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Magnetic properties of magnetostrictive $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ / polyurethane composite materials

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ABSTRACT

Purpose: The aim of this work was to obtain polyurethane matrix composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles and to observe changes of magnetic properties and magnetostriction of samples with different particle size distributions of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder and varying volume concentration.

Design/methodology/approach: The studies was performed on composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders. The morphology of powder was examined by scanning electron microscopy (SEM) and analysis of the grain distribution of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder was made using the Mastersizer 2000 analyser. The changes of magnetostriction and magnetic properties, including hysteresis loops and effective permeability with changing applied field are tested.

Findings: Analysis establishes a direct connection between magnetic properties and structural characteristics of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder size, which was used as reinforcement phase. The increase of particle size distribution of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder in composite materials amplify the magnetostrictive responses, moreover the change of magnetic properties as a function of volume fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder were confirmed.

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

Practical implications: The polymer matrix in investigated composite materials causes growth of resistivity, limiting this way losses for eddy currents at the high operating frequency of the transducers. In addition the values of permeability of composite materials is nearly constant in investigated frequency range.

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 properties in cost effective way in comparison to conventional giant magnetostrictive materials (GMM).

Keywords: Composites; Multifunctional and smart materials; Magnetic properties

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MATERIALS

1. Introduction

Intensive interest of researchers all over the world in the so called smart materials may be observed for the last few dozen years. Magnetostrictive materials deserve special interest in this group, including the Giant Magnetostrictive Materials (GMM), which are the subject of the intensive investigations in the leading laboratories in the world [1, 2] because of their interesting magnetic properties and possibility of the eventual applications in the actuator/sensor converters. These materials use the physical phenomena thanks to which they can convert the magnetic energy to the mechanical one (magnetostriction) and have the capability to reverse this process, i.e., conversion of the mechanical energy to the magnetic one (phenomenon opposite to magnetostriction).

The $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ alloy is the most commonly used material of this group, characteristic of the giant magnetostriction (up to 1600 ppm depending on fabrication method), very short reaction time and capability to carry the loads of the order of 600 MPa.

In spite of their many advantages, some properties of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ restrict possibilities of its applications. Among others, losses during material use in the high frequency magnetic fields, high material costs, and high brittleness of the material are counted to them [3-5].

Introducing a magnetically neutral polymer material as the matrix of the magnetostrictive $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles causes improvement of their crack resistance making employment possible of the tensile stresses, reduction of losses resulting from the induced eddy currents, costs reduction, and possibility of fabrication of materials of any shapes.

Intensive development of the composite materials [6-8], especially magnetostrictive ones, began in mid-nineties of XX century – then the researchers began investigations focused on extending the frequency band of the magnetostrictive converters operation. Thanks to the increased electrical resistivity compared to the monolithic material heat generated by eddy currents at operating frequencies above 1 kHz was reduced making it possible to extend the field of possible applications of the magnetostrictive converters.

Depending on the consolidation method of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder with the polymer matrix, the magnetostrictive composite materials are characteristic of various magnetic-, physical-, and mechanical properties. The fabrication methods include:

- bonding of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with the polymer material and their orientation in the magnetic field [5,9-14];
- laminated materials, including thin layers [14-15];
- materials in which $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ is infiltrated with polymer [16-17].

The acquired magnetostriction values are lower than for the monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$, and therefore, solutions are sought so far that would merge advantages of composite materials with good magnetomechanical properties.

The goal of this work is investigation of structure and properties of the magnetostrictive composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder particles.

2. Experimental

Examinations were made on samples of the composite material reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ (Etrema Co, USA) powder particles, demonstrating the giant magnetostriction. The Smooth-cast 325 (Smooth-on Inc., USA) two-component polyurethane resin with low viscosity was used as the matrix. Composite materials with cylindrical shape and dimensions of 8.7x40 mm were made by casting. For the particles sedimentation reasons, moving the mould continuously was required until the matrix gelation was completed. The samples differed with the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ concentration in matrix (Table 1) and with the powders particle sizes, denoted by their manufacturer as 38-106 µm and 212-300 µm were made.

Table 1.
Concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder 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	48.72
15	60.14
20	68.13

Analysis of the grain distribution of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder was made using the Mastersizer 2000 (Malvern Instruments) analyser employing the laser diffraction phenomenon. The measurement has been done in the equivalent diameter range up to 2000 µm. As a dispersing agent deionised water has been used, and the measurements have been carried out in constant temperature taking into consideration viscosity and density of liquid.

