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# Stresses Impact on Magnetic Viscosity During Initiation of Annealing for $Fe_{65}Co_9Y_6B_{20}$ Bulk Metallic Glass Below Crystallization Temperature

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The phenomenon of magnetic susceptibility disaccommodation is one of the effects related to magnetic viscosity. This magnetic delay effect is based on the decrease, over time, of the magnetic susceptibility of a sample — the sample being previously demagnetised using an alternating current with amplitude decreasing to zero. This paper presents the results of a study into the structure and disaccommodation of magnetic susceptibility for a massive amorphous ferromagnetic alloy with soft-magnetic properties. The tests were carried out on alloy samples, both in the state following production and also after an isothermal vacuum-annealing process — at a temperature below the crystallisation temperature, but above the Curie temperature.

topics: magnetic susceptibility disaccommodation, activation energy of elementary processes, bulk amorphous alloys, structural relaxation

## 1. Introduction

Phenomena affecting the properties of materials are very important for application reasons. They are especially important when they have a decisive influence on the material condition. This can apply to two types of structures for magnetic materials. Structures describing the structure of the material, such as the most common crystal structure, and structures responsible for magnetic properties of the material, i.e., the magnetic structure. Of course, it is not possible to definitively separate these structures from the material because they interact with each other. Important electrotechnical materials are magnetically soft materials, which are widely used in all electrotechnical and electronic equipment. An interesting group of materials with magnetically soft properties are amorphous and nanocrystalline FeCoB-based materials produced by rapid cooling techniques [1–5].

Initially, already in the 1990s, such materials were produced in the form of thin ribbons. The process of obtaining such materials is now very common and this technique involves rapidly cooling of a liquid alloy on copper rotating at a high linear speed cylinder [6, 7]. Thus, produced samples were liquids with a thickness of several dozen  $\mu$ m, usually 25–35  $\mu$ m, with a limit thickness close to 100  $\mu$ m. Obtaining such ribbons is associated with obtaining a very high cooling speed of 10<sup>5</sup>–10<sup>6</sup> K/s [6, 7]. It was quickly concluded that the thickness of these samples was insufficient and attempts were made to obtain amorphous materials of greater thickness. Such studies were carried out both in research laboratories of electrotechnical companies as well as in scientific units.

It was not until 1989 that Professor Inoue of the University of Tohoku and his colleagues developed criteria that made it possible to produce amorphous samples with a thickness well exceeding 100  $\mu$ m [8, 9]. Since then, a new group of amorphous materials, called the bulk amorphous materials, has appeared in science. They concluded that at least three components should be used to obtain a bulk amorphous sample, the atomic rays of which will differ by more than 12% atomically and in addition pairs of at least the main components of the alloy will show a negative mixing heat [10]. Properties of conventional alloys (amorphous ribbons) and bulk alloys are slightly different, which is related to the stresses caused by the manufacturing process itself. As is well known, amorphous alloys are metastable materials with interesting properties including reversible and irreversible relaxations that can even occur at room temperature.

The amorphous structure itself is not quite understood to accurately characterize it as it is in the case with crystalline materials. In crystalline alloys, structural relaxation occurs in a narrow temperature range and the corresponding activation energies have discrete values. In the case of amorphous alloys, these relaxations occur over a wide temperature range and correspond to a wide spectrum of activation energy. One of the more interesting research areas in the field of magnetization of amorphous materials in low magnetic fields, where reversible relaxation processes can be studied, is the study of the effect of magnetic delays, called magnetic susceptibility disaccommodation [11–13]. This phenomenon consists in reducing the susceptibility of a sample that has previously been demagnetized by alternating current with an amplitude decreasing to zero (Fig. 1).

This phenomenon in the case of amorphous materials is much more complex than for crystalline materials. The model relating to amorphous materials was developed by H. Kronmuller [14], who stated that in amorphous alloys the considered phenomenon is related to the ordering of pairs of atoms near free volumes. Free volume in amorphous materials plays a similar role as point defects in crystalline materials. In his considerations, Kronmuller used the model of two-level systems developed earlier by Anderson to fully describe the phenomenon of disaccommodation [15]. Due to the very nature of amorphous materials, it should be concluded that there are many two-level systems with different energies in their volume. Change in the position of atoms is well described by the model of rigid spheres moving close to free volumes (see Fig. 2).

