Effect of nanocrystallization conditions on the structure and magnetic properties of amorphous alloys

N Noskova, V Shulika and A Potapov
Institute of Metal Physics, Ural Branch of Russian Academy of Sciences, 18 S.Kovalevskay str., Ekaterinburg 620041, Russia
E-mail: noskova@imp.uran.ru

Abstract. The influence of annealing temperature, rate of cooling and magnetic field frequency under thermomagnetic treatment on the shift field value of hysteresis loop of Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ and Fe$_{60}$Co$_{20}$Si$_{5}$B$_{15}$ amorphous alloys samples has been studied. The relation of the structural state of amorphous alloys with the shift field value of the hysteresis loop has been studied. The physical explanation of this effect is proposed on the basis of obtained data.

1. Introduction
It has been known that annealing of the amorphous Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ ribbon in a direct current (DC) magnetic field leads to the shift of the hysteresis loop (SHL) along the $H$ field axis [1]. We found the shifted hysteresis loops after DC magnetic field annealing at temperatures above 250°C also in Fe$_{60}$Co$_{20}$Si$_{5}$B$_{15}$ amorphous alloy [2]. The origin of this effect is still not definitely established. The authors of [1] assumed that the SHL is originated from the precipitation of hard magnetic regions in a soft magnetic matrix. In [3] it is suggested that the SHL is a specific form of manifestation of domain structure stabilization in amorphous alloys. With a view to treat the physical mechanism of the hysteresis loop shift origination in Fe-Co-based amorphous soft magnetic alloys the research of influence of thermomagnetic treatment (TMT) conditions (heating temperature, cooling rate, magnetic field frequency) on the shift field value ($\Delta H$) has been carried out. The relation of the structural state of amorphous alloys with the shift field value of the hysteresis loop has been studied. On the basis of obtained data the methods preventing SHL while conserving high rectangularity of the hysteresis loop have been developed.

2. Experimental
The amorphous ribbons were obtained by melt quenching on a rotating disk (ribbons are 20–25 µm thick and 5 mm wide). Toroidal specimens of outer diameter 30 mm and inner diameter 25 mm were wound of these ribbons. The studies of specimens of Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ ($\lambda_{s}$=0.5·10$^{-6}$) and Fe$_{60}$Co$_{20}$Si$_{5}$B$_{15}$ ($\lambda_{s}$=30·10$^{-6}$) amorphous alloys with different magnetostriiction have been carried out. To release internal stresses the specimens were annealed in vacuum. Then the TMTs in a longitudinal magnetic field were carried out. Complex thermomagnetic treatment involved the annealing of specimens in specific temperature range with simultaneous effect of high frequency ($f$=80 kHz) and DC magnetic fields. The temperature of the TMT was determined from the minimum of the curve of the initial magnetic permeability temperature dependence [4]. Some specimens of Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ were quenched into water in an alternative current (AC) magnetic field (AC field amplitude 15 A·m$^{-1}$, frequency 50
Hz) from the Curie temperature (cooling rate was 5000°C/min). Static hysteresis loops, initial magnetic permeability $\mu_i$ and magnetic losses $P_{0.2/20000}$ at frequency 20 kHz and induction 0.2 T were measured. Losses were estimated by areas of dynamic hysteresis loops.

The study of the structure of amorphous ribbons after different TMTs was carried out using the electron transmission microscope [5].

3. Results and Discussion
The hysteresis loops of specimen of Fe$_5$Co$_{70}$Si$_{15}$B$_{10}$ alloy in different structural states: after fast quenching on a rotating disk, after annealing without a magnetic field, after TMT in the DC field and TMT in the high frequency field ($f=80$ kHz) are presented in figure 1.