Observations of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder particles structure was made on Zeiss SUPRA 25 scanning electron microscope at the accelerating voltage of 20 kV and maximum magnification 500x.

Magnetizations versus applied field curves were registered using the Oxford Instruments Ltd. vibrating sample magnetometer (VSM) and on the basis of these relationships the hysteresis loop curves were drawn.

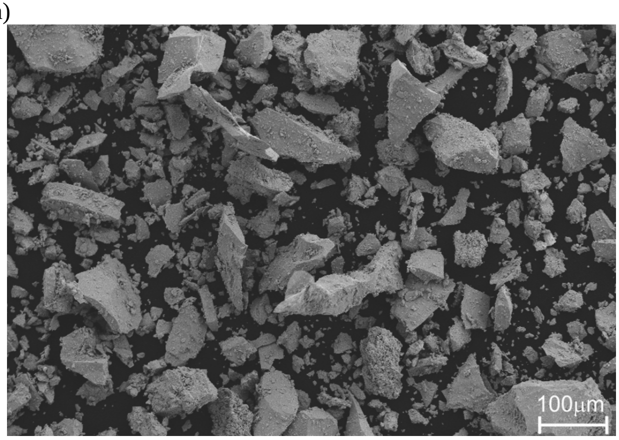
Testing of the magnetic permeability was made using the measurement system whose main element was the alternating current Maxwell-Wien bridge.

Measurements of magnetostriction formulated as the sample length increase in comparison with its initial length were carried out on a stand equipped with the capacitance dilatometer with three capacitors connected in series [18] in which sample deformations was observed as capacity change of the condenser with the movable electrode in the external magnetic field. The measurements were made in the transverse and longitudinal directions in relation to the sample axis, at room temperature and in the magnetic field with maximum magnetic field up to 800 kA/m.

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 powders' morphology observed on the scanning electron microscope is presented in Fig. 1. The powders differ both with the size and shape of grains, which is irregular in the entire range, moreover, in the population of fine particles (Fig. 1a) agglomerates occur made from them.

a)



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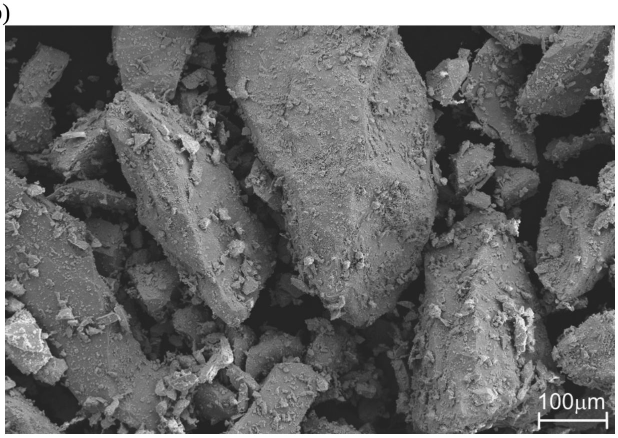

Fig. 1. Morphology of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with granulation denotes as: a) 38-106 μm, b) 212-300 μm; scanning electron microscope

Table 2.
Parameters characterizing grain-size distribution of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders

Designation of powder particles size / Parameter	38-106 μm	212-300 μm
Specific surface area, m^2/g	0.26	0.19
Quantile $q_{0.1}$, μm	10.27	16.09
Quantile $q_{0.9}$, μm	105.73	257.47
Median, μm	39.86	96.19
Mode, μm	60.25	158.5

Analysis results of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders grain-size distribution are shown in Fig. 2, whereas the statistical parameters characterising them are presented in Table 2. Powder with granulation denoted as 212-300 μm has grains whose median is 96.19 μm, whereas its quantile $q_{0.9}$ is equal to 257.47 μm (it means that 90% of grains have the dimension smaller or equal this value). The mode value (i.e., the most probable value in the powder grains population) for powder with this granulation is 158.5 μm, whereas for powder denoted as 38-106 μm: 60.25 μm. This powder is characteristic of grains with the clearly smallest dimensions, which is confirmed by median and quantile 0.9 μm values, equal to 39.86 μm and 105.73 μm respectively.

