Magnetic Properties of Composites Based on Amorphous Iron Alloys Produced with the Use of a Non-Magnetic Binder and Covered with High Temperature Varnish

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Abstract. The continuous increase in the demand for electricity causes the need to search for new energy sources and to look for savings during its transmission. One of the areas of interest are materials dedicated to low loss transformer cores. Due to good magnetic properties, amorphous and nanocrystalline iron-based alloys are used in this area. The limited dimensions of these materials make their application difficult. Magnetic composites based on ferromagnetic amorphous alloys are a new direction of research. Usually, composites are made using a non-magnetic binder. However, the separation of magnetic particles worsens the magnetic properties. The solution to this problem may be production of composites coated with high temperature varnish. As part of the work, the results of research on the structure and magnetic properties of composites based on amorphous iron alloys are presented. Composites were made with 0.5% resin by mass and without resin covered with high temperature varnish. Material produced without the use of resin had a lower coercive field value.

1. Introduction
The rapid economic development that has been taking place for several decades has brought a very large increase in the demand for electricity. Unfortunately, despite the development of technologies in the field of obtaining and processing energy resources, the production of electricity is becoming more and more expensive. The development of renewable energy sources alone does not cover the growing demand for energy. The development of energy technologies must be supported by progress in energy transmission and processing. One of the aspects many scientists are working on is the losses caused by transformers. Modern magnetic materials are used for their construction. Commonly used materials for transformer cores are Fe-Si sheets [1-2] and amorphous alloys based on Fe or Co [3-7]. Another object of research are materials intended for wedges in nurseries of electric motors or magnetic screens [8]. In recent years, rapid-quenched alloys with an amorphous structure and nanocrystalline alloys created by appropriate thermal treatment have enjoyed special interest [9-12]. Depending on the alloy composition used and the type of structure obtained, these materials are characterized by soft magnetic properties such as low coercive field value and high saturation magnetization [13-16]. Materials of this type are produced on an industrial scale by the melt spinning method (casting a liquid alloy on a copper rotating drum in the form of ribbons) [17]. Another way to make amorphous material is to use a method of injected a liquid alloy into a copper mold. The method is characterized by a much lower
cooling rate but allows the production of alloys with amorphous structure with much larger dimensions [18].

An important disadvantage of rapid-quenched alloys is their limited geometry. One of the possible solutions to this problem are magnetic composites. Composite is a material consisting of at least two physically and chemically separate phases, which give the material new properties. In the case of magnetic composites, these phases are ferromagnetic powder obtained during crushing of an alloy characterized by the desired magnetic properties and a type of bonding binder giving forming properties, for example epoxy resins [19]. The addition of resin negatively affects the magnetic properties but significantly extends the application possibilities of the material. The challenge is to find optimal solutions during the production of composites so as to obtain the appropriate mechanical properties enabling the formation of desired shapes while maintaining good magnetic properties. Studies show that the ratio of resin to powder and the type of powder (particle size and shape) have an impact on the properties of magnetic composites [19].

However, the fact of magnetic particle separation limits the occurrence of exchange interactions, which significantly affects the deterioration of magnetic properties, increase the value of the coercive field and decrease the saturation magnetization [19-21]. A way to reduce this negative effect is to create a resin-free composite, ensuring the consistency of the material with a high-temperature varnish layer. In this way, an electrical insulator material can be obtained while maintaining good magnetic properties. The differences between the resin-containing composite and the high-temperature varnish are illustrated in the diagram in Figure 1.

![Figure 1. Scheme of composites made with resin and high temperature varnish.](image)

As part of this work, magnetic composites based on Fe$_{70}$Nb$_5$Y$_5$B$_{20}$ amorphous alloy were produced. Composites were made with and without resin - these materials were covered with high-temperature varnish. The purpose of the work was to compare the magnetic properties of composites made according to two different methods.

2. Methods and materials
The starting material for making the magnetic composite is an alloy with the chemical composition Fe$_{70}$Nb$_5$Y$_5$B$_{20}$. To produce the alloy, components with a purity of 99.99% were used. The alloy components were measured using a laboratory balance with an accuracy of 0.001 gram. The ingredients were melted in an arc furnace to combine and mix them. The process takes place in an argon protective atmosphere after pumping out the furnace working chamber. In order to obtain the most even distribution of alloy components in the ingot volume, the melting is repeated several times. The ingot produced in this way was subjected to mechanical cleaning and using an ultrasonic cleaner. On the basis of a polycrystalline ingot, a rapid-quenched alloy was made by injection molding. The process was carried out in an argon atmosphere. The charge was melted by means of eddy currents.
The liquid alloy was injected into a water-cooled copper mold. The cooling rate achieved by this method is up to $10^3$ K/s. The alloy obtained in the form of 0.5 mm thick plates was ground. The ferromagnetic powder was divided into fractions (20-50 μm, 50-100 μm, 100-200 μm) using a set of laboratory sieves with shaker. Based on the selected powder fraction, two types of magnetic composites were produced: with 0.5% resin content (by mass) and without the addition of resin with a high-temperature varnish coating. Composites were made in the form of toroids with an outer diameter of 10mm, an inner diameter of 5mm and a thickness of 2mm. The sample production scheme is shown in Figure 2.

**Figure 2.** Scheme for producing magnetic composites.

Composites were made using a hydraulic press. The powder was pressed at 200 bar for 30 minutes at room temperature. The molding without the addition of resin during removal from the mold was covered with a layer of high temperature varnish. The resulting composites were allowed to dry at room temperature. Figure 3 contains photos of produced materials.
Figure 3. Photographs of produced materials: (a) polycrystalline ingot, (b) amorphous alloy in solid state, (c) composite with 0.5% resin content, (d) composite coated with high temperature varnish.

