 min and then at 673 K for 1 h leads to the highest magnetic susceptibility of the nanocrystalline sample. Furthermore, the nanocrystalline samples of the Fe$_{77}$Cu$_{17}$Nb$_{5}$Si$_{13}$B$_{10}$ alloy obtained by the heat treatment of the amorphous ribbons preannealed at 673 K or 603 K exhibit also the higher magnetic susceptibility than the sample annealed only at 823 K (without preannealing).

Table 1 The effective hyperfine field at $^{57}$Fe nuclei ($B_{hf}$), volume fraction of the crystalline phase ($V_{cr}$), iron content in the crystalline and amorphous phases ($Fe_{cr}$, $Fe_{am}$) and short range order parameter ($\alpha_s$) for the Fe$_{77}$Cu$_{17}$Nb$_{5}$Si$_{13}$B$_{10}$ alloy.

<table>
<thead>
<tr>
<th>Annealing conditions of the samples</th>
<th>$B_{hf}$[T]</th>
<th>$V_{cr}$</th>
<th>$Fe_{cr}$ [at%]</th>
<th>$Fe_{am}$ [at%]</th>
<th>$\alpha_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>603 K/20 min+673 K/1 h+823 K/10 min</td>
<td>23,57</td>
<td>0,47</td>
<td>85</td>
<td>61</td>
<td>-0,36</td>
</tr>
<tr>
<td>603 K/20 min+823 K/10 min</td>
<td>23,15</td>
<td>0,48</td>
<td>84</td>
<td>63</td>
<td>-0,28</td>
</tr>
<tr>
<td>673 K/1 h+823 K/10 min</td>
<td>23,21</td>
<td>0,48</td>
<td>84</td>
<td>63</td>
<td>-0,29</td>
</tr>
<tr>
<td>823 K/10 min</td>
<td>23,42</td>
<td>0,48</td>
<td>84</td>
<td>63</td>
<td>-0,30</td>
</tr>
</tbody>
</table>
The Mössbauer studies of the annealed amorphous samples show that they exhibit a different structure. The average hyperfine fields for the amorphous samples annealed at 603 K for 20 min, at 673 K for 1 h and at 603 K for 20 min plus 673 K for 1 h are equal to 21.4, 21.5 and 21.7 T, respectively. Moreover, the enhancement of the second line intensity in the Zeeman sextets with the increase of the preannealing temperature and time was observed. These results indicate that preannealing leads to the change of the packing density and the stress relief of the samples.

The results obtained from Mössbauer spectra analysis (Table 1) and the magnetic properties studies (Fig. 1) for the nanocrystalline samples may indicate that, due to the preannealing of the samples, the narrowed size distribution of α-FeSi grains in them occurs. This may lead to more effective averaging of the magnetocrystalline anisotropy and explains the increase of the magnetic susceptibility in the nanocrystalline samples of the Fe₇₅Cu₃Nb₂Si₂B₁₀ alloy, subjected to preannealing below the crystallization temperature.

As for the magnetic susceptibility disaccommodation in nanocrystalline samples, it is independent of preannealing conditions (Fig. 2). From these results one may conclude that distribution of grain size does not effect the magnetic susceptibility disaccommodation. Moreover, the observed temperature independent background in the isochronal disaccommodation curves of the nanocrystalline samples in the temperature range from 150 K up to 475 K indicates that the relaxation processes occurring in the amorphous matrix play the dominant role in the magnetic susceptibility disaccommodation of the nanocrystalline samples. The very low disaccommodation intensity in this temperature range is connected with the annealing out of the free volumes in the amorphous matrix during the crystallization of the samples. The increase of the disaccommodation intensity (for all investigated samples) above 475 K is connected with phase transition (from ferromagnetic to paramagnetic state). Similar behaviour was observed for the nanocrystalline samples annealed at 823 K for 10 s.

5. Conclusions

- The preannealing of the amorphous ribbons below the crystallization temperature leads to the increase of the magnetic susceptibility of the nanocrystalline samples.
- The disaccommodation intensity of the nanocrystalline samples does not depend on the preannealing conditions.

References