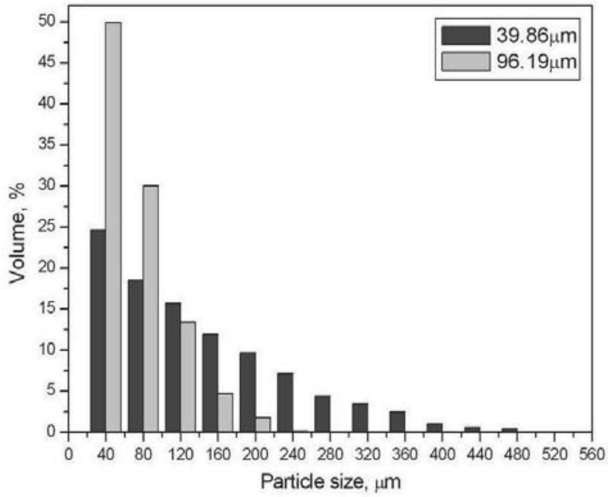

Fig. 2. Grain-size distribution of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders

Figures 3 and 4 shows magnetization versus applied field curves for composite materials reinforced with $Tb_{0,3}Dy_{0,7}Fe_{1,9}$ powders. From magnetic hysteresis loops obtained from these relationships characteristic magnetic properties was determined (Table 3).

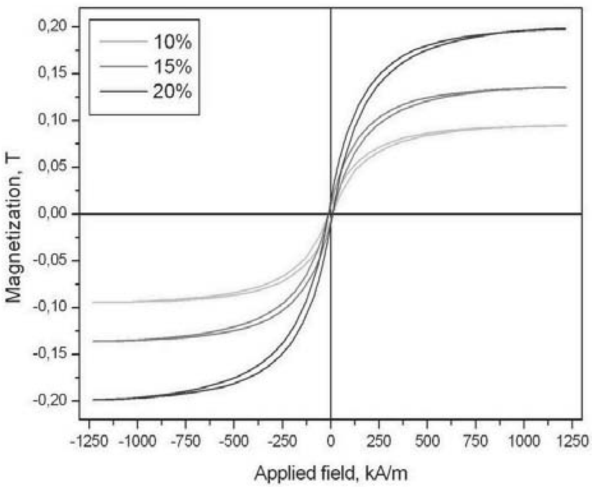

Fig. 3. Magnetic field dependence of magnetization for composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with median of 39.86 μm

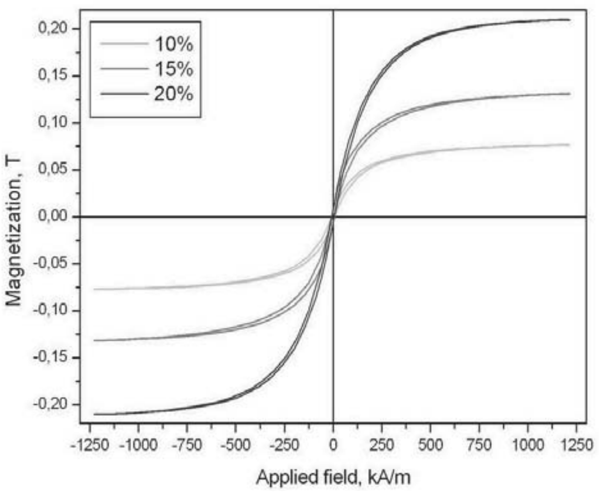

Fig. 4. Magnetic field dependence of magnetization for composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with median of 96.19 µm

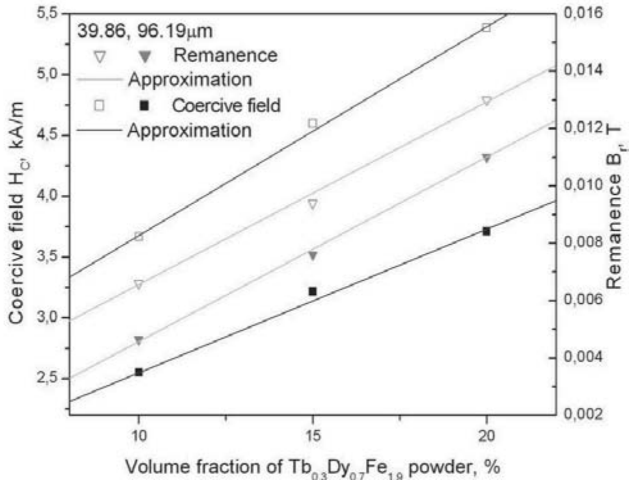

Fig. 5. Dependence of coercive field and remanence on $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder concentration and its granulation in composite materials

Table 3.
Magnetic properties of composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders

Median of the powder, µm	Volume fraction of powder, %	M_s, T	H_{max}, kA/m	H_c, kA/m	B_r, T	B_s, T
39.86	10%	0.094	1230.03	3.67	0.007	1.626
	15%	0.136	1228.48	4.60	0.009	1.664
	20%	0.199	1224.59	5.39	0.013	1.723
96.19	10%	0.077	1226.92	2.55	0.005	1.602
	15%	0.131	1226.92	3.22	0.008	1.659
	20%	0.209	1226.92	3.71	0.011	1.738

The saturation magnetization (M_s) of tested composites has a value of 0.094 T for materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with median of 39.86 µm and increase up to 0.199 T for materials with 20% volume fraction of reinforcement. What is more, this value has change continuously with the change of volume fraction of powder in matrix for both type of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles.

Figure 5 presents relationship of coercion and saturation magnetization with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder concentration in composite materials and its granulation. The coercive force (H_c) value is inversely proportional to the power particles size and assumes value of about 2.55 kA/m for composite materials reinforced with powder particles with median of 96.19 µm and about 5.39 kA/m for granulation with median of 39.86 µm. On the other hand the value of the attained remanence is proportional to concentration of the magnetic fraction in the polymer matrix and for composite materials with powder particles with median of 39.86 µm increase from 0.007 T to 0.013 T as the volume fraction of magnetostrictive powder increase. As expected a similar trend is observed for granulation with median of 96.19 µm, that is, an increase from 0.005 T values to close to 0.011 T for the highest volume concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder.

The course of magnetostriction curves for composite materials are shown in Fig. 6 and Fig. 7. Measurements were carried out of magnetostriction both the transverse- and longitudinal ones, for different orientations of the examined sample in respect to the external magnetic field, which confirmed the occurrence of the lower values of transverse magnetostriction in comparison to longitudinal ones (Tables 4 and 5). For transverse measurements the highest value of magnetostriction (i.e. $165 \cdot 10^{-6}$) was obtained for composite material with 20% volume concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder with median of 39.86 µm.

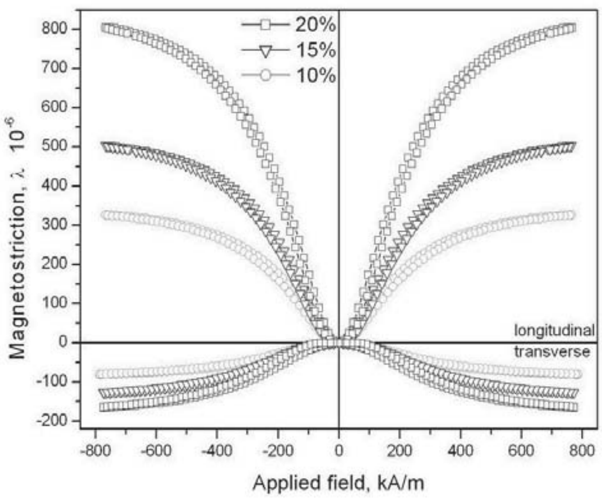

Fig. 6. Magnetostriction curves for the composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with median of 39.86 µm in directions perpendicular and parallel to sample axis

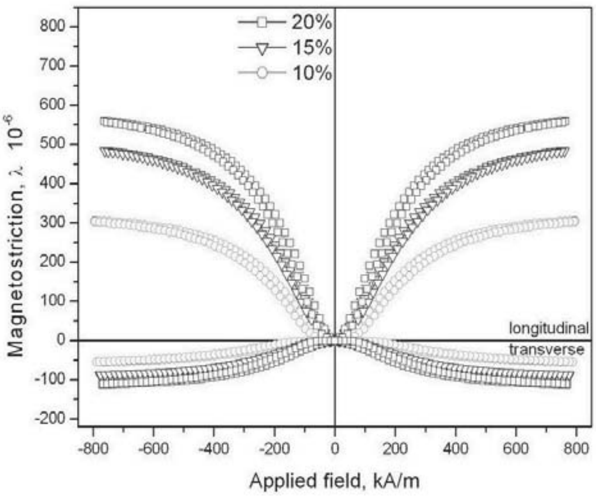

Fig. 7. Magnetostriction curves for the composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with median of 96.19 µm in directions perpendicular and parallel to sample axis

Table 4.
Magnetostriction values for the composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders from measurements taken in the longitudinal direction

Median of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder, µm	Volume fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$, %	H, kA/m	$\lambda \cdot 10^{-6}$
39.86	10%	766	326
	15%	764	503
	20%	766	805
96.19	10%	790	306
	15%	760	484
	20%	763	560