The structure of the materials produced was examined using an BRUCKER model Advanced 8 X-ray diffractometer. X-ray irradiation was performed in an angle of 2θ with an exposure time of 5 s and a measuring step of 0.02°. Magnetic properties were tested using a LakeShore vibrating magnetometer to obtain static hysteresis loops. The measurement was carried out in the range of external magnetic field strength up to 2T.

Using the relationship (1), the spin wave stiffness parameter parameter $D_{spf}$ was calculated (based on the primary magnetization curve).

\[
b = 3.54gμ_0μ_B\left(\frac{1}{4πD_{spf}}\right)^{3/2}k(Tgμ_B)^{1/2}
\]  

(1)

Where:

- $b$ – slope of the linear fit corresponding to the thermally induced suppression of spin waves by a magnetic field of high intensity,
- $μ_0$ – magnetic permeability of vacuum,
- $k$ – Boltzman's constant,
- $μ_B$ – Bohr magneton,
- $g$ – gyromagnetic factor,
- $T$ – temperature.

3. Research results

Figure 4 contains X-ray diffraction patterns for the materials tested.

Figure 4. X-ray diffraction patterns for: (a) Fe$_{70}$Nb$_5$Y$_5$B$_{20}$ alloy after solidification, (b) 20-50 μm powder fraction, (c) 50-100 μm powder fraction, (d) 100-200 μm powder fraction, (e) composite with resin, (f) composite coated with high temperature varnish.

Testing of the alloy after solidification was carried out for the volume of the material, i.e. for the alloy in the powder state. Diffractograms for composites were measured only for the surface of the materials. The recorded diffractograms are typical as for materials with an amorphous structure. X-ray
diffraction images show only one maximum in the 40-50° two theta angle range. Figure 5 contains static magnetic hysteresis loops for the materials tested.

Figure 5. Static magnetic hysteresis loops for: (a) Fe⁷⁰Nb⁵Y⁵B²⁰ alloy in solidified state; powder fractions: (b) 20-50 μm, (c) 50-100 μm, (d) 100-200 μm; (e) composite with 0.5% resin for 50-100 μm fraction, f) composite for 50-100 μm fraction covered with high temperature varnish.

Defining the structure significantly affected the magnetic properties of the tested materials. The static magnetic hysteresis loop measured for a solid alloy has an almost rectangular pattern. This curve shape is typical for materials with so-called soft magnetic properties. Measurements carried out for various fractions of amorphous powder indicate a significant impediment to the magnetization process. The tested samples achieve saturation value at a much higher intensity of the external magnetic field. Amorphous powder with grain size 50-100 μm has the best properties among the three fractions isolated. On the basis of this fraction, composites with resin or high-temperature varnish were made. The registered loops are characterized by a similar course to the loops measured for the separated fractions. After increasing the origin of the M-H coordinate system, the coercive field value was determined. In addition, the value of saturation magnetization and the value of the external magnetic field at which the magnetization sample was associated with the damping of thermally excited spin waves was determined.

The results of magnetic properties are shown in Table 1.

Table 1. Magnetic properties of produced materials.

|                  | Hc [A/m] | Ms [T] | HcIP [T] | Dspf [meV/nm²] |
|------------------|----------|--------|----------|----------------|
| ASQ              | 10       | 0.75   | 0.35     | 42.9           |
| 20-50 μm         | 565      | 0.68   | 0.7      | 35.4           |
| 50-100 μm        | 31       | 0.73   | 0.7      | 35.5           |
| 100-200 μm       | 116      | 0.73   | 0.74     | 35.2           |
| composite with resin | 780   | 0.62   | 0.7      | 42.1           |
| composite with varnish | 690   | 0.61   | 0.67     | 43.3           |
As expected, the 20-50 μm powder has the worst magnetic properties due to the higher degree of magnetic particle separation. This fact weakens the exchange interactions. In this case, the magnetic properties are created to a greater extent by dipole interactions than exchange interactions. The composites produced have a lower saturation magnetization value, which is associated with a much lower material density. The value of the coercive field for composites increased more than 20 times compared to the value measured for the powder itself. Composite coated with high-temperature varnish is more easily re-magnetized compared to a molded with resin. This means that a smaller degree of separation of magnetic particles improves the magnetic interactions that occur in the volume of this material. In higher magnetic fields, the process of magnetizing powdered samples is similar. The tested materials achieve magnetic saturation at an external magnetic field strength of about 0.7T.

4. Conclusions
As part of this work, the impact of manufacturing technology on the soft magnetic properties of composites based on bulk amorphous alloys was investigated. The alloy was tested after solidification and after division into fractions: 20-50 μm, 50-100 μm and 100-200 μm. The 50-100 μm fraction shows the best soft magnetic properties slightly different from the values measured for the solid alloy. The composites produced had a similar saturation magnetization value of 0.61-0.62T and a much higher coercive field value compared to the solid alloy and 50-100 μm powder fraction. In a composite coated with high-temperature varnish, the arrangement and distances between magnetic particles are more favorable than for a composite with resin, which results in a lower coercive field value of almost 100 A/m. In addition, the composite coated with high-temperature varnish is characterized by a significantly higher applicability due to the lack of brittleness (which ensures the mechanical properties of the varnish) and better insulation properties (due to the "closure" of magnetic particles in the varnish matrix).

5. References
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