Physical properties of magnetostrictive composite materials with the polyurethane matrix

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ABSTRACT

Purpose: The purpose of this study was to determine the thermal and electrical conductivity of composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with different particle size distributions and varying volume concentration.

Design/methodology/approach: The investigated samples were obtained by casting of the composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles. There were determined the samples density, electrical properties (by a resistivity measurements), thermal conductivity (by Physical Property Measurement System with thermal transport option), as well as the metallographic investigations (by stereo microscope).

Findings: It was found from obtained results that the resistivity value for composite materials filled with larger particle size $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ was lower than the smaller particles size filled composites. Moreover, it may be noticed that thermal conductivity has an approximate value for different $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particle size and the same its volume fraction in matrix. Simultaneously it was also observed that the thermal conductivity of the composite materials did not depend on the temperature within the tested range from 293 to 333 K.

Research limitations/implications: Contributes to research on structure and physical properties of magnetostrictive composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles.

Practical implications: The polyurethane matrix in investigated composite materials causes growth of resistivity, limiting this way losses for eddy currents at the high operating frequency of the transducers.

Originality/value: The obtained results show the possibility of manufacturing the magnetostrictive composite materials based on the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles, with desired physical properties (including thermal and electrical one) in cost effective way in comparison to conventional giant magnetostrictive materials (GMM).

Keywords: Composites; Smart materials; Terfenol-D; Giant magnetostrictive materials; Physical properties

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PROPERTIES

1. Introduction

The dynamic engineering and technology development rate and constant search for new technological solutions also refer to the materials of special magnetic properties. Moreover, the interest in the magnetostrictive materials permanently grows, due to the continuously expanding opportunities of their practical application by industry. The factors restricting the application of Giant Magnetostriction Materials (GMM) at an industrial scale motivate the continuous search for alternative solutions, so that the requirements given to such materials by state-of-the-art technology are met, including [1-3]:

- low correlation between the physical properties and the temperature;
- high Curie temperature value (enabling work within wide temperature ranges);
- plasticity enabling the production of thin layers (which is necessary for reduction of energy losses caused by eddy currents);
- high corrosion resistivity;
- simple production technologies;
- low price.

The advantages resulting from dispergation of the magnetostrictive particles in polymer matrix mainly are in the broader frequency band of the transducers' operation, lesser fragility, adaptability of the properties to specific applications and competitive price of the composite materials, in comparison to the monolith magnetostrictive alloys. The target of the scientists throughout the last 30 years (mainly in the USA and in East Asia for the last decade) [4-7] is to produce composite materials of the highest magnetostriction possible with the simultaneous reduction of the participation of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ fractions in the matrix [8-12].

The use of components is determined by their mechanical, strength and physical properties, which in turn - in case of composite materials - depend on the properties of the components, i.e. volumetric share of the reinforcement material, its form and distribution method in the matrix. For example the significant difference in the thermal dilatation and conductivity coefficients of the components leads to the occurrence of thermal stresses in the composites and such stresses may exceed the matrix level of strength [13-29].

Proper selection of chemical constitution and technology allow to fabricate the magnetostrictive composite materials with exact utilitarian features, which magnetic, electric, and mechanical properties depend on the shape and size of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles, as well as the way of connecting the reinforcement with the matrix [9, 12, 19]. Moreover - from practical point of view - it is important to gain the biggest magnetostriction possible with the lowest magnetic field intensity in order to provide the most effective energy converting.

Integration of components with opposite properties in magnetostrictive composite materials and diversity of factors influencing on their magneto-mechanical parameters makes the problem of proper selection of matrix material and $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ volume fraction more complex. Taking into consideration factors mentioned above it is recognized to be expedient to take up a research concerning elaborating new group of magnetostrictive composite materials in the way that makes it possible to gain high magnetostriction, simultaneously lowering material costs and

increasing resistivity (lowering occurrence of losses caused by eddy currents), as well as decreasing thermal conductivity.

Nowadays, the magnetostrictive composite materials are subject of many different research consideration [1-12,30], but at the same time no information can be found concerning the effect of particles content on physical properties in $Tb_{0.3}Dy_{0.7}Fe_{1.9}$/polyurethane composite materials.