Table 5.
Magnetostriction values for the composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders from measurements taken in the transverse direction

Median of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder, µm	Volume fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$, %	H, kA/m	$\lambda \cdot 10^{-6}$
39.86	10%	786	80
	15%	772	127
	20%	772	165
96.19	10%	786	54
	15%	776	88
	20%	767	93

One can state, based on these curves, that all samples attain the saturation magnetostriction easily in the applied field intensity near to 800 kA/m, however, the composite material with 20% volume concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder with median of 39.86 µm attains the biggest magnetostriction value in the entire range of field intensity, acquiring the maximum magnetostriction equal to $\lambda_{max} = 805 \cdot 10^{-6}$.

As expected, concentration growth of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ in composite material (from 10 to 20% by volume) causes growth of the resultant magnetostriction. This results not only from the volume concentration growth of the magnetostriction phase, but also from the increase of the direct contact of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder particles with each other, so that better conditions occur for energy transfer in the composite material.

Properties of the matrix material are also of key importance here. The polyurethane resin used is characteristic of low viscosity which ensures the relevant moistening of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder particles, improving the magnetomechanical energy transfer.

In case of the composite material with 10% volume concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ a higher portion of polymer material causes that magnetostriction attains low values. With the excessively high matrix material content in respect to the magnetostrictive particles content, the composite material becomes the electrical insulator with the low magnetic permeability, in which dielectric properties predominate the magnetic ones.

It was found based on the permeability versus frequency measurements made (Fig. 8), that permeability of the composite materials is proportional to the magnetostrictive fraction concentration and attains highest values for materials reinforced with powder with median of 39.86 µm. The resin used as the matrix is diamagnetic material, so it has great influence on the values of obtained permabilities.

Moreover, in the investigated frequency range, i.e., from 20 Hz to 2 MHz, no effect was observed of frequency on the permeability value for the particular material, which – in connection with the high resistivity of the investigated composites – suggests possibility of using these materials in the high-frequency magnetic fields.

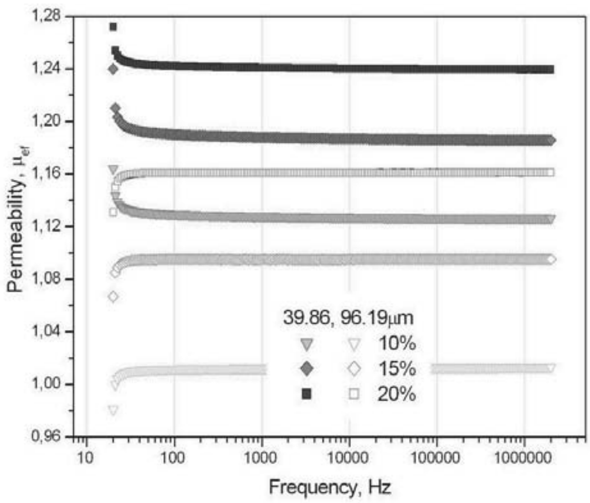

Fig. 8. Permeability - frequency relationship for the magnetostrictive composite materials

4. Conclusions

Results are presented in the article of the magnetic and magnetomechanical examinations of the composite materials with the polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder particles with median of 39.86 µm or 96.19 µm and varying volume concentration. It was found that saturation magnetization, remanence and saturation induction of these materials change proportionally to changes of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ volume concentration in the matrix. Moreover, it was found that materials reinforced with powder particles with smaller granulation demonstrate better magnetostrictive responses: differentiation of powder particles sizes in such range causes increase of their packing density, and therefore, growth of the resultant magnetostriction. However, there exists the boundary range of differentiation of their sizes, below which the packing density of particles will be smaller because of the effect of the electrostatic forces. Moreover, it is necessary to take into account the demagnetizing effect accompanying reduction of the powder particles sizes.

The biggest magnetostriction ($\lambda_{max} = 805\cdot10^{-6}$) was obtained for the material with 20% volume concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with median of 39.86 µm. The polymer matrix causes, moreover, growth of resistivity of composite materials, limiting these way losses for eddy currents at the high operating frequency of the transducers. The values of permeability of composite materials are nearly constant in investigated frequency range.

Lower cost, no need to use the initial stress and possibility to design actuator elements with the arbitrary shape decide the advantage of these materials as compared with the monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ and composite materials reinforced with magnetostrictive particles oriented in the magnetic field.

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