It should be added that the jump of a single atom, as in Fig. 2, is possible only if the displacement is accompanied by movements of other atoms similar in position. A magnetic delay itself resulting in a reduction in magnetic susceptibility can be illustrated by a diagram showing the deepening of the potential well in which the domain wall is located (see Fig. 3).



Fig. 1. Magnetic susceptibility desacommodation curve  $\chi_0$ -susceptibility to t = 0,  $\chi_\infty$ -susceptibility at the time of  $t \to \infty$  [13].



Fig. 2. Rigid sphere model showing two axis orientations of atom pairs corresponding to two energy levels [16].



Fig. 3. Domain wall stabilization potential creation scheme [16].

A change in the properties of the amorphous material can be achieved by relaxing it at temperatures below the crystallization temperature and above the Curie temperature. This work presents the results of a structure test and the disaccommodation of magnetic susceptibility for the massive amorphous alloy of the alloy in the state after solidification and after the heating process. The paper presents the results of structure research and magnetic susceptibility disaccommodation for a bulk amorphous alloy in solidified state and after the annealing process.

# 2. Materials and methods

Using the radiation cooling method from high purity ingredients (min. 99.98%),  $Fe_{65}Co_9Y_6B_{20}$  amorphous samples were produced. The produced samples had the shape of plates with 0.5 mm thickness and had an area of 100 mm<sup>2</sup>.

Initially, crystalline ingots were melted from alloy components weighed in the right proportions. Ingots were made in an arc furnace and the process of melting and mixing alloy components was done in the atmosphere of argon. The components of the alloy have been melted several times to obtain a good degree of mixing. Then, the ingots — mechanically cleaned from impurities — were divided into smaller portions, which were used to produce rapidly-cooled plates. The plates were made using the method of injecting liquid alloy into a water-cooled copper mold. Magnetic properties were conducted using an automated system to measure susceptibility and its disaccommodation. These measurements were made using the transformer method in a magnetic field with a frequency of 2 kHz and an amplitude of 0.16 A/m. Each measurement was initiated by earlier demagnetization of the sample with a variable magnetic field with a frequency of 100 Hz and an amplitude decreasing from 1500 A/m to 0in 1.1 s. The samples were heated in a vacuum pipe furnace at 650 K and for 30 min.

#### 3. Results

Figure 4 shows the curves of initial magnetic susceptibility to temperature for the tested alloy after solidification and after the annealing process.



Fig. 4. Initial magnetic susceptibility as a function of temperature for the  $Fe_{65}Co_9Y_6B_{20}$  alloy after solidification (a) and after the annealing process (b).



Fig. 5. Isochronous diacommodation curves measured for test samples: alloy after the solidification (a) and after the annealing process (b).

The initial magnetic susceptibility to a temperature of about 550 K increases for both tested samples. In the vicinity of temperature 650 K, the initial compliance drops sharply.

This decrease is related to the transition of the sample from the ferromagnetic to paramagnetic state. It is clearly visible that the susceptibility increases after thermal treatment, which means that irreversible structural relaxations took place in the sample volume. Figure 5 presents the measurements of time stability of magnetic susceptibility.

Curves in Fig. 5 are as typical as for amorphous materials. They consist of a wide maximum. As a result of heat treatment, free volumes were released into the sample surface and the number of the so-called relaxors was reduced, which is manifested by a decrease in the intensity of disaccommodation.

#### 4. Conclusions

This work presents the results of studies of initial magnetic susceptibility and its disaccommodation. Samples of alloy  $Fe_{65}Co_9Y_6B_{20}$  were tested in a state after solidification and after the annealing process carried out at temperatures below the crystallization temperature and above the Curie temperature. Such a conducted annealing process does not affect the domain structure. It has been observed that a sharp decrease in magnetic susceptibility is associated with the transition of the magnetic material from the ferromagnetic to paramagnetic state. The tested samples, both in the solidified state and after the annealing process, have good time and temperature stability of magnetic properties. If the amount of free volumes within the alloy decreases, its density increases, i.e., the packing coefficient of atoms in the volume unit increases. Therefore, after the isothermal heating process below the crystallization temperature and above the Curie temperature, the intensity of magnetic susceptibility disaccommodation is reduced.

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