Previously [30], the authors performed studies on composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder particles and varying volume concentration, including magnetic and magneto-mechanical examinations. This paper is an attempt to complete data base with physical properties for these materials.

2. Experimental

2.1. Material

Examinations were made on composite materials reinforced with Terfenol-D (i.e. $Tb_{0.3}Dy_{0.7}Fe_{1.9}$) powder (Etrema Co, USA), demonstrating the giant magnetostriction. The density of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ is 9.25 g/cm^3 and its thermal conductivity equals to 13.5 W/m·K. The matrix material was a low-viscosity two-component polyurethane resin Smooth-cast 325 (Smooth-on Inc., USA) with a density of 1.07 g/cm^3. Samples were prepared by casting and the mixture was stirred until the matrix gelation was completed to prevent the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles sedimentation. After cross-linking process finished, specimens were placed in an oven at 338 K for 5 h to ensure full cure of the resin. The samples differ with the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ concentration in matrix (Table 1) and with the powders particle sizes, denoted by its manufacturer as 38-106 µm and 212-300 µm were made.

Table 1.
Concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles in composite materials

Volume fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$, %	Mass fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$, %
10	48.7±0.5
15	60.1±0.5
20	68.1±0.5

2.2. Methodology

Analysis of the grain distribution of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles was made in [30] and according to obtained results it had been noticed that powder with granulation denoted by manufacturer as 212-300 µm has grains whose median is 96.19 µm, whereas for powder denoted as 38-106 µm this value equals to 39.86 µm.

In this work the metallographic, density, electrical and thermal properties investigations were carried out on composite materials.

The metallographic investigation of composite materials was made on SteREO Discovery.V12 ZEISS microscope at the magnification 25x.

The density of the composite materials was evaluated by determining the test piece mass using an analytical balance with an accuracy of $\pm10^{-4}$ g and its volume basing on the apparent mass loss by immersing in water. Five measurements were made for each composite version.

Electrical properties testing were made on stand consisting of the stabilized DC power unit, slide rheostat, ammeter, voltmeter, and sample holder (Fig. 1). To ensure precise contact of the sample surface with electrodes, and the homogeneous distribution of electrical charge, as well as to minimize the effect of load of electrodes on the measured current value, samples were prepared for testing with 3 mm thick copper disks mounted at the ends (Fig. 2). By using Ohm principle and on the base on the following relation, the resistivity values were estimated:

$$\rho = \frac{U}{I} \cdot \frac{A}{l}$$

(1)

where ρ is the resistivity ($\Omega \cdot$m), l is the length of the sample (m), A is the cross-section area of transverse section of the rod (m^2), U is the voltage (V), and I is the current intensity (A).
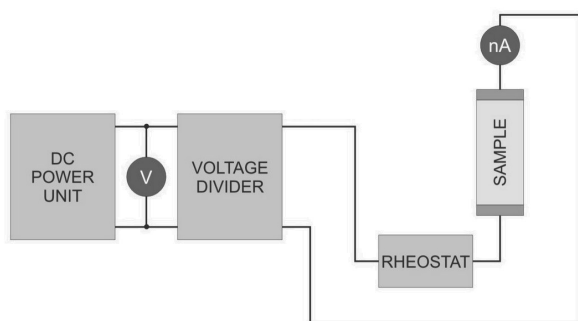

Fig. 1. The test stand for resistivity measurements (nA - the nano-ammeter, V - the voltmeter)

Fig. 2. The view of the sample ready to electrical properties investigation

The thermal conductivity investigation was performed using the thermal transport option of the QUANTUM DESIGN Physical Property Measurement System located at the Institute of Physics of the Polish Academy of Sciences in Warsaw, Poland. In this system, a two-contact method is applied (Fig. 3) to determine the thermal conductivity of composite samples rectangular in shape with dimensions of 8×2×2 mm, over the temperature range from 293 to 333 K and for magnetic induction (B) equals to 1 T.
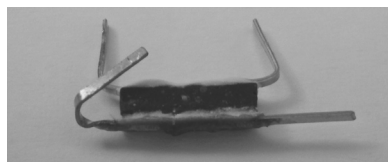