![Hysteresis loops of Fe$_5$Co$_{70}$Si$_{15}$B$_{10}$ amorphous alloy after different treatments: (a)–as-quenched; (b) –annealing (325°C–1 h) without magnetic field; (c) –TMT in DC field; (d) –TMT in high frequency field.](image)

The temperature of the TMT is 325°C, the time of the TMT in high frequency field is 1 hour, DC field strength is 15 A·m$^{-1}$, AC field amplitude is 15 A·m$^{-1}$. As-cast specimen is seen to have a non-rectangular symmetrical hysteresis loop. After thermal treatment without field the hysteresis loop becomes distinctly wasp-waisted. After the TMT in the DC field the hysteresis loop becomes rectangular, but the shift of the hysteresis loop along $H$ field axis ($\Delta H$) is obtained. The TMT in a field altered with frequency 80 kHz results in the symmetrical non-rectangular hysteresis loop with the smallest coercivity. The TMT in the high frequency magnetic field is analogous to the TMT in rotating magnetic field [6]. It should be noted that the quenching into water from the Curie temperature for the specimen of Fe$_5$Co$_{70}$Si$_{15}$B$_{10}$ alloy gives the same result as the TMT in the high frequency magnetic field. The dependence of shift field $\Delta H$ on temperature of TMT for the specimens of Fe$_5$Co$_{70}$Si$_{15}$B$_{10}$ alloy is shown in figure 2. The annealing time is 1 hour for all specimens. The DC field strength is 15 A·m$^{-1}$.

One can see that the TMT up to 250°C actually does not shift hysteresis loops. Treatments in temperature interval 250–350°C result in small shift (order of 1–2 $H_c$). At higher temperatures of TMT the shift abruptly increases and reaches 10–15 $H_c$.

Analogous results are obtained for Fe$_{60}$Co$_{20}$Si$_5$B$_{15}$ amorphous alloy. The hysteresis loops of Fe$_{60}$Co$_{20}$Si$_5$B$_{15}$ amorphous alloy after fast quenching on a rotating disk, after annealing without a magnetic field, after TMT in DC magnetic field and after complex TMT are presented in figure 3.

The properties of the specimens of the alloys after different treatments are presented in table 1. The TMT temperature is 350°C, the time of the TMT in high frequency field is 1 hour, the DC field strength is 25 A·m$^{-1}$, the AC field strength is 25 A·m$^{-1}$.

It can be seen that the annealing in a DC magnetic field leads to shifted hysteresis loop, low values of initial permeability and high magnetic losses. As a result of complex TMT the hysteresis loop becomes symmetrical with a high coefficient of rectangularity ($B_r/B_m=0.95$), high initial permeability and low coercivity and magnetic losses.
Table 1. Magnetic properties of Fe\textsubscript{60}Co\textsubscript{20}Si\textsubscript{5}B\textsubscript{15} amorphous alloy after various treatments.

| Treatment          | $\mu_i$ | $H_c, \text{A} \cdot \text{m}^{-1}$ | $P_{0.2/20000}$/$\text{W} \cdot \text{kg}^{-1}$ |
|--------------------|---------|-------------------------------------|-----------------------------------------------|
| Annealing          | 1050    | 2.5                                 | 55                                            |
| TMT in DC field    | 1200    | 2.5                                 | 60                                            |
| Complex TMT        | 7500    | 1.0                                 | 7                                             |

Table 2. Magnetic properties of Fe\textsubscript{5}Co\textsubscript{70}Si\textsubscript{15}B\textsubscript{10} amorphous alloy after various treatments.

| Treatment                        | $\mu_i$ | $H_c, \text{A} \cdot \text{m}^{-1}$ | $P_{0.2/20000}$/$\text{W} \cdot \text{kg}^{-1}$ | $B_r/B_m$ |
|----------------------------------|---------|-------------------------------------|-----------------------------------------------|-----------|
| Annealing                        | 4500    | 1.3                                 | 30                                            | 0.3       |
| TMT in DC field                  | 5200    | 0.6                                 | 37                                            | 0.97      |
| Fast cooling in AC field         | 45000   | 0.4                                 | 5                                             | 0.96      |

Figure 2. Dependence of shift field on temperature of TMT in DC magnetic field for the specimens of Fe\textsubscript{5}Co\textsubscript{70}Si\textsubscript{15}B\textsubscript{10} amorphous alloy.

