Magnetic properties of the composition of nanocrystals and amorphous matrix in an alloy of the Fe-Nb-Si-B-Cu system

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Abstract. It is shown that amorphous alloys of the Fe-Nb-Si-B-Cu system are practically not used as soft-magnetic materials in the initial (freshly quenching) state. It is proposed to perform post-quenching annealing of products to improve the soft-magnetic properties. Annealing modes, which remove winding and quenching stresses are studied. It is established the optimal annealing mode, in which a favorable structure is formed. It is an amorphous alloy-nanocrystal composition. This composition can significantly increase the magnetic permeability, reduce the coercive force and the loss of magnetization reversal. This treatment will provide optimal performance characteristics for the amorphous alloy used. The paper studies and compares the magnetic properties of the amorphous alloy-nanocrystal composition obtained at different annealing temperatures. It is shown that the most significant comparison is the dependence of the magnetic induction $B (Tl)$ by the magnetic field strength $H (A/m)$.

Keywords: amorphous alloys, soft-magnetic materials, annealing, composition, nanocrystal.

1. Introduction

Amorphous alloys and nanocrystalline compositions are a relatively new class of magnetic materials compared to traditional crystalline materials [1]. These materials are of great interest because they can have unique physical (including magnetic) properties. Amorphous alloys are used to make both DC device cores and magnetic cores for AC equipment converters in a wide range of magnetization reversal frequencies (from 50 Hz to 200 kHz). In the quenched state, amorphous alloys do not have high magnetic properties [2]. To obtain optimal properties, the alloys are subjected to various thermal and thermomagnetic treatments, similar to those used to improve the properties of electrical steels [3, 4]. Any plastic deformation and stress degrades the soft magnetic properties of these steels [5, 6].

The aim of this work is to improve the soft magnetic properties of Fe-Nb-Si-B-Cu alloys by determining the optimal modes of post-quenching annealing, as well as to study the amorphous alloy-nanocrystals composition formed during annealing.
2. The methodology of the experiment
The research was carried out in production conditions. All properties were measured at room temperature. For the study, samples were made from an amorphous ribbon of the Fe-Nb-Si-B-Cu alloy system. The resulting samples were subjected to heat treatment, namely annealing.

An amorphous alloy of the Fe-Nb-Si-B-Cu system was prepared by rapid quenching of a molten metal jet on the outer surface of a rotating copper drum with a cooling rate of approximately \(10^6 \text{ K/sec}\). The width of the resulting tape was \(0.014-0.016 \text{ m}\), the thickness of \(20-25\times10^{-6} \text{ m}\). The crystallization temperature was determined using a differential thermal analyzer at a heating rate of \(20 \text{ K/min}\).

The resulting samples were subjected to annealing. Samples placed in the furnace were heated to the crystallization temperature, which was measured using a thermocouple placed in the sample itself. Two series of samples were examined: 1) to determine the dependence of the crystallite size on the annealing temperature \((t = 400-700^\circ \text{C})\); 2) to measure the magnetic properties (annealing was varied in the temperature range \(t = 530-620^\circ \text{C}\)). After reaching the crystallization temperature, the samples were removed from the furnace and cooled in air.

Structural studies were performed by x-ray diffractometry. The optimal annealing mode, in which the best magnetic properties are obtained, was established by comparing the dependence of the magnetic induction \(B (T_I)\) on the magnetic field strength \(H (\text{A/m})\).

3. The results of experiments and discussion
Structural studies have shown that the phase composition of the alloy changes after heat treatment. The formation of nanocrystals and a significant simplification of the domain structure are observed (the magnetization is aligned along the magnetic field applied during processing). The formation of a new structure consisting of an amorphous alloy-nanocrystals composition is accompanied by the fact that the total electromagnetic losses are almost halved. The latter is probably mainly related to the eddy current component [7-9].

Studies have shown that in the Fe-Nb-Si-B-Cu alloy, nanocrystals appear after annealing at temperatures above 500-520°C and reach a minimum value of about 25 nm at about \(t = 520^\circ \text{C}\). Heating to higher temperatures leads to a significant increase in grain size (Fig. 1).