Fig. 3. The view of the sample prepared to thermal conductivity measurements

3. Results and discussion

The magnetostrictive composite materials with the polyurethane matrix were reinforced with the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ magnetostrictive particles. The homogenization grade of the particles in the matrix has been determined basing on the materialographic images made with a stereo microscope (Fig. 4). The observations enabled finding the even distribution of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles in the polyurethane matrix with no voids in the structure.

a)

b)
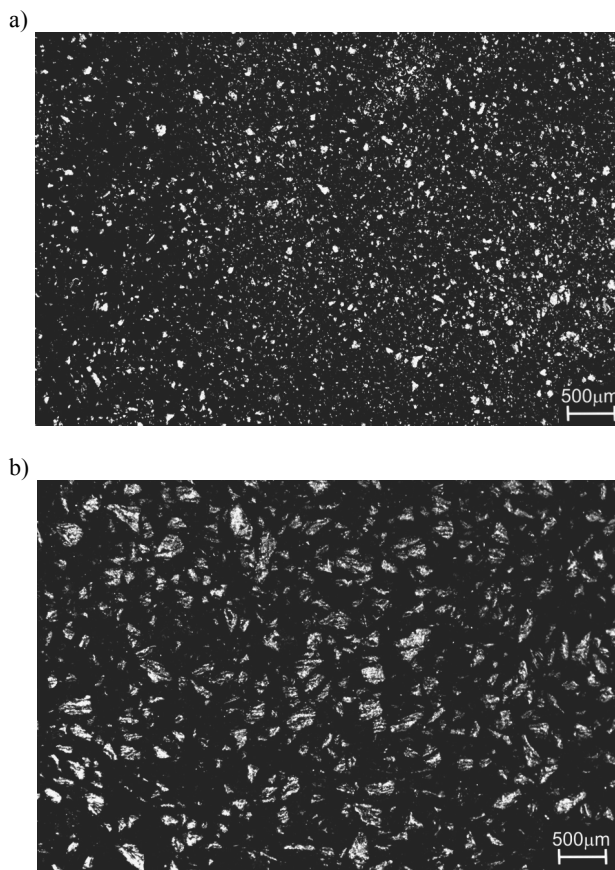

Fig. 4. Topography of the surface of the composite materials with the polyurethane matrix reinforced with 20% volume fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with median of 39.86 μm (a) and 96.19 μm (b)

Based on the density measurement results (Table 2, Fig. 5) it has been found that - disregarding the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particle size, being the reinforcement - the density of the composites increases monotonically along with the increasing share of the reinforcement material from 1.79 g/cm^3 (for materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles and 39.86 μm median and 10% volumetric share) to 2.62 g/cm^3 (for materials reinforced with the same particles and 20% its volumetric share), assuming values below the theoretical density determined according to the rule of mixtures.

Table 2.
Density of the composite materials

Median of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles, μm	$Tb_{0.3}Dy_{0.7}Fe_{1.9}$ volume fraction in matrix	Theoretical density, g/cm³	Density of the composite materials, g/cm³
39.86	10%	1.89	1.79±0.12
	15%	2.29	2.18±0.04
	20%	2.70	2.62±0.08
96.19	10%	1.89	1.81±0.04
	15%	2.29	2.15±0.06
	20%	2.70	2.55±0.06

The composite materials density decreasing along with the increasing volumetric share of the matrix material results from significantly lower value of density for polyurethane as compared $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ used as reinforcement. For the composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with 96.19 μm median, the density value oscillates within the range from 1.81 g/cm³ (for materials with 10% volumetric share) to 2.55 g/cm³ (for materials with 15% share of reinforcement in the matrix).
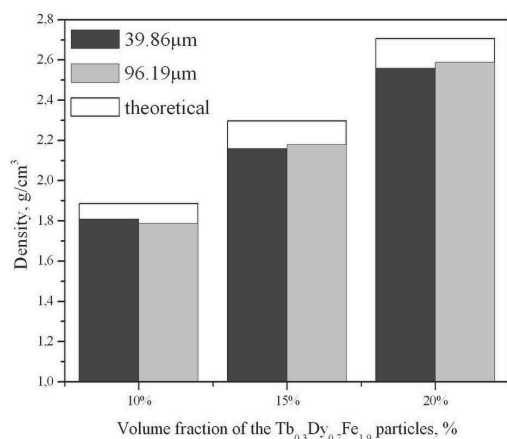