Figure 3. Hysteresis loops of Fe\textsubscript{60}Co\textsubscript{20}Si\textsubscript{5}B\textsubscript{15} amorphous alloy after different treatments: (a) as-quenched; (b)– annealing (350°C-1 h) without magnetic field; (c)– TMT in DC field; (d)– complex TMT.
Magnetic properties of Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ amorphous alloy after annealing without a magnetic field, after TMT in DC magnetic field, and after quenching into water from the Curie temperature in an AC magnetic field are presented in table 2. It is seen that the new method of TMT, namely, quenching into water from $T_c$ in AC magnetic field (frequency 50 Hz, AC field amplitude 15 A·m$^{-1}$) substantially improves the magnetic properties of Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ amorphous alloy.

The structure studies of Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ amorphous alloy show that after TMT in a DC magnetic field at 250°C disperse clusters of α-Co appear in amorphous specimen matrix. After such treatment the clusters are lighted in the form of sharp and non-sharp points on dark field pattern of the alloy structure; the inner ring is smeared on microdifraction pattern (see figure 4a).

After TMT at 380°C in a DC magnetic field the disperse segregations of Co$_2$Si and Fe$_3$Si phases (with size <5 nm), which are not seen after TMT at lower temperature, appear in the structure (see figure 4b). One can note some feature of these phases – arrangement of the disperse segregations in the form of chains.

Figure 4. Electron-microscopic micrograph of Fe$_{5}$Co$_{70}$Si$_{15}$B$_{10}$ amorphous alloy structure and microdifraction pattern after different TMTs: (a)–TMT in DC field at 250°C; (b)–TMT in DC field at 380°C.

The present study allows one to conclude that in amorphous soft magnetic alloys in TMT temperature range 250–350°C the small hysteresis loop shift is due mainly to the structural changes leading to the domain structure stabilization. This conclusion can be confirmed by:

- Small value of shift field;
- The destabilization of domain boundaries by quenching from the Curie temperature or TMT under the high frequency eliminated SHL;
- The reversibility of changes of magnetic properties after the treatments.

The hysteresis loop shift of the specimens annealed in DC magnetic field at higher temperatures probably occurs due to precipitations of the disperse phases with high coercive force, in this case Co$_2$Si, the direction of magnetization of which is determined by the direction of magnetic field at TMT, which leads to the large value of the shift field. On the basis of the results obtained in the present study the methods preventing the occurrence of hysteresis loop shift and conserving the high rectangularity of the loop may be recommended. They are:

- Complex thermomagnetic treatment including simultaneous action of high frequency (f=80 kHz) and DC magnetic fields for amorphous soft magnetic materials with different magnetostriction;
- Fast cooling from temperature above the Curie temperature at 5000°C/min rate in AC magnetic field (f=50 Hz) for amorphous alloys with zero magnetostriction.

The changes of magnetic properties after this treatment result from uniaxial magnetic anisotropy appearance and domain structure destabilization. The destabilization of domain walls, which is realized by the treatment in a high frequency magnetic field or fast cooling from above the Curie temperature, leads to symmetrical hysteresis loops. The uniaxial magnetic anisotropy originating due to annealing in DC or AC magnetic field results in high rectangularity of the hysteresis loop.
4. Conclusions
In the amorphous soft magnetic alloys tested the hysteresis loops shift in the temperature range of the TMT 250–350°C is probably related with domain structure stabilization. At higher temperatures the shift is due to the precipitation of fine grains with high coercivity.

The new methods of preventing SHL, namely the complex TMT and quenching into water in AC magnetic field may be recommended for improving properties of amorphous alloys.

References
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