![Figure 1. The dependence of the size of crystals on annealing temperature](image)

As can be seen from figure 1, after annealing, starting from \(t = 580^\circ \text{C}\), the size of nanocrystalites is about 25-30 nm and does not change until \(t = 710^\circ \text{C}\). Studies of the second series of samples showed that after annealing at \(t = 520^\circ \text{C}\) and below, the nanocrystalline phase in the alloy is not formed. In order to determine which annealing mode contributes to obtaining the best magnetic properties and forming a favorable structure consisting of the composition – amorphous alloy-nanocrystals, it was necessary to choose more indicative magnetic parameter for comparison. Studies have shown that the values of magnetic induction \(B (T_I)\) are suitable for this purpose. The magnetic induction is influenced...
by the chemical composition of the substance and the magnetic field strength $H \, (A/m)$, which in turn is set by the measuring device. The steepness of the Hysteresis loop will obviously indicate which annealing mode is the best [10, 11]. Thus, it is possible to choose the annealing mode that is most effective and provides the best magnetic properties of the material under study.

After heat treatment, magnetic characteristics were obtained using the control unit for magnetic cores in production conditions «UKMP-0.05-100». The unit provides determination of dynamic magnetic characteristics at a frequency of remagnetization up to 100 kHz in accordance with GOST 12119.4 and GOST 12119.5. Using the program, the values of the magnetic field strength $H \, (A/m)$, magnetic induction $B \, (Tl)$, the value of the loss angle $\phi \, (rad)$, and magnetic permeability $\mu$ were obtained. The program also allows you to build a hysteresis loop that best reflects the magnetic properties of the material. The measurement results are shown in table 1.

### Table 1. The results of measurements of magnetic properties

| Process parameters | sample number | 1   | 2   | 3   | 4   | 5   | 6   | 7   | 8   | 9   | 10  |
|--------------------|--------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| $t_m, ^\circC$     |              | 530 | 540 | 550 | 560 | 570 | 580 | 590 | 600 | 610 | 620 |
| $t_c, ^\circC$     |              | -   | 561 | 569 | 585 | 589 | 591 | 609 | 613 | 618 | 648 |
| $B, Tl$            |              |     |     |     |     |     |     |     |     |     |     |
| $H, A/m$           |              | 0.2 | 0.004 | 0.009 | 0.008 | 0.016 | 0.016 | 0.037 | 0.022 | 0.023 | 0.029 | 0.024 | 0.022 |
|                   |              | 0.4 | 0.019 | 0.035 | 0.039 | 0.042 | 0.118 | 0.058 | 0.059 | 0.069 | 0.062 | 0.049 |
|                   |              | 1.0 | 0.050 | 0.190 | 0.192 | 0.224 | 0.491 | 0.329 | 0.323 | 0.359 | 0.302 | 0.253 |
|                   |              | 2.0 | 0.090 | 0.430 | 0.655 | 0.678 | 0.879 | 0.752 | 0.710 | 0.771 | 0.668 | 0.704 |
|                   |              | 3.0 | 0.190 | 0.520 | 0.751 | 0.782 | 0.965 | 0.864 | 0.829 | 0.874 | 0.764 | 0.809 |
|                   |              | 6.0 | 0.519 | 0.710 | 0.857 | 0.921 | 1.077 | 1.004 | 0.938 | 1.001 | 0.907 | 0.964 |
|                   |              | 11.0 | 0.728 | 0.860 | 0.924 | 1.027 | 1.135 | 1.075 | 0.995 | 1.065 | 0.971 | 1.028 |
|                   |              | 14.0 | 0.803 | 0.919 | 0.953 | 1.048 | 1.136 | 1.097 | 1.028 | 1.092 | 0.999 | 1.060 |
|                   |              | 18.0 | 1.007 | 1.008 | 0.990 | 1.084 | 1.157 | 1.123 | 1.058 | 1.107 | 1.015 | 1.088 |
|                   |              | 22.0 | 1.052 | 1.033 | 1.010 | 1.106 | 1.172 | 1.143 | 1.075 | 1.120 | 1.053 | 1.096 |

Legend: $t_c$ - crystallization temperature; $t_m$ - melting temperature.

Under all annealing conditions, the temperature of the beginning of crystallization of the amorphous alloy ($t_c$) could be observed. However, when annealing $t = 530^\circ C$, the crystallization temperature was not detected. Obviously, the above-mentioned furnace temperature was not sufficient for the formation of amorphous alloy-nanocrystals compositions. The authors [13] obtained similar results. The formation of a nanocrystalline structure in an amorphous alloy is possible only under the following conditions: 1) many crystallization centers will be formed; 2) the growth of crystallites will not be too fast. In this case, the crystallization centers appear near local inhomogeneities, and their sizes are larger than the critical one [13-15].

Based on the experimental data obtained, the dependences of the magnetic induction on the magnetic field strength were constructed (Fig. 2).
Figure 2. Curves of the dependence of the magnetic induction $B$ by the magnetic field strength $H$ with a change of annealing temperature from $t = 530^\circ\text{C}$ to $t = 620^\circ\text{C}$ (indicated near curves)

Analysis of the obtained curves (figure 2) allows us to conclude that the best annealing is $t = 570^\circ\text{C}$. After this annealing, the values of magnetic induction ($B$) are higher than after annealing at other temperatures (at the same field strength). It can also be seen that the sample that was annealed at $t = 530^\circ\text{C}$ has significantly lower values of magnetic induction ($B$) compared to other samples. Table 1 shows that there is no crystallization temperature at this annealing temperature. Apparently, the small values of magnetic induction are due to the fact that the nanocrystalline structure was not formed in this sample during annealing. In addition, it can be observed that when the temperature rises above $580^\circ\text{C}$ the magnetic properties of the sample do not improve, but deteriorate. Probably, the latter indicates that for high-quality heat treatment, it is necessary to observe the optimal temperature regime and not to overheat the parts during annealing.

After the experiments, it is safe to assume that annealing $t = 570^\circ\text{C}$ contributes to the achievement of the highest magnetic properties in an amorphous alloy of the Fe-Nb-Si-B-Cu system. This is very important for the production of magnetic amplifiers and high-frequency current transformers [16, 17].

For a more detailed study of the effect of the annealing temperature on the magnetic properties of the alloy under study, the curves of the magnetic induction dependences on the field strength are presented below: after annealing $t = 530-570^\circ\text{C}$ (figure 3) and after annealing $t = 580-620^\circ\text{C}$ (figure 4).
Figure 3. Curves of the dependence of the magnetic induction $B$ by the magnetic field strength $H$ with a change of annealing temperature: 1 - $t = 530 \, ^\circ C$; 2 - $t = 540 \, ^\circ C$; 3 - $t = 550 \, ^\circ C$; 4 - $t = 560 \, ^\circ C$; 5 - $t = 570 \, ^\circ C$

Figure 3 shows that in this temperature range (from $t = 530 \, ^\circ C \, \text{до} \, t = 570 \, ^\circ C$) there is a large spread of curves. This is due to the fact that in this temperature range, an intensive process of crystallization of the amorphous alloy occurs, accompanied by a large variation in the values of magnetic induction. However, the lowest values of induction are found in sample 1, which was annealed at $t = 530 \, ^\circ C$. As mentioned above, the latter is due to the fact, that the temperature is $530 \, ^\circ C$ is not enough for the nucleation of nanocrystals.

Figure 4. Curves of the dependence of the magnetic induction $B$ by the magnetic field strength $H$ with a change of annealing temperature: 1 - $t = 580 \, ^\circ C$; 2 - $t = 590 \, ^\circ C$; 3 - $t = 600 \, ^\circ C$; 4 - $t = 610 \, ^\circ C$; 5 - $t = 620 \, ^\circ C$
After analyzing figure 4, we can conclude that the curves have a more uniform distribution. Apparently, this is primarily due to the fact that this temperature range (from $t = 580 \, ^\circ\text{C}$ to $t = 620 \, ^\circ\text{C}$) is higher than the temperature at the beginning of crystallization (see Fig. 1). However, this annealing temperature range does not provide high values of magnetic induction.

4. Conclusion

Studies have shown that the increase in the soft magnetic properties of amorphous alloys of the Fe-Nb-Si-B-Cu system is possible by post-quenching annealing. Annealing not only removes winding and quenching stresses, but also forms a favorable structure consisting of an amorphous alloy-nanocrystal composition. It is found that after annealing at the optimal mode $t = 570 \, ^\circ\text{C}$, the alloy under study has a higher value of magnetic induction than after annealing at other temperatures (at the same field strength). The higher magnetic properties of the alloy are very important for the production of high-frequency current transformers. This study will make it possible to create more economical transformers, since it will reduce the cross-section of the magnetic circuit at the same accuracy class.

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