Fig. 5. Influence of the content of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles on the density of the composite materials

The resistivity/voltage diagrams for composite materials with polyurethane matrix, reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles have been presented in the Figs. 6a and 6b . The composite materials tested are characterised with high resistivity depending on the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles volumetric ratio in the matrix and the size of such particles. The resistivity of the newly developed materials with U = 200 V voltage fits within the range from 1 kΩ·m (for composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder with particle size 212-300 μm and volumetric ratio of 20%) to 30.5 kΩ·m (for 38-106 μm size $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ and volumetric ratio 10% (Fig. 7)). Based on the recorded current-voltage characteristics it has been found that their course is typical for dielectric materials, and - for the analogous nature the changes of voltage in function of current intensity reasons - there has been presented only the curve for composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles of 39.86 μm median and volumetric share 10% (Fig. 8).
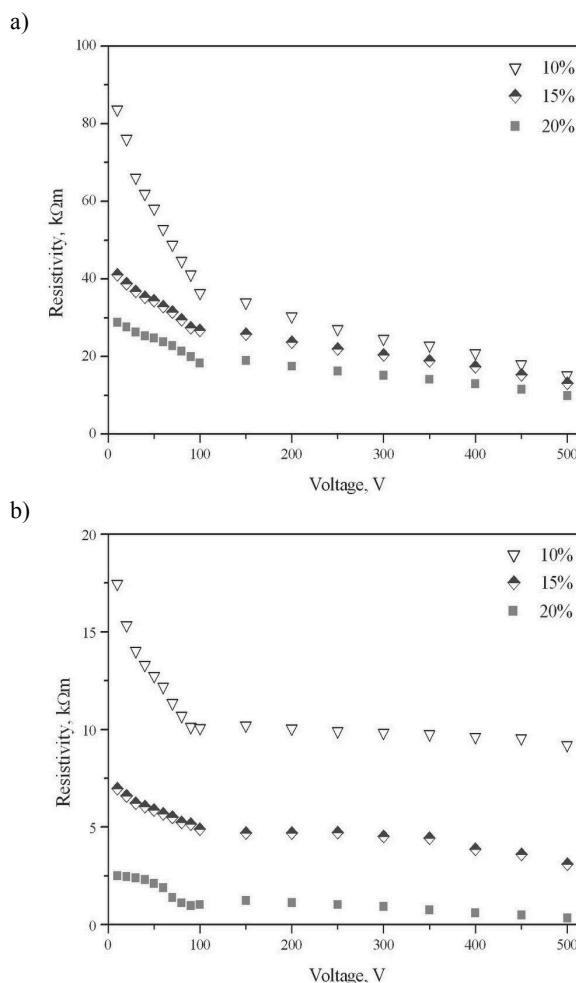
a)

b)

Fig. 6. The voltage dependence of resistivity for composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with median of 39.86 μm (a) and 96.19 μm (b)
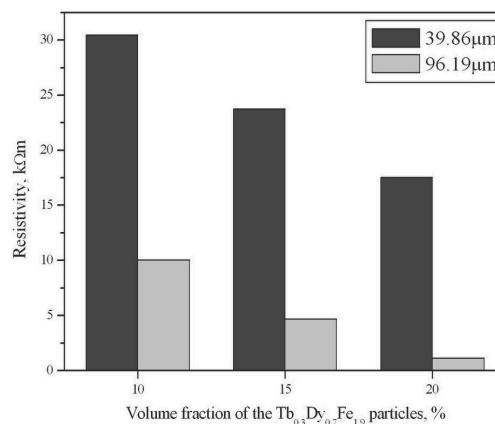

Fig. 7. Relation between the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles volume fraction in the composite materials and the resistivity (for U = 200 V)
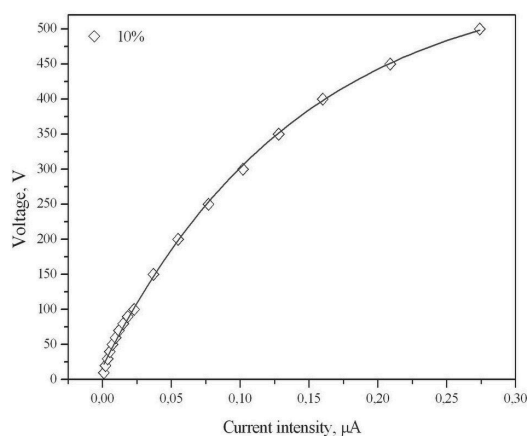
Fig. 8. Current - voltage characteristics for composite material with polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with 39.86 μm median and 10% volumetric share

a)

b)

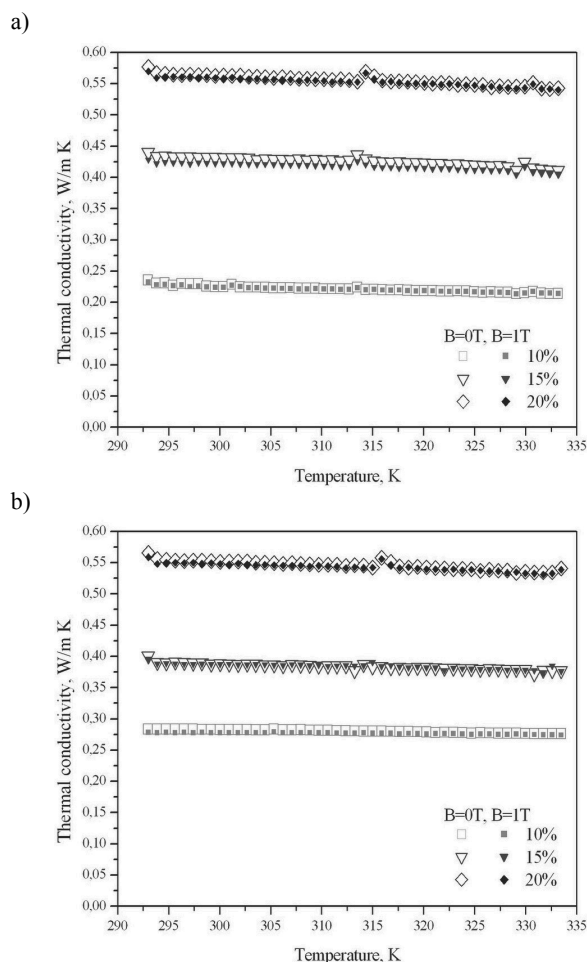
Fig. 9. The variation on thermal conductivity of composite materials with polyurethane matrix, reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles of median equal to 39.86 μm (a) and 96.19 μm (b) versus temperature

Figs. 9a and 9b illustrates the diagrams of thermal conductivity dependency on the temperature for composite materials with polyurethane matrix, reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles, obtained in magnetic field induction B = 1 T and in absence of the same. The thermal conductivity of composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles (Fig. 10) is within the range from 0.221 W/m·K (for materials with particles of 39.86 μm median and 10% reinforcement share), increasing its value monotonically, along with the increase of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles share in the matrix to the value of 0.556 W/m·K (for materials with particles of the same median and 20% share of particles). This is an obvious dependency, considering the value of thermal conductivity for monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ equal to 13.5 W/m·K.

It was found that the lowest thermal conductivity value – equal to 0.221 W/m·K – is presented by composite materials with polyurethane matrix, reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with 39.86 μm median and 10% volumetric share, however an approximate value of 0.280 W/m·K has been obtained for samples with the same volumetric share of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ reinforced with particles with 96.19 μm median. It was also observed that the thermal conductivity of the composite materials did not depend on the temperature within the tested range from 293 to 333 K.

The magnetic field set during the thermal conductivity measurements of the newly developed composite materials with 1 T induction has no impact (in case of composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles of 39.86 μm and 96.19 μm medians and volumetric share: 10% and 15%, respectively) or slightly reduces (for other materials) the thermal conductivity of the materials tested. The anomaly visible in the diagrams for temperature about 314 K (most conspicuous in the samples of higher thermal conductivity values) may be related to the physical effect that was not analyzed in details in this paper.
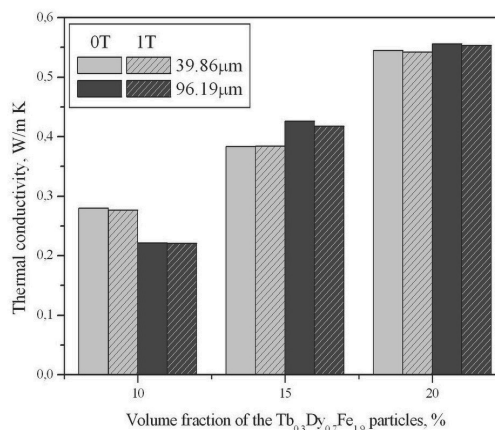

Fig. 10. The concentration of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles dependence of the mean thermal conductivity of the composite materials with the polyurethane matrix

4. Conclusions

In this work the density, electrical properties, thermal conductivity, as well as the metallographic investigations of the

composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles were determined. The study indicates that – as a result of reinforcing composite materials with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles of gradually increasing median (according to the statistical distribution of their particle sizes under analysis) - slight changes of the composite materials density were found (ranging from 1.79 to 1.81 g/cm^3 for the composite materials with 10% volumetric share of the reinforcement) more and more noticeable, along with the increasing volumetric share of reinforcement in the matrix (Fig. 5). The dispergation of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles in the polymer matrix obviously causes reduction of density and increased resistivity (Fig. 7) of the composites, in comparison to the monolithic alloy.

Based on the recorded current-voltage characteristics (Fig. 8) it has been found that the expected improvement of newly developed composite materials electrical properties making them an attractive alternative for the monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ alloy (with resistivity value equals to 58·10^{-8} Ω·m), which is of particular importance by taking into account that the resistivity values correspond to the losses on eddy currents in materials with low values of resistivity.

With the decreasing ratio of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles in the composite materials, the weak contact between the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles causes high resistivity, while for materials with 20% volumetric ratio, the resistivity decreases due to the conducting properties of the reinforcement material. The lowest resistivity value of 1 kΩ·m (for U = 200 V voltage) recorded for composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles of 96.19 μm median and 20% volumetric share is several times higher than for monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ - which, however, should be considered in the context of dielectric losses related under the operating conditions of such materials, causing heating of the polymer material [8, 19].

The thermal conductivity values of the composites developed range from 0.221 W/m·K (for material reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with 39.86 μm median and 10% volumetric share) to 0.556 W/m·K (for the composite material reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with the same median and 20% volumetric share of the reinforcement in the matrix) and – like the density values increase monotonically along with the increasing $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ share in the matrix. Such changes result from higher density (9.25 g/cm^3) and thermal resistivity (13.5 W/m·K) of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ as compared to the values for polyurethane resin (being 1.07 g/cm^3 and 0.035 W/m·K, respectively). The independent values of thermal conductivity of the temperature and 1 T induction magnetic field set during the measurements (Figs. 9 and 10), prove that the parameters are insignificant in relation to the operating properties of the composite materials developed. So the heat generated during the operation of the materials in rapidly varying frequencies will hardly affect their thermal properties.

Moreover, with 20% volumetric share of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles in the matrix, the metallic particles are in mutual touch, which affects the increase of electrical conductivity of the composites. The lack of percolation paths for the composite materials with 10% volumetric share of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ in the matrix causes that the properties – from the position of the operation of materials developed in rapidly varying magnetic fields – are worse. In contrast to electrical properties, the concentration dependence of thermal conductivity does not show the percolation behavior.

At the subsequent stage of research, it is planned to carry out measurements of resistivity in the function of temperature, which seems necessary, due to the fact that, according to [14, 23], slight temperature increases may cause the drop of the polymer resistivity by more than two orders, while in metals the resistivity in the same temperature range remains unchanged or minimally